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HIGH-TEMPERATURE INTERMITTENT DRYING:

QUALITY CHARACTERIZATION OF CRUMB RUBBER

NG MEI XIANG, B.SC. (HONS)

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DEDICATION

Dedicated to my beloved mother, brother and Jun Yan

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ABSTRACT

In rubber research, drying is one of the most prominent stage to preserve the material. Numerous drying studies had been conducted by industrial specialists and researchers for decades, but the difficulties in producing consistent quality rubber remained unresolved. Hot air drying is the preferred artificial drying method to produce dried crumb rubber, but the final products tends to have wet rubber trapped in the dried rubber, which was undesirable. Prolonged drying at high hot air heating temperature was typically used to prevent the wet rubber pieces in dried rubber. However, the prolonged drying period leads to severe deterioration of rubber's properties. One way to overcome this quality problem is to apply two-stage drying techniques. The basis of this research work was to study the possibility of quality enhancement through two-stage intermittent vacuum convective drying of crumb rubber via time-varying stepwise temperature profile. The main objectives were to compare the quality and drying kinetics of crumb rubber subjected to the two-stage vacuum convective drying (VCD) techniques. The experimental strategy was started with drying the crumb rubber using vacuum drying (VD), hot air drying (HAD) and two-stage VCD. The application of two-stage VCD in rubber drying gave the shortest drying time compared to VD and HAD. It was found that the two-stage VCD gave the lowest colour change and high plasticity retention index. Thus, further experimental works were carried out based on two-stage VCD. To objectively access the quality changes of rubber, the effect of varying operating parameters, including vacuum pre-drying duration, hot air drying temperature and rubber sample diameter were investigated for two-stage continuous and intermittent VCD at high drying temperature (90 - 150°C). Their drying kinetics (drying time, drying rate, effective moisture diffusivity) and product quality (visual attributes, textural attributes) were considered. The results indicated that the drying kinetics and the overall quality results of dried rubber under two-stage intermittent VCD at high temperature performed better than the corresponding continuous tests. Using two-stage intermittent VCD, dried rubber had an acceptable colour, hardness and stickiness that required further test plan. Using a time-varying stepwise temperature profile, the rubber samples were dried sequentially with vacuum pre-drying at 90°C (30 min), followed by hot air drying at 150°C (60 min), 130°C (40 min) and 110°C (80 min). For every 15 min in the hot air convective dryer, the heat input was switched off for 5 min tempering intervals, which is intermittent ratio of 0.75. The time-varying stepwise intermittent process resulted in an acceptable colour of rubber products with moderate hardness and high stickiness. The total colour change (ΔE) values of the dried crumb rubber was 7.82, which was lowered than hot air drying and vacuum drying (12.75 and 9.93). The dried crumb rubber hardness and stickiness values were 69.42 and -27.55 g, respectively whereas continuous hot air dried crumb rubber had hardness and stickiness values of 620.69 and -140.29 g, respectively. The results indicate that the time-varying stepwise temperature profile helps in maintaining the sample temperature to prevent severe oxidative degradation to the samples. The samples tend to form impermeable rubber layer when undergone prolonged drying. The main contribution of this study is to provide comprehensive information on the high temperature drying characteristics of rubber, concerning both drying kinetics and the product quality. The effect of controllable factors such as vacuum pre-drying duration, hot air heating temperature and sample diameter were investigated for both two-stage continuous and intermittent VCD. The industry should adopt two-stage VCD approach with time varying stepwise profile for better rubber quality and shorter drying time.

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LIST OF PUBLICATIONS

Journals

1. **Ng, M.X**., Tham, T.C., Ong, S.P., Law, C.L. (2015). Drying Kinetics of Technical Specified Rubber, *Information Processing in Agriculture, 2*(1), 64–71.

2. **Ng, M.X**., Ong, S.P., Chin, N.L., Chuah, L.A., and Law, C.L. (2015). Review of Food Toxicological Issues Associated in Rubber Products. In *Processing of Foods, Vegetables and Fruits: Recent Advances*, eds. C.L. Hii, S.V. Jangam, S.P. Ong, P.L. Show, A.S. Mujumdar, 73-84.

3. Tham, T.C., Hii, C.L., **Ng, M.X.**, Ong, S.P., Law, C.L. (2018). Application of microwave-assisted drying on specific energy consumption, effective diffusion coefficient and topological changes of crumb natural rubber (Cis-1, 4- polyisoprene). Chemical Engineering & Processing: Process Intensification

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1. **Ng, M.X**, Tham, T.C., Ong, S.P., Lai, K.T., Law, C.L. (2014). Analysis of Vacuum Drying and Hot Air Drying on Temperature Profile and Heat Transfer of Rubber. *2nd International Conference on Agricultural and Food Engineering, CAFEi2014,* Kuala Lumpur, 01-03 Dec.

2. **Ng, M.X**, Ong, S.P., Chin, N.L., Chuah, L.A., and Law, C.L. (2014). Review of Food Toxicological Issues Associated in Rubber Products. Review of Food Toxicological Issues Associated in Rubber Products. *In proceedings of the 3rd International Symposium of Processing of Foods, Vegetable and Fruits.* Kuala Lumpur, 11-13 August, 122-128.

3. **Ng, M.X**, Tham, T.C., Ong, S.P., Law, C.L. (2015). Rubber Drying: Understanding the Mechanism and Recent Developments. *8th Asia-Pacific Drying Conference,* Kuala Lumpur, 10-12 August, 243-250.

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5. **Ng, M.X**, Ong, S.P., Law, C.L. (2017). Energy Saving by Hot Air Drying with Vacuum Pretreatment of Crumb Rubber. *9th Asia-Pacific Drying Conference,* Wuxi, China, 24-26 September.

LIST OF ABBREVIATIONS

A	Area (m^2)
a*	CIE - redness
b*	CIE - yellowness
DR	Drying rate (g H_2O min $^{-1}$)
DM	Dry matter
D_o	Diffusion coefficient (m^2 s^{-1})
D_{eff}	Effective moisture diffusivity (m^2 s^{-1})
ΔE	Total colour change
E_a	Activation energy (kJ mol^{-1})
H	Hardness (g)
HAD	Hot air drying
HPD	Heat pump drying
ID	Intermittent duration
IR	Intermittent ratio
L*	CIE - lightness
LPSSD	Low-pressure superheated steam drying
M	Moisture content (gH_2O gDM^{-1})
M_o	Initial Moisture content (gH_2O gDM^{-1})
M_f	Final Moisture content (gH_2O gDM^{-1})
M_e	Equilibrium Moisture content (gH_2O gDM^{-1})
M_{cr}	Critical Moisture content (gH_2O gDM^{-1})

MR	Moisture ratio
P_o	Initial plasticity
PRI	Plasticity retention index (PRI)
S	Stickiness (g)
SSD	Superheated steam drying
T_{rubber}	Absolute temperature
t_{on}	Total time with heat input (min)
t_{off}	Total time without heat input (min)
TSR	Technical specified rubber
VD	Vacuum drying
VCD	Vacuum convective drying

CHAPTER 1

INTRODUCTION

1.1 Background

Drying is a highly effective method to dehydrate and preserve a material. It is also the most critical process in producing good quality dried rubber. In rubber processing industry, ensuring consistent quality of the dried rubber is a major concern. Some of the common low cost processing techniques include open sun drying, greenhouse solar drying, ambient air drying and smoke drying often have limitation to the process control and consistency in quality. Commercial dried rubber sheets typically takes about four to seven days to dry using solar drying to reach complete dryness, but the rubber quality, in term of visual and textural attribute are not always in acceptable form (Tekasakul *et al.*, 2008; Siriwardena *et al.*, 2010; Suchonpanit *et al.*, 2011; Dejchanchaiwong *et al.*, 2016).

Natural rubber products are prepared from latex, which is the tree saps that are collected from a tree species called Hevea *brasiliensis*, widely known as the rubber tree. The latex is harvested by slicing a groove into the bark of the tree at a depth of a quarter inch with a hooked knife and peeling back the bark. Figure 1.1 shows the rubber tapping process:

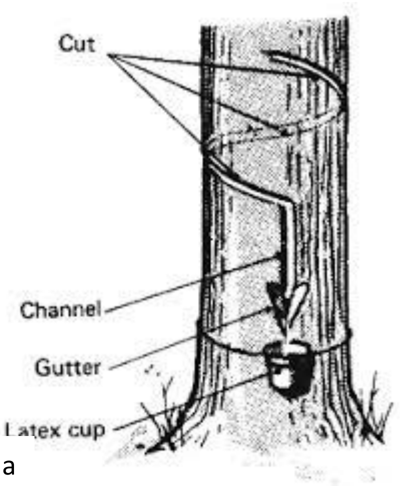


Figure 1.1 a: Tree saps flow out from rubber tree into a cup; Figure 1.1 b: Schematic diagram of rubber tapping process (FAO, 2019)

The latex is a sticky, milky colloid. It generally consisted of 5- 10% of non-isoprene components, such as proteins, carbohydrates, lipids, metal ions and other minerals, that will degrade and affect rubber quality during the drying process (Vaysse *et al.*, 2012; Roux *et al.*, 2000). Figure 1.2 below shows the fraction of natural rubber latex after centrifugation:

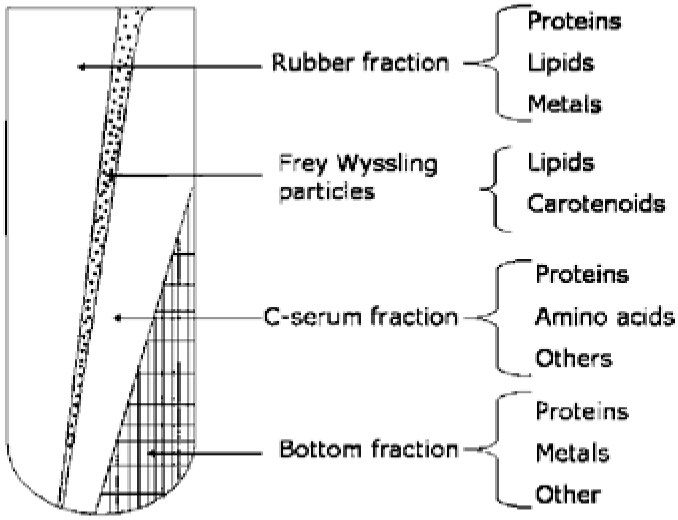


Figure 1.2: Fraction of natural rubber in general

During drying, the gradient in water content helps to migrate the non-rubbers to the outer surface. As the water evaporated, the latex particles will deform and pack together. Some non-rubbers or water soluble materials, such as protein migrate to the surface together with water, especially those incompatible with rubber. Then, the polymer surface chain inter-diffuse until a rubber sheets or rubber block formed. Prolonged drying will leads to a reduction in crosslink density. This occur earlier and much substantial at high drying temperature. As the delinking reaction prevails, the rubber softens and turns sticky. A softer rubber with high green strength is generally preferable for the automotive industry as it ease the mastication process.

1.2 Origin and historical aspect of rubber

Natural rubber is a plantation crop that was cultivated systematically since year 1877, when the British colonists introduced Hevea *brasiliensis* rubber to Singapore and Malaysia. The climate in Malaysia with fairly distributed rainfall is suitable for rubber tree cultivation. Hence, rubber production increased dramatically after the 1890s when there was a huge surge in demand for rubber. The favourable socio-economic factors also contributed to rapid expansion of rubber cultivation in Malaysia.

By the 1930s, Malaysia accounts almost 50% of the world's rubber production. Rubber products accounted for 3.9 percent of Malaysia's total exports for manufacturing products (Jeffrey, 2015). The main destination of natural rubber exports was China (55.2%), followed by Germany (11.1%) and Iran (9.6%) (Rubber Statistics Malaysia, 2018). The main downstream industry of natural rubber is the automotive industry which contributes over 70% of the rubber consumption in China. Global natural rubber production had increased from 4.4 million tons in 1983 to more than 13 million tons in 2016 to cope with the increasing need of rubber (Charles, 2016).

The Malaysian rubber products industry is made up of more than 500 manufacturers can be categorised into latex products, tyres and tyre-related products, as well as, industrial and general rubber products. The industry itself dominated by small and medium enterprises (SMEs). In 2016, a total of RM1.09 billion was invested into 13 approved projects, which four of the approved projects were wholly Malaysian-owned (RM12 million) (MIDA, 2017). Figure 1.3 below shows the stock, import, export and domestic consumption of rubber in Malaysia:

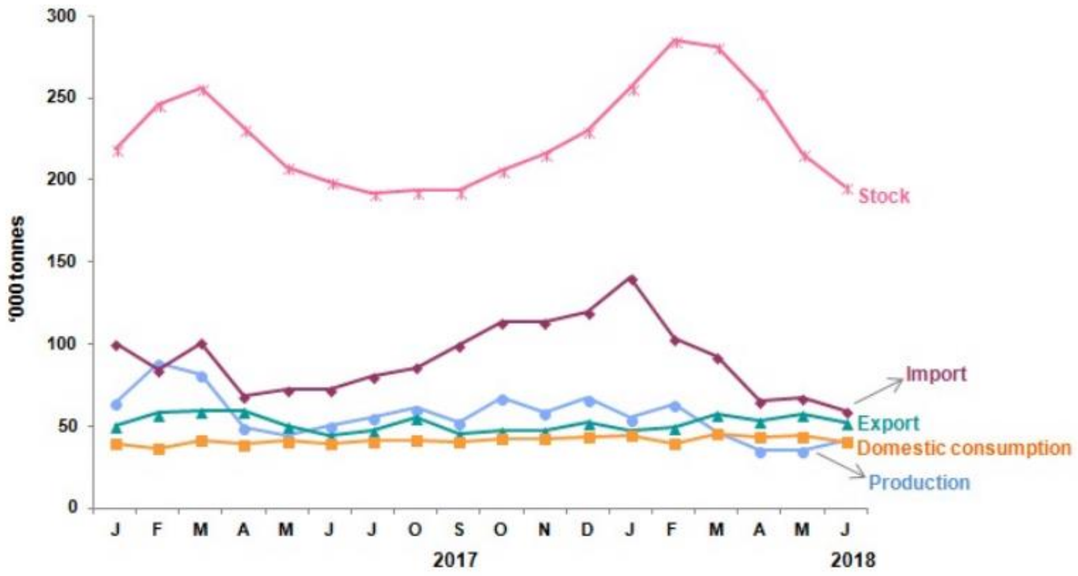


Figure 1.3: Monthly production of rubber 2017/2018

(Rubber Statistics Malaysia, 2018)

Natural rubber trees generally have a lifespan of 35 years and start yielding after 7 years of planting. Malaysia's natural rubber sector used to affect the national economy in term of production and consumption. However, the declining rubber prices and competition has forced many farmers to abandon rubber trees and converted to palm oil plantations. Production of rubber in Malaysia declined from over a million tons in the 1990s to 615,222 tons in 2000 and 41,578 tons in 2018 (Rubber Statistics Malaysia, 2018). Malaysia now accounts for only 4.29% of the world's rubber and is the world's sixth largest producer behind Thailand, Indonesia, China, Vietnam and India.

1.3 Basic principle of rubber processing

Prior to the introduction of technical specified rubber (TSR), rubber sheets are the only solid rubber product on the market, sold in the form of coagulated latex sheets. In order to turn latex into solid rubber, acid is added to the collected latex for the coagulation process. The coagulum, now a soft solid slab, is then squeezed through a series of rolls which drive out most of the moisture and reduce diameter to about 3 mm (1/8 in). The sheets are then draped over wooden frames and dried either in smokehouses or ventilated areas. Several days are normally required to complete the drying process for both methods. The darker rubber sheets in figure 1.4b are prepared with smoking process, while the lighter coloured rubber sheets in figure 1.4a are not smoked but heat dried in hot air chamber, under a shed or tunnel without smoke or additives. Figure 1.4a, b shows two types of dried rubber sheets:

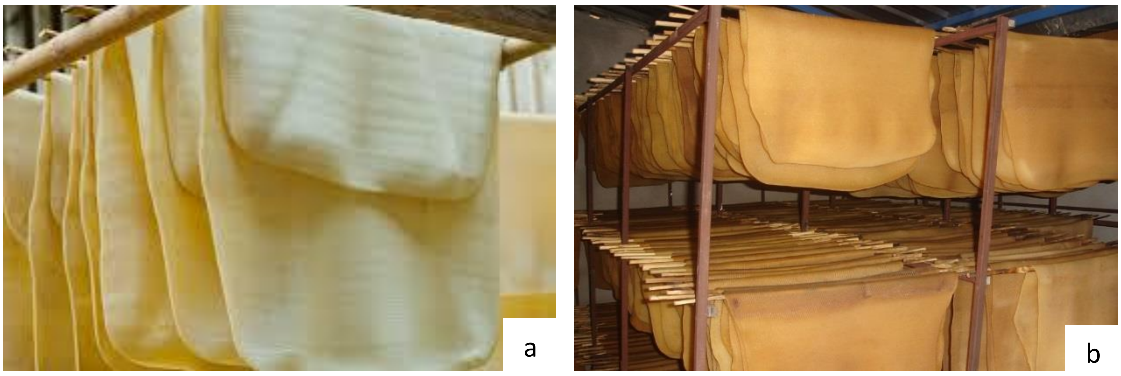


Figure 1.4 a: Air dried sheet;
Figure 1.4 b: Ribbed smoked sheet

1.3.1 Processing of crumb rubber to Technical Specified Rubber (TSR)

As the global demands increased, a faster processing method is in need for the rubber processing industry. The research works involving processing and drying of crumb rubber was carried out since early 1930s (John, 1935). From the residual of dripping latex after the latex collection (cup lump), to the drying of crumb rubber and the processing of technical specified rubber (TSR), the entire process might took more than 30 days (including maturation period). The cup lump formation is through natural coagulation of the tree sap from Heave *brasiliensis* tree. After the fresh latex from different Heave *brasiliensis* clones was naturally coagulated, the coagulum is subjected to different durations of maturation, processed into solid rubber by resizing and drying, then compounded into pure gum stocks and vulcanized.

Technical specified rubber (TSR) is produced from a mixture of cup-lump, crepe cup-lump and unsmoked rubber sheet. In the block rubber production process, the raw materials are cut and washed before being flattened into a crepe shape. The industry use slab cutter, rotary cutter and pelletizer to bring down the crumb rubber size to less than $<0.4 cm^3$. Figure 1.5 shows a very basic description of the crumb rubber processing prior drying in hot air convective dryer:

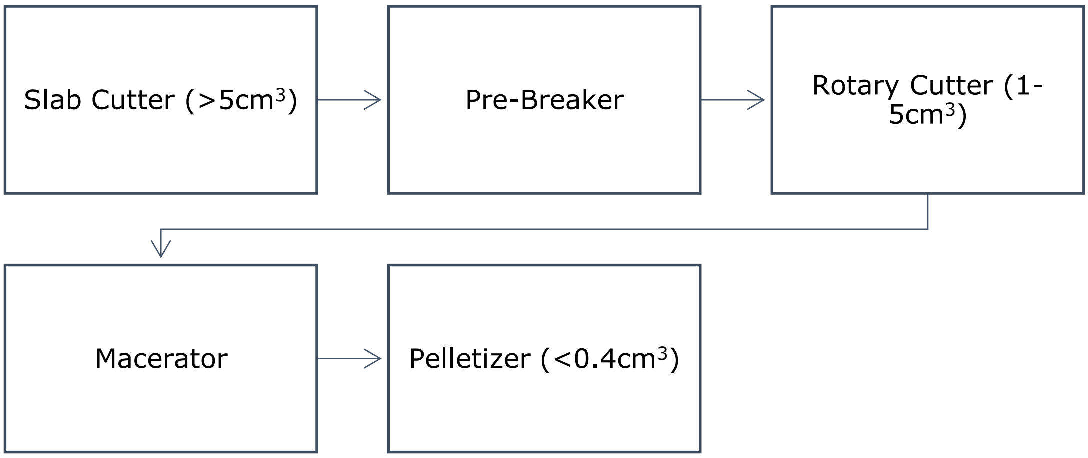


Figure 1.5: A process flow of rubber resizing process

During the rubber resizing process, the force exerted to the crumb rubber will mechanically dewatered the materials. This effectively reduce the thermal load and assist rubber to dry faster. The small crumb rubber pieces are transferred and inspected throughout the processing line using belt conveyors system. Figure 1.6 shows the conveyor belt that are used to transfer the crumb rubber from station to station:

Figure 1.6: Belt conveyor system to transfer rubber

The crumb rubber then goes through the drying process to remove excess water and is then compressed into a block shape for storage or shipment. There is no addition of any chemicals to the rubber during any of the processes involved: coagulation, washing, crumbing, drying, baling. Drying of crumb rubber is usually carried out by hot air convective dryer, namely tunnel dryer. Figure 1.7 shows the typical tunnel dryer for crumb rubber drying process:

Figure 1.7: Tunnel dryer for crumb rubber

The tunnel dryer is a machine where a container (tray) filled with the rubber is placed on a trolley or the material is loaded directly on the trolley and passed them through circulation convection drying at atmospheric pressure (Karunaratne, 1971). The heat is conveyed by hot air and the moisture is removed simultaneously from the rubber by the dry air. The final product, block rubber or TSR, as in figure 1.8 is the raw material used in the production of tyres for automobiles and aeroplanes.

Figure 1.8: Technical specified rubber (TSR)

TSR20 grade is the most widely sold solid rubber for technical specified rubber (TSR). It has the highest market demand from the automobile industry and most commonly exported, for its good technological properties, such as plasticity retention index (PRI). PRI is defined as the measure of the susceptibility of raw rubber to thermal oxidative degradation, which directly related to the process ability of rubber materials.

1.3.2 Technical Specified Rubber (TSR) grading system

All the block rubber is graded according to precise technical parameters such as dirt content, ash content, nitrogen content, volatile matter and properties of the rubber, based on the International Standards Organization (ISO 2000:2003) as shown in Table 1.1.

Table 1.1 Specification of Technical Specified Rubber (TSR)

Parameters	TSR-CV	TSR-L	TSR-5	TSR-10	TSR-20
Dirt content, %wt, Max	0.05	0.05	0.05	0.10	0.20
Ash content, %wt, Max	0.60	0.60	0.50	0.75	1.00
Nitrogen content, %wt, Max	0.60	0.60	0.50	0.60	0.60
Volatile matter % wt, Max	0.80	0.80	0.80	0.80	0.80
Initial Wallace plasticity Po, Min	-	30	30	30	30
Plasticity Retention Index, PRI, Min	60	60	60	50	40
Colour, Max (Lovibond units)	-	6	-	-	-

*Remark: Those are not specified are controlled by rubber processor.

Basically, there are five main grades of crumb rubber, namely consistent viscosity rubber (TSR-CV), light-coloured rubber (TSR-L), rubber grade number 5 (TSR-5), rubber grade number 10 (TSR-10), and rubber grade number 20 (TSR-20). The differentiation of the five grades is based on the dirt and ash content %. Plasticity Retention Index (PRI) and colour limit, but not all values were affected significantly by drying conditions. The actual quality changes during the drying process was mainly focused on visual and textural attributes.

1.4 Research Aims

The research aims to:

- Investigate the effect of vacuum drying (VD), hot air drying (HAD) and two-stage vacuum convective drying (VCD) strategies on the drying kinetic of crumb rubber to improve drying rate and reduce total drying time
- Determine a suitable intermittency strategy for the two-stage vacuum convective drying techniques to improve quality of crumb rubber
- Analyse the effect of vacuum duration, heating temperature and product size on the quality changes of crumb rubber under two-stage intermittent vacuum convective drying
- Optimize the two-stage intermittent vacuum convective drying (drying kinetics, colour, hardness and stickiness) via time-varying drying scheme

1.5 Research scope

1.5.1 Drying kinetics

Drying of natural rubber typically carried out at high drying temperature in the range of 105°C - 140°C with hot air convective dryer (Tham *et al.*, 2014; Tirawanichakul *et al.*, 2009; Ngolemasango *et al.*, 2003). However, the high drying temperature would result in lower plasticity, material softening and loss of strength in rubber properties (Tham *et al.*, 2014). This study aimed to improve the quality attribution of crumb rubber in comparison with the effect of intermittent drying and continuous drying to the property changes of crumb rubber (colour, plasticity, hardness and stickiness). The effect of intermittent drying on the drying kinetic and properties changes of rubber in combination of various intermittency strategy, based on intermittent ratio and tempering time were studied.

By utilizing two-stage drying technique at high drying temperature and short drying time for the intermittent drying, the surface moisture and internal moisture can be removed quickly to a higher precision of moisture control. The two-stage drying technique could reduce operating cost and minimize produce degradation through a more versatile drying process. Tham *et al.* (2014) also suggested the use of various drying technique to increase the energy efficiency of the drying process and improve the product quality. Similarly, Kumar *et al.* (2001) and Chou and Chua (2001) agreed that multi-stage drying techniques maximize the product quality and improve the energy efficiency, as discussed in a review of fruits and vegetables drying (Sagar and Kumar, 2010). There were many relevant studies on using two-stage drying system to improve product quality (Wojdyło *et al.*, 2014; Chong *et al.*, 2014; Tian *et al.*, 2016; Chou *et al.*, 2017). Wojdyło *et al.* (2014) discussed on preserving the biocompounds and colour of dried sour cherries through two-stage drying with varying operating parameters. Likewise, a combined drying of apple cubes showed that a suitable combination of two-stage drying would help in retaining colour quality (Chong *et al.*, 2014). The study on infrared intermittency also managed to reduce drying time while minimizing colour degradation (Chou *et al.*, 2017). The choice of dryers is critical to ensure the drying environment match with the product drying characteristic. In current research, the drying strategy would be optimized by details analysis of total processing time, heating temperature and critical moisture content to determine the fundamental rubber drying characteristic.

There are several factors that could influence the drying mechanism of crumb rubber, which mainly categorized into two aspects, process conditions and internal mass transfer mechanism in rubber. From these two factors and consideration of industrial expectation on product quality and time saving, the main focus of this research is to develop a suitable drying technique and improve existing drying scheme. Typically, crumb rubber is dried in a drying tunnel with hot air drying (HAD) for prolonged period at drying temperature (105-140°C), which resulting in unnecessary quality degradation. In this study, the experimental works included two-stage vacuum convective drying (VCD) method, intermittent drying (varying intermittent ratio and tempering time) and time-varying drying scheme. The proposed two-stage intermittent VCD with time-varying drying scheme is evaluated based on the drying rate and moisture content changes of crumb rubber. As the variation of moisture content of rubber depends on process conditions, the drying kinetic of rubber was investigated in this study. The drying parameter that gave highest moisture diffusivities is chosen as the most suitable setting.

The two-stage vacuum convective drying (VCD) proposed in this study aims to improve the drying uniformity of crumb rubber to solve the “white-spots” problems and prevent oxidative degradation due to high drying temperature. The selection of suitable combination of two-stage vacuum convective drying (VCD) and determination of suitable intermittency strategy for hot air drying (HAD) part is based on the least property changes in crumb rubber. The evaluation of product properties in both continuous and intermittent VCD were done before carrying out the time-varying drying scheme. The comparison of drying kinetic and product properties in continuous drying and intermittent drying make the selection of time in the stepwise drying scheme easier to understand. The optimized drying scheme can be applied in all existing rubber processing plant in Malaysia with some modification to existing dryer, but the design, fabrication, simulation and modelling are not included in this study.

Drawing on this idea, the drying experiment was designed to minimize the total drying time via two-stage intermittent vacuum convective drying (VCD) process with stepwise drying scheme. All the drying experiments were done using laboratory dryer in University of Nottingham Malaysia, but all test results were compared with reference samples obtained from the same factory that provide the crumb rubber samples. The drying procedure used for the experiment are identical to the process used by the factory. The crumb rubber sample dried with laboratory dryer can obtain properties like the rubber processed in the factory.

1.5.2 Final product quality

All the raw materials were sourced locally from the same manufacturer to ensure consistency in the comparison study. This study does not include analysis of structural characterization of rubber, clonal origin of rubber and the type of coagulation used in the factory. The crumb rubber samples obtained were frozen upon procurement to retain its properties and prevent it from any mean of degradation. Prior to the drying process, samples were defrosted at room temperature to return to its original state. Reference samples are used to assess the quality of raw materials prior drying. This study limited only in improving the physical (colour quality) and mechanical properties (hardness and stickiness) of the crumb rubber. Some calculations, assumptions and selections were made as a consideration of a proper and realistic design. A theoretical calculation of energy used during the drying process is done as a reference to the improvement of the drying scheme. The anticipate outcomes from this study would be a good base for further investigation of the topic.

1.6 Contribution of study

In this research, the drying kinetics of crumb rubber such as the drying rate, temperature - moisture content distributions, as well as theoretical approaches to moisture movement were studied. The fundamental understanding in the drying kinetics of crumb rubber is essential for the prevention of quality degradation and for the achievement of fast and effective drying. This present study would be able to close the gap in the literature regarding the drying kinetic of crumb rubber. Through experiment determination, the initial constant rate period might not happen in the rubber drying process as the available free moisture on the rubber surface is removed quickly, whereas the bound moisture in the rubber rely on moisture transport to the surface during falling rate period.

A new combination process of two-stage intermittent vacuum convective drying (VCD) with a time-varying stepwise temperature profile was developed. Effective moisture diffusivity (D_{eff}) is one of the most important transport properties to find out the heat transfer in rubber drying process. However, there are limited publications on the public domains that could be used to compare the results in current rubber drying study, thus the results found in current research would be a knowledge input for the domain of rubber drying studies.

A high drying temperature for hot-air drying (HAD) stage was coupled with multiple short tempering periods applied in succession to prevent quality degradation of crumb rubber. The stepwise drying scheme and the tempering period during HAD could reduce the effective drying time. Similarly, the usage of two-stage VCD ease the temperature schedule control and improve the drying efficiency. The two-stage vacuum convective drying (VCD) give rubber processors a better understanding of the available technologies to enhance their existing drying process.

The research provided novel information on the drying kinetic and quality control during two-stage VCD process and the possible consequences on NR properties, especially on hardness and stickiness. Rubber softening is a phenomenon specific to natural rubber (NR), during which links deform between the poly-isoprene chains and non-rubber compounds. This work provided new knowledge on how to reduce the existing quality degradation problems during rubber drying process by detect any changes in the physical structure as well as solute transport that can occur in the rubber drying process.

Many rubber-based products depend on natural rubber for quality and performance. The addition of vacuum pre-drying to the traditional tunnel drying method would benefit the industry in terms of better product quality and production efficiency. The crumb rubber processing industry has huge potential to improve the rubber production time using two-stage vacuum convective drying approach with time varying drying scheme. In short, technological innovation is pivotal for the future sustainability of the rubber industry.

CHAPTER 2

LITERATURE REVIEW

2.1 Research on crumb rubber drying

Forced convective hot air drying has been the most popular method for rubber drying due to its simplicity and low operational cost. The existing drying method is sufficiently versatile to process all forms of rubber, but the energy cost of thermal drying is high (Kudra and Mujumdar, 2004; Chua *et al.*, 2001). A few studies regarding drying efficiency and total energy consumption of crumb rubber drying process had been carried out (Khongchana *et al.*, 2007; Tasara *et al.*, 2009; Utomo *et al.*, 2010). A rubber processing plants typically consumed 1.17MJ kg^{-1} of diesel fuel, while the drying process itself used about 7.67% of the total production energy (Utomo *et al.* 2010). Therefore, a change to the drying process is of interest to rubber processing industry.

The basic principles in manufacturing process of crumb rubber has remained almost unchanged since heveacrumb process invention in 1964 (Sekhar, 1967; Karunaratne, 1971). The manufacturing process of rubber is constantly under development, with the basic principles and raw materials remained the same as half a century ago. The basic manufacturing process has not changed from the beginning of 1969 when block rubber manufacturing first produced in Malaysia: the rubber is blended, pressed and formed to sheets, then loaded into trays and dried. There is a need to understand the fundamental drying kinetic of crumb rubber to design a better drying scheme.

2.1.1 Solar drying system

Investigation of continuous drying on the crumb rubber quality was done by Tirawanichakul (2008) and Khongchana (2007), since decade ago. Most of the recent drying study is focused on improving the solar drying system for rubber sheet, in example, solar dryer (Dejchanchaiwong, 2016), solar greenhouse dryer (Arekornchee, 2014), and Sandwich Greenhouse (Tanwanichkul, 2013). The quality of the rubber dried using the methods abovementioned are normally visually graded based on purity and colour. There is only a few recent publication or studies relating crumb rubber drying or the effect of drying conditions to the drying characteristic of crumb rubber and rubber quality (Ng, 2015; Tham, 2014). The publication on alternative drying technology to dry the rubber materials and the effect of drying method on the other dried rubber's properties of are limited.

Felix (2015) studied the effect of various drying methods to rubber sheet quality and concluded that the best continuous air drying temperature of rubber sheets with initial moisture content of 42% - 50% were 27 - 33°C. However, crumb rubber dried by solar drying and air drying is susceptible to fungal proliferation. The crumb rubber properties may deteriorate, including appearance, colour and textural properties during the long hours of drying time. The drying time depending on the dryer design and total dryer capacity to produce block rubber. The residence duration of rubber inside the dryer might be prolonged due to the change in machinery design. The major disadvantage of using hot air convective drying system associate with the degradation of material quality (Rahman, 1985; Tirawanichakul *et al.*, 2009). Thus, from a practical perspective, the changing of heating temperature would be one of the quickest method for the industry to adapt and to improve their product quality.

Tirawanichakul and Tirawanichakul (2008) did a simulation study, based on empirical equation and diffusion model. Drying kinetic was predicted with theoretical thin layer drying equation, which showed that effective diffusion coefficient is directly proportional to drying temperature. A comparison of a series of drying temperatures ranged from 40 to 150°C showed that the highest theoretical effective diffusion coefficient is 2.15 x 10^{-5} m^2 s^{-1}. In Tirawanichakul's studies, there were no drastic changes on colour for drying temperature of 110 - 130°C, which was preferable results. Thus, the experimental design in current study would be comparing drying temperature of 90, 110, 130 and 150°C.

2.1.2 Machine design improvement

Existing drying technique by tunnel dryer, a hot air convective drying technique, is carried out by moving the rubber from one stage to another with time interval between five to fifteen minutes. Drying in this technique took relatively short time (maximum 240 minutes) for industrial scale drying, comparing with the drying studies carried out in laboratory scale (minimum 300 minutes) at same drying temperature. The differences in machinery design for industrial scale and lab scale changed the proportion of rubber surface in contact with circulated air, hence affected the total drying time taken. Thus, several constrains had been taken into consideration to handle the scaling down process to prevent any artefact to the experimental results.

Alternative drying method other than hot air convective drying or solar drying system was heavily studied starting 1978 (Rahman, 1985). Many research on crumb rubber drying were mostly confined to mechanical and design improvement on the drying equipment (Park *et al.*, 1928; Berthomieu *et al.*, 1997). The process began with allowing wet rubber coagulum passed through a series of rollers and resizing by granulators or pelletisers (Graham, 1968; Morris and Sekhar, 1969; Karunaratne, 1971). The study on the effect of varying machinery pressure to remove the moisture of rubber mechanically do not bring much changes to rubber drying time (Kemp, 1938). At this stage, the moisture of rubber was reduced from the initial moisture content of 35 – 55% dry-basis to 20 - 40% dry-basis as the wet rubber coagulum was reduced to a smaller size (Graham, 1968). Similarly, current mechanical dewatering by fine pelletizer is capable to bring down the initial moisture content of crumb rubber to 35% prior drying. As the rubber is cut into small-size crumbs, the surface area become larger and improve the heat transfer, hence the drying process would be relatively effective compared to rubber sheets (Rahman, 1985). Thus, rubber size is an important criterion to improve the drying efficiency.

Tirawanichakul *et al.* (2009) simulated the effect of drying condition on TSR 20 based on empirical equation and diffusion model, which the simulation prediction of theoretical thin layer drying equation best fit the experimental data. However, the use of thin layer drying equation for a high rubber bed depth of 100mm might be inappropriate as there was significant temperature gradient throughout the drying experiment and unable to achieve uniform temperature distribution. In Tirawanichakul's studies, the first hour of drying showed a temperature difference between the top layer and bottom layer was higher than 20°C, whereas the air temperature measured difference between rubber layers of 40 - 80mm from base was minimal. This phenomenon indicated that sample diameter lower than 50mm might be a better fit to the simulated results with theoretical thin layer drying equation. Thus, experiments carried out in current study would be focusing on sample diameter varies between 10 – 50mm. The simulated and descriptive showed the importance of rubber sizing to the drying process.

2.1.3 Two-stage vacuum convective drying (VCD)

The usage of different dryer type at each drying stage to perform heat integration, which commonly known as multiple-stage drying is gaining popularity in recent years. Multiple-stage drying has been tested on agricultural and food products to elevate the drying efficiency (Chou *et al.*, 2001). For rice drying, the two-stage drying systems normally combine a fast-drying technology with low temperature dryer to reduce the cracking defects in paddy (Dokurugu, 2009; Srzednicki and Driscoll, 2008). Similarly, two-stage drying was commonly applied on rubber drying for a better control of the drying process. The higher heat transfer rate and better process flexibility for two-stage drying would be able to improve the drying kinetic of crumb rubber and maximize product quality (Mujumdar, 2004).

Typically, crumb rubber drying process is carried out over two-stage, with the first stage focusing on removal of the available free moisture from the surface; and, second stage focusing on improvement of the internal mass transfer (Rahman, 1985). The heating process improve the rubber uniqueness, especially on physical characteristic of rubber, such as elasticity, flexibility, and strength. However, the heat also caused oxidative thermal degradation and depolymerised of the molecular chain. This chain scissioning process create a high amount of low molecular weight rubber particles, which will lead to a loss in strength (Ngolemasango *et al.*, 2003).

The suitability of a drying condition, normally based on preferable heat transfer mechanism, state and shape of the products (Barbosa and Vega-Mercado, 1996). Drying of crumb rubber can be done in several ways, including convective drying chamber (Ng *et al.*, 2015; Tirawanichakul *et al.*, 2009; Tirawanichakul *et al.*, 2008; Khongchana *et al.*, 2007; Berthomieu *et al.*, 1997), vacuum drying chamber (Ng *et al.*, 2015; TropenpRanzer, 1905), microwave drying chamber (Lin *et al.*, 2008) or fluidized bed drying chamber (Pollock, 1996), based on a review carried out by Tham *et al.* (2014). For rubber, there is no capillary movement as it is a non-porous material that would not absorb moisture easily, hence all liquid movement would be dominated by diffusion process. Diffusion is the main transport mechanism during the entire rubber drying process, including the initial heating time and falling rate period.

Rubber properties tend to vary with drying methods. Rubber drying is essential for the rubber processing plant, where efficient moisture removal plays an important role. The heat transfer during drying, either convective or conduction, would bring changes on the polymer's chemical characteristic, mainly due to the heat transfer effect (Park *et al.*, 1928; Berthomieu *et al.*, 1997; Tasara *et al.*, 2009; Khongchana *et al.*, 2007). This study began with several controlled experiments to identify the effect of drying techniques, included vacuum drying (VD), hot air drying (HAD) and two-stage vacuum convective drying (VCD) on the drying kinetic and other property changes of rubber. The advantage of combining vacuum and hot air to perform a drying operation included a higher mass transfer rate (Péré and Rodier, 2002), a more homogeneous heating and a shorter drying time.

On top of that, Khongchana *et al.* (2007) also tested the effect of three different drying conditions on thick rubber (250mm), comparing single stage drying, two - stage drying and airflow direction. The researcher concluded that two - stage drying with high drying temperature could enhance the evaporation of moisture from rubber, based on a combination of drying temperature at 130°C for 40 min and 110°C for 120 min.

Two-stage VCD has the advantage of both drying system, which provides the synergistic effect to improve the effectiveness of drying. Rubber samples that were pre-dried using vacuum drying were further dried in hot air convective dryer. Pre-drying is a potential way to increase the overall drying efficiency and product quality as majority of surface moisture can be removed during the process. The high heat transfer to crumb rubber under sub-atmospheric pressure, also enhance the moisture migration to the surface and prevent oxidative thermal degradation of rubber. Determination of suitable vacuum drying (VD) duration and hot air drying (HAD) heating temperature to obtain high drying rate of rubber without affecting the product quality was included in this study. Thus, the drying of crumb rubber in vacuum condition is compared with the drying of crumb rubber in hot air drying condition at the initial state.

Studies of rubber drying, particularly by two stages drying, are limited and span mostly on modelling work. Many studies conducted on crumb rubber drying have shown that drying temperature is proportional to the rate of moisture removal (Aguele *et al.*, 2015; Tasara *et al.*, 2009; Tirawanichakul *et al.*, 2009; Varghese *et al.*, 2004; Berthomieu *et al.*, 1997; Naon *et al.*, 1995; Cousin *et al.*, 1993). However, this is often accompanied by degradation in the rubber properties, in term of colour (Suchonpanit *et al.*, 2011; Jitjack *et al.*, 2016) and physical properties (Felix, 2015; Tasara *et al.*, 2009; Khongchana *et al.*, 2007). The colour change from white granules to a uniform gel is preferable compared to a darker or sticky surface (Suchonpanit *et al.*, 2011). The rubber with highest grade is typically gold or light yellow in colour with good colour consistency (Jitjack *et al.*, 2016). A consistency in dried rubber quality is utmost important for tyre manufacturing plants as this semi-processed product accounts for about 40 percent of the total raw material cost to produce a good quality tyre (Ferrer, 1997). A combination of soft, strong and tacky rubber could shorten the mastication process and improved the processability. An evaluation of the sample colour and texture will be able to determine the effect of drying on the physical changes. The current scientific knowledge regarding the changes of rubber properties during the drying process is very limited and not determined in detail. Thus, this present study would be able to close the gap in the literature regarding the effect of crumb rubber drying to TSR 20 quality.

2.1.3.1 Hot Air Drying (HAD)

Hot air convective dryer is easy to operate and low maintenance, which is the simplest and the most economical choice in comparison with various advance drying technology (Fernandes *et al.*, 2011). This drying process basically transfer heat energy by forced convection of hot dry air. Figure 2.1 is a schematic diagram of the hot air convective dryer showing how the hot air is blown by the fan to the oven chamber.

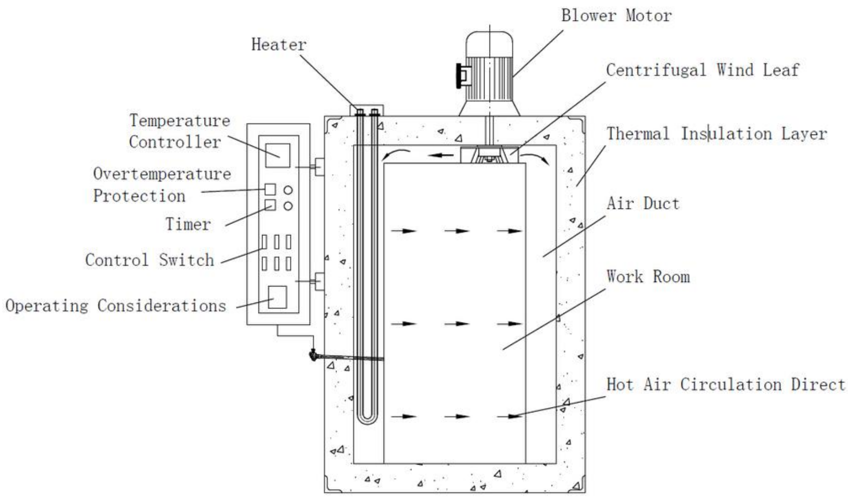


Figure 2.1: A schematic diagram of hot air flow in hot air convective dryer (Guangyi, 2019)

Natural rubber has a low thermal conductivity. When a forced convection of hot dry air blow onto the rubber, the surface moisture dried out too quickly and form a sticky, oily layer on rubber surface. This eventually leads to an inhibition of moisture migration to surface. The slow internal mass transfer (by diffusion) and low external heat transfer (by conduction) are typically mitigated by strong airflow during rubber drying. As the predominant transport mechanism of hot air convective drying is convection, the simultaneous exchange of heat and mass transfer between rubber and the heated air are intense. The slow heat transfer process and long rubber drying time are mostly due to both external and internal resistance to heat or mass transfer (Yau *et al.*, 2012). Thus, a pre-drying without strong forced convection of hot dry air would be beneficial for rubber drying process.

Naon *et al.* (1995) had analysed the heat and mass transfer phenomena during rubber drying process using portable laboratory dryer. Some of the experimental design in this research are extended from Naon's studies, especially in determining the effect of heating temperature to rubber drying process. From several earlier published works, Khongchana *et al.* (2007), Tirawanichakul *et al.* (2009) and Suchonpanit *et al.* (2011) had been trying to simulate and design industrial dryer for rubber processing industry, which focused primarily on technical specified rubber (TSR-20) and air dried rubber (ADR) with the use of self-fabricated hot air convective dryer. Hot air convective drying is a common drying techniques in rubber drying research. Table 2.1 has summarized some of the selected rubber drying studies, including the drying method, drying time and rubber quality. In the rubber drying studies, most of the drying process were carried out by either single - stage or two - stage drying techniques.

Table 2.1 below is a summary of selected work on crumb rubber drying with hot air convective dryer.

Table 2.1: Selected works on hot air drying (HAD) and product quality

No.	Rubber Types	Drying Methods	Drying Temp. (°C)	Drying Time (Min)	PO %	PRI value
1	TSR-20 (Khongchana *et al.*, 2007)	Two – stage HAD (cyclic drying)	130/110	40/180	34.5	77
2	TSR-20 (Tirawanichakul *et al.*, 2009)	Single stage HAD	100	180	42.5	52
3	TSR-20 (Tirawanichakul *et al.*, 2008)	Two - stage HAD (cyclic drying)	130/110	40/180	41.5	63
4	TSR-20 (Jutarut *et al.*, 2009)	Two - stage HAD (cyclic drying)	140/110	20/130	38.5	65
5	ADR (Suchonpanit *et al.*, 2011)	Single stage HAD	64.1	2340	44.5	100

In example, Tirawanichakul *et al.* (2009) used a short time high temperature for first stage drying and followed by high air flow low temperature for second stage drying. This stepwise drying technique, typically effectively reduced the oxidation of rubber due to prolonged drying period at high heating temperature, however, unable to resolve the problems of wet particles incorporation in the dried rubber. The second stage of the drying at low temperature with high air flow might lead to the formation of rubber skin. A high temperature drying process lead to immense heat stress to the physical properties of the products, which resulting in significant quality losses and affect the process-ability of the dried rubber.

In another study, the results showed that drying of rubber in thick layer could enhance the moisture diffusivity of rubber and reduce the total drying time (Cousin *et al.*, 1993), which was in agreement with Jutarut *et al.* (2009), who found the drying efficiency of a rubber drying process was correlated with rubber bed depth. However, it is necessarily to determine the influence of sample diameter to the rubber drying characteristic. By analysing the drying time, drying rate, and effective moisture diffusivity, the rubber drying mechanism can be better understood.

2.1.3.2 Vacuum Drying (VD)

Vacuum drying (VD) is an efficient method functioned at low atmospheric pressure to improve effective moisture content at low drying temperature (Bhattacharya, 2014). At normal atmospheric pressure, 1000 mbar, water boils at 100°C; whereas, at sub-atmospheric pressures, water boiling temperature is lowered (Reis, 2014). Heat sensitive materials typically employed vacuum drying at reduced pressure of 25 –

100 mbar as a pre-drying stage (Keey, 1991). Figure 2.2 below shows a schematic diagram of the vacuum dryer:

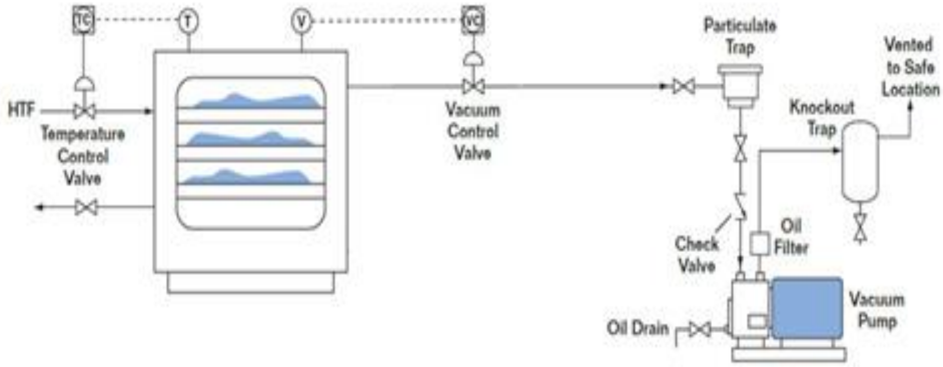


Figure 2.2: A schematic diagram of rubber drying in vacuum dryer (Dicimo, 2018)

Due to the low vapour pressure and oxygen-free condition, the diffusivity coefficient of moisture transfer increased without causing severe degradation to the drying materials. Drying studies from Hee and Chong (2015), Lee and Kim (2009) and Reis (2014) showed an improvement in diffusivity when the heating temperature in vacuum drying increased. However, vacuum drying at high temperature might resulting in poorer colour quality (Alibas, 2007).

The earliest scientific articles on the effect of vacuum drying to quality change of fruits and vegetables were published on 1930s (Webb, 1938). Krokida (1998) compared the effect of conventional hot air drying (HAD) and vacuum drying (VD) on fruits and found that the colour quality deteriorated when drying was carried out at high temperature and low humidity for both drying methods. The comparative studies showed that heating temperature does not affect the darkening of the materials, but change the colour parameter, redness (a*), yellowness (b*) and increase the total colour change for both HAD and VD up to the drying temperature of 90°C. Vacuum drying of Indian gooseberry flake also showed a higher total colour change in comparison with low-pressure superheated steam drying (LPSSD) at 75°C (Methakhup, 2005).

A summary of drying studies on various materials using vacuum dryer is as shown in Table 2.2. Table 2.2 shows that majority of experimental works were carried out at temperature of 75˚C, except some that were tested at higher temperature of 90˚C. When a drying material was dried with a high drying temperature and low absolute pressure in vacuum drying, a higher rubber drying rate reduced the drying time and save energy cost (Patil, 2016). However, the intense heat provided by the vacuum dryer often lead to formation of a non-porous surface skinning (Rahman, 1985). The non-porous layer inhibited the moisture diffusion to the surface and prolonged the drying time. Hence, vacuum drying is preferable to be used as pre-drying for the two-stage vacuum convective drying (VCD) process.

Table 2.2: Selected works on vacuum drying and product quality

Products	Drying Conditions	Drying Kinetics	Product Quality	References
Hempedu bumi (*Andrographis paniculata*, AP)	Varying temperatures of 40 - 60°C with vacuum pressures at 100 and 300 mbar	Effective moisture diffusivity values were ranged from 10-13 m^2 s^{-1}.	Nil	Hee *et al.* (2015)
Asian white radish (*Raphanus sativus* L.) slices	Varying temperatures of 40 - 60°C and slice diameter (4 and 6mm) with vacuum pressures at 100 mbar	Effective moisture diffusivity values were ranged from 6.92×10^{-9} to 14.59×10^{-9} m^2 s^{-1}	Nil	Lee *et al.* (2009)
Nettle Leaves (*Urtica dioica* L.)	Varying temperatures of 50 - 75°C at 30 -67 mbar	Nil	There is a loss in brightness after vacuum drying. The best drying temperature was 75°C in nettle leaves drying.	Alibas *et al.* (2007)
Coconut presscake	Varying temperatures of 65 - 75°C and slice diameter (2 - 4mm) at 87 mmHg absolute pressure	Effective moisture diffusivity values were ranged from 7.026×10^{-10} and 3.326×10^{-9} m^2 s^{-1}	Nil	Jena *et al.* (2007)

Pumpkin (*Cucurbita maxima*)	Varying temperatures of 50 - 70°C at vacuum pressures of 50 – 250 mbar	Effective moisture diffusivity values were ranged from 1.30×10^{-9} to 4.03×10^{-9} m^2 s^{-1}	Nil	Arévalo-Pinedo *et al.* (2006)
Apple, guava and potato	Fixed temperature of 45°C at vacuum pressures of 150 mbar	Nil	High firmness and h* value for all materials over prolonged drying time.	Hawlader *et al.* (2006)
Mango pulp	Varying temperatures of 65 - 75°C and slice diameter (2 - 4mm) at 40 – 67 mbar	Nil	Colour change depends on pulp diameter. The best drying temperature was 72.3 °C on pulp diameter lower than 2.6mm	Jaya and Das (2003)

2.1.3.3 Other drying techniques

Other drying techniques, such as superheated-steam drying (SSD), heat pump drying and infrared drying are briefly described as below. Some researchers attempted to improve the drying uniformity by solar drying, superheated steam drying and microwave drying, but could not commercialize due to capital intensive and economically unjustifiable (Blow, 1944). Most of the principle of these drying techniques were known to be able to dry materials without having any negative effects on the product qualities, with limitations to dry heat-sensitive materials. Several research studies conducted by Rosana (2001), Pronyk *et al.* (2004), Soponronnarit *et al.* (2006), Ezhil (2010), Mujumdar (2014) and Li (2018) on superheated-steam had shown the possibility to implement this dryer for fast drying purpose. High-temperature steam transfers the sensible heat to the products quickly, with moisture evaporated without diffusion resistance. Figure 2.3 below shows a schematic diagram of impingement drying with superheated steam:

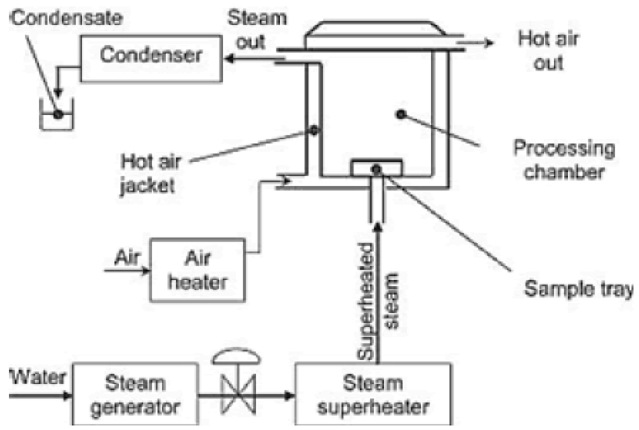


Figure 2.3: A schematic diagram of superheated steam dryer (Tang, 2000)

This old technology has increasing popularity for not only higher drying rate (Jangam, 2010), but better products quality (Ezhil, 2010). Steam boiler type of dryer has a promising future saving as it produced superheated steam by adding sensible heat to the saturated steam, which reduced at least 50% of energy compared to normal hot air drying (Pronyk, 2004). However, the drying of heat sensitive products by purely SSD caused severe deterioration, with both colour and texture of products affected by the high - temperature superheated steam (Nathakaranakule, 2007). Hence, this method is not suitable for this study.

Drying with heat pump dryer (HPD) was typically carried out at relatively low drying temperature (<65°C) by relying on dehumidification process. Figure 2.4 below shows a schematic diagram of heat pump drying process:

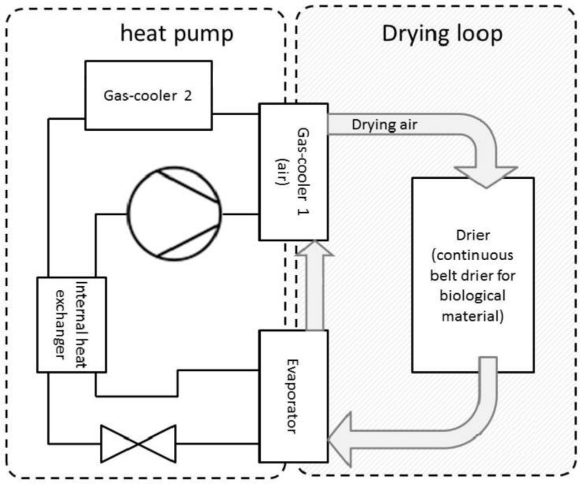


Figure 2.4: A schematic diagram of heat pump dryer (Michael, 2017)

There were many studies on heat pump dehumidifier drying but there was limited application for food drying (Perera and Rahman, 1997). The comparison of apple drying in a HPD and solar dryer showed that HPD had a higher effective moisture diffusivity compared to solar dryer (Aktaş *et al.*, 2009). As the heating temperature is low, heat sensitive materials can be dried with minimal quality deterioration, with high drying efficiency up to 95% can be achieved (Perera and Rahman, 1997). However, the drying rate was slow and prolonged drying at low heating temperature will lead to microbial growth and rubber skinning problems, especially for drying thick rubber materials (Rahman, 1985). Thus, low temperature drying via heat pump dryer is not a good option for rubber drying.

For infrared (IR) drying, it is a proven method to dry thick materials at low temperature with radiative heat. Figure 2.5 below shows a schematic diagram of infrared drying process:

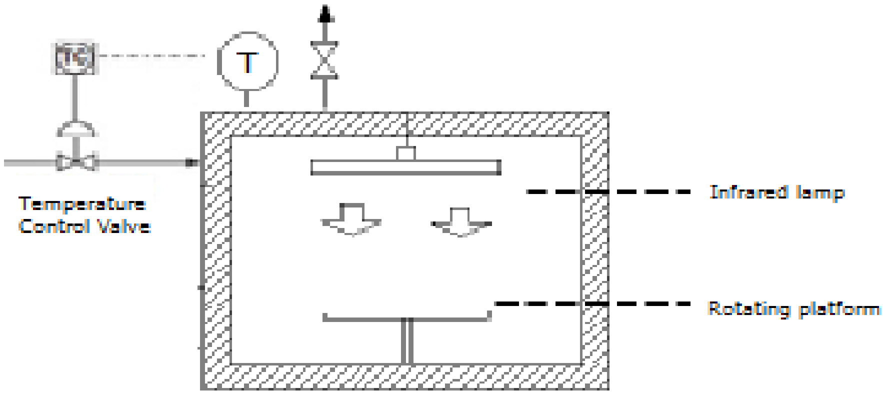


Figure 2.5: A schematic diagram of infrared dryer (Thanit, 2007)

In example, diffusion in potato during far infrared radiation drying was affected by radiation intensity level (Afzal and Abe, 1998). By using far-infrared as the heat source, the heat will penetrate the products via a form of electromagnetic wave, with wavelength of 0.75-100 µm, without any medium (Pan *et al.*, 2011). The increase of molecular vibration and the rate of moisture migration to surface as the energy of radiation is converted to heat (Ginzburg, 1969). However, the infrared radiation properties and the drying material must synchronize, in term of the material's emissivity and absorption wavelength for a success application (Onwude, 2016). When the infrared radiation properties and the material was not a good match, the total drying time will be prolonged. As compared to infrared drying techniques, vacuum and hot air drying showed more promising drying results and high drying efficiency. Table 2.3 shows the comparative advantages of infrared drying, vacuum drying and hot air drying.

Table 2.3: General comparison of infrared drying with vacuum and hot air drying

	Hot air drying	Vacuum drying	Infrared Drying
Products	Standard Thai Rubber 20 (Tirawanichakul *et al.*, 2009; Suchonpanit *et al.*, 2011)	Nettle Leaves (Alibas, 2007; Motevali *et al.*, 2011)	Dried Parboiled Rice (Das *et al.*, 2004; Motevali *et al.*, 2014)
Specific energy consumption value (MJ kg^{-1})	6.84 – 141.05	2.52 – 447.62	14.703-277.81
Operating temperature range (°C)	30 - 150	30 - 90	30 - 65
Operating %RH range	Variable (10 – 50%)	Low (0 – 5%)	High (60 – 90%)

- Remark: RH= Relative Humidity

When the three drying methods were compared with respect to the specific energy consumption in literature, vacuum drying has the lowest and the highest value, which are 2.52 and 447.62 MJ kg^{-1}, depending on the drying duration and vacuum pressure. The operating temperature for infrared drying is the lowest, which was not a satisfactory drying temperature for rubber drying process. The fabricated infrared dryer unit was unable to have good air ventilation, which lead to a high humidity content trapped in the dryer. Hence, vacuum drying and hot air drying methods were selected as the two-main drying medium for subsequent experimental works. The study in this research attempts to fill the gap between machinery design and dry rubber processing.

2.1.4 Intermittent drying technique

Other than the proposed two-stage vacuum convective drying technique, intermittent drying (rest period or tempering) is used for moisture redistribution purpose. It has been established that intermittent drying is the most energy efficient drying processes to prevent quality deterioration of drying materials by a controlled supply of heat on a predetermined intermittent ratio of heating time, to shorten the overall effective heating time (Baradey, 2017; Kumar, 2014; Kowalski, 2011; Chua, 2003; Chou, 2000; Pan, 1998; Zhang, 1991). Intermittent drying was commonly applied to heat sensitive materials improve the material properties. The examples of intermittent studies included drying of agro-products, such as timber (Salem, 2017), grain (Ghasemi, 2018), fruits (Defraeye, 2016) or other heat sensitive agricultural products, excluding crumb rubber. Pistachio nuts (Kermani, 2017), edible bird's nest (Gan, 2017), red pepper (Soysal, 2009), rough rice (Aquerreta, 2007) demonstrated that intermittent drying reduced the effective drying time and improve the physical attribute (colour and texture) compared to continuous drying.

Intermittent drying is a non-continuous drying process with tempering periods, which normally involved pulsation of drying temperature at various time intervals with some advantages in reducing deterioration of products quality due to heat treatment. Intermittent drying can be done in several ways, such as heat supplied intermittently (on-off method), removal of products to chilling storage area (dry aeration) and heat supplied following a fixed pattern (cyclic drying) (Cavusoglu, 2008). The tempering periods allowed more time for the moisture diffusion from internal to surface. A series of variation on the energy input by involving heating and resting period (on-off method) have been applied in the experimental works to determine the improvement of energy efficiency by comparing with the total effective drying time of continuous drying. As the tempering period would provide a time for the moisture diffusion, the intermittent drying process could be beneficial to the overall drying process and rubber quality. Previous pilot scale drying studied by various researchers (Table 2.1) also employed cyclic drying, an air reversal intermittent drying, to enhance the drying process. Considering of the skyrocketing fuel cost, intermittent drying which have been proven for cost reduction and quality assurance drying process would be a good alternative for the rubber processor without evolutionary changes required. When intermittent drying is applied at a reduced drying rate period, the product quality could be preserved with lower drying cost (Chua *et al.*, 2003). The existing literature regarding intermittent drying of other agricultural products is limited to traditional sun drying or low temperature drying process, which the drying temperature is lower than 100°C. Drying of crumb rubber is typically carried out at a temperature ranges from 105 – 140°C (Rahman, 1985). Any drying temperature that is lower than this range tends to result in unnecessarily long drying time.

In this study, it is hypothesized that by using intermittent two-stage vacuum convective drying (VCD) in the falling rate period of rubber drying process, the heat exposure to the rubber would be reduced and the moisture transfer from inner to surface could be enhanced (Chua *et al.*, 2003). The tempering period allows the internal moisture of rubber to be diffused to the surface with smooth moisture gradient, which in turn shortens the effective drying time and lowers the energy consumption, even though the overall drying time might be prolonged (Putranto *et al.*, 2011). The moisture diffusion, as determined by effective moisture diffusivity (D_{eff}), is predicted to increase with the tempering period. In comparison with continuous drying, the peak values of D_{eff} during intermittent drying should be higher. Thus, the intermittency of the drying process was chosen based on the lowest total effective drying time and acceptable product quality (colour, hardness, and stickiness).

2.1.5 Time-varying stepwise profile

Stepwise drying is one of the popular drying method, which can be categorize into step-up drying and step-down drying to optimize the drying process for better quality products (Gan *et al.*, 2017). It was studied extensively by several researchers on other materials, including Cuervo-Andrade and Hensel (2015), Chua *et al.* (2001) and Devahastin and Mujumdar (1999). One of the earlier research on optimization of air drying of food was carried out by Banga and Singh (1994), which the key optimization problems were pointed out.

Several studies have indicated that intermittent drying accompanied with step-wise temperature profiles could bring improvement in product quality (Kowalski and Pawlowski, 2011; Kumar *et al.*, 2014; Cavusoglu, 2008). In cabbage outer leaves study, the results showed that the stepdown drying process able to maximize the product quality and minimize quality degradation compare to continuous drying process (Lekcharoenkul *et al.*, 2014). In addition, Chua *et al.* (2001) also compared both step-up and step-down temperature profile for banana samples using a two-stage heat pump dryer for 300 minutes. In comparison of those stepwise air temperature variation, the researcher found that both step-up and step-down techniques improved the product colour.

Several other optimization studies on Naranjita (*Citrus mitis* B.) pomace (Delgado-Nieblas *et al.*, 2017), Scrophulariae Radix (Yan *et al.*, 2016), Banana (Omolola *et al.*, 2015), cauliflower (Gupta *et al.*, 2013), Tomato (Abano *et al.*, 2012), Artemisia absinthium leaves (Karimi *et al.*, 2012), Ganoderma lucidum slices (Chin and Law, 2012), olive leaf (Erbay and Icer, 2009), and parboiled paddy (Rao *et al.*, 2007) had showed an improvement in product quality, including phenolic content and colour change. The changing of operating parameters to maximize the quality retention and minimize the drying time while meeting the constraints was typically done for optimization of drying process (Banga and Singh, 1994; Banga *et al.*, 2003). To optimize the response, the effect of hot air drying temperature, air velocity and drying time to the moisture content, drying rate, energy efficiency and exergy efficiency of Artemisia absinthium leaves were studied (Karimi *et al.*, 2012). Similarly, Chin and Law (2012) studied the optimized drying parameters towards a better Ganoderma lucidum slices' quality.

The effect of process variable, including drying temperature (50.03–83.97°C) and air velocity (0.48–1.32 m s^{-1}) to the equilibrium moisture time (EMT) of Naranjita (*Citrus mitis* B.) pomace was optimized to preserve the product quality (Delgado Nieblas *et al.*, 2017). At high drying temperature, the temperature gradient between samples and heating air is high, which drying rate is increased and the EMT is reduced. However, the high temperature gradient typically leads to a rapid removal of moisture and collapse of rubber structure. Cuervo-Andrade and Hensel (2015) stated the stepwise drying process gave a better product quality without affecting the EMT for the dried medicinal plants. The changing points of temperature is typically based on the moisture content of product, to reduce the effect of colour change (Cuervo-Andrade and Hensel, 2015). The process dynamic of rubber drying process, including combination of heating temperature with point of change is determined by drying curve. Similarly, an optimized rubber drying carried out by Dejchanchaiwong *et al.* (2016) reduced the deterioration of rubber quality through using of mixed mode dryer. The mixed mode dryer used in the study is able to reduce 21.4% - 30.3% (w.b.) moisture content within 96 hours. A higher quality product is obtained when the drying time is reduced by 42.86%. Thus, time-varying stepwise drying process is an important step to improve rubber quality. In this study, step-up drying was adopted for transition of rubber from vacuum drying (VD) to hot air drying (HAD), then step-down during intermittent HAD to enhance the drying performance without the need to sacrifice product quality. Thus, stepwise drying helps to reduce the product quality degradation and maintain the drying kinetic.

2.2 Engineering properties

The most widely used experimental parameter for evaluation of drying process is through a sequential measurement of the mass change throughout the drying process. The mass loss is typically converted to moisture content and moisture ratio to further determine the drying kinetic and moisture diffusivity. Then, a systematic approach to improve the quality of dried rubber were decided based on a thorough analysis of drying kinetic of crumb rubber.

2.2.1 Drying kinetics

Any changes involved in drying process often governed by the environmental condition, including heating temperature, relative humidity and air velocity (Derossi *et al.*, 2011). As most bio-materials have low thermal conductivity, the penetration of the heating energy into a product often took a long period and thus, high temperature heating would quicken the heat and mass transfer process (Erbay and Icier, 2010). When there is a high temperature difference between the supplied heat and product temperature, there will be a higher driving force to remove moisture, which shorten overall drying time.

Below is the typical graph to indicate the drying mechanism of rubber:

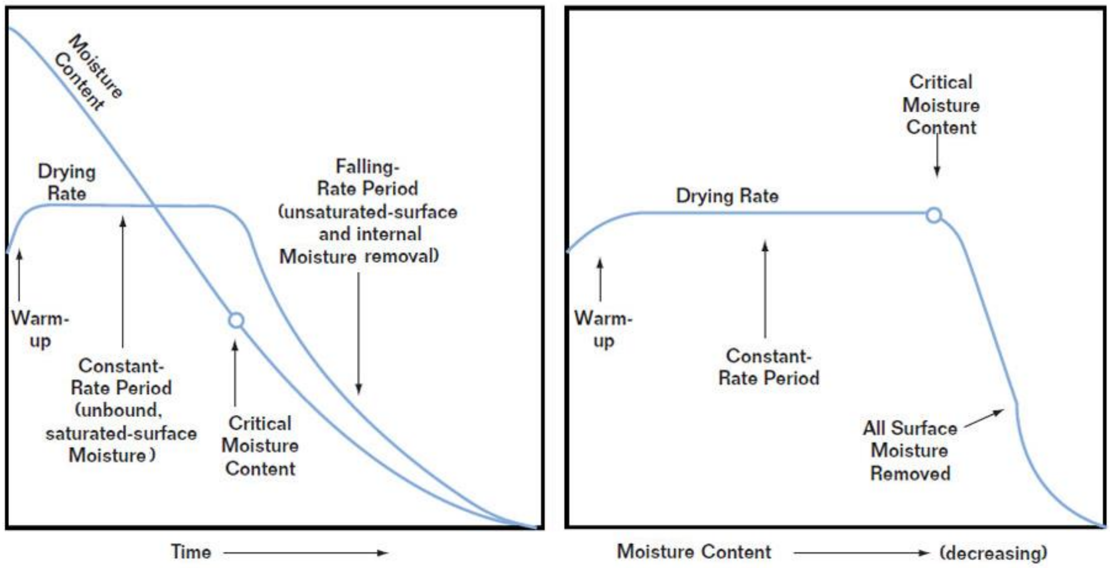


Figure 2.6: Characteristic of a typical drying curve (Rosen, 1983)

A common drying rate curve will consist of three different drying periods, which included initial transient period, constant rate period and falling rate period. However, the typical constant rate period was not shown in Figure 2.6 as the available free moisture on the rubber surface removed rapidly. At the beginning of any drying process, the product temperature will rise rapidly until the temperature difference is negligible. The increasing temperature will lead to a higher drying rate, which is shown at the initial transient period, based on Figure 2.6. Thus, the constant drying rate period does not exist in rubber drying curve.

After the point B, namely critical moisture content, the falling rate begin. The drying at this stage mainly depends on the product characteristics, internal perceived temperature and the vapour pressure of hot air until the second falling rate region begin. The time taken for rubber drying at the second falling rate period to reach point D, the equilibrium moisture content is often longer than the first falling rate period. This is due to the existence of external and internal mass transfer resistance that slow-down the drying process. Internal resistance of mass transfer is a limitation of internal moisture migration of bound moisture in a food product, which took a considerable longer time to overcome compared to the external resistance, which made the external resistance to mass transfer become negligible (Cavusoglu, 2008). However, formation of impermeable skin on rubber surface lead to an additional external resistance, especially when the moisture content is low or drying for a prolonged period (Rahman, 1985). Thus, it is necessary to consider a multi stage drying technique to dry rubber to a desired final moisture content of 5% d.b.

2.2.2 Moisture diffusivity

For rubber, all liquid movement is dominated by diffusion process, which follows the Fick's second law of diffusion. It is the main transport mechanism during the entire rubber drying process, including the initial heating time and falling rate period, hence the transport model should be inclusive of diffusivity. Both diffusivities (thermal and moisture) and mass transfer resistance could be a determining factor of the effective diffusivity rate (Coulson *et al.*, 1999). Most of the time, effective diffusivity of dried bio-materials could be very different depends on its shape and structure at microcellular level.

The estimation of diffusivity in rubber could be calculated by considering there is no chemical reaction between the moisture and rubber. Both thermal diffusivity and moisture diffusivity are important transport properties to find out the heat transfer in rubber drying process, and the estimation of diffusivity could be determined through modelling of the drying process. Although most of the diffusivity of liquids relies on correlations, based on Fick's law, there is no fixed method to determine it. By assuming constant diffusivity for a product, there are three types of diffusivity estimation equation that can be used, based on the surface geometry (Zogas, 1996):

Slab: $$\boldsymbol{MR} = \frac{\boldsymbol{M_t - M_e}}{\boldsymbol{M_{cr} - M_e}} = \frac{\mathbf{8}}{\pi} \sum_{\boldsymbol{n}=\mathbf{0}}^{\infty} \frac{\mathbf{1}}{(\mathbf{2}\boldsymbol{n}+\mathbf{1})^2} \exp\left[-\frac{(\mathbf{2}\boldsymbol{n}+\mathbf{1})^2 \pi^2 \boldsymbol{D_{eff} t}}{\boldsymbol{L}^2}\right] \quad (2.1)$$

Cylinder: $$\boldsymbol{MR} = \frac{\boldsymbol{M_t - M_e}}{\boldsymbol{M_{cr} - M_e}} = \sum_{\boldsymbol{n}=\mathbf{1}}^{\infty} \frac{\mathbf{4}}{\boldsymbol{b_n}^2} \exp\left(-\boldsymbol{b_n}^2 \frac{\boldsymbol{D_{eff} t}}{\boldsymbol{r_s}^2}\right) \quad (2.2)$$

Sphere: $$\boldsymbol{MR} = \frac{\boldsymbol{M_t - M_e}}{\boldsymbol{M_{cr} - M_e}} = \frac{\mathbf{6}}{\pi^2} \sum_{\boldsymbol{n}=\mathbf{1}}^{\infty} \frac{\mathbf{1}}{\boldsymbol{n}^2} \exp\left(-\boldsymbol{n}^2 \frac{\boldsymbol{D_{eff} t}}{\boldsymbol{r_k}^2}\right) \quad (2.3)$$

where MR is the dimensionless moisture ratio; M_t, M_e and M_{cr} is the moisture content at time t, equilibrium moisture content, and critical moisture content, respectively; D_{eff} is the effective diffusivity; L, r_s and r_k are the 50% thickness of slab, radius of cylinder and radius of sphere, respectively; n and b_n are the positive integer and characteristic root of zero order functions.

However, there are limited publications on the public domains that could be used to compare the results in current rubber drying study, thus the results found in current research would be a knowledge input for the domain of rubber drying studies.

2.3 The effect of rubber drying to quality changes

Raw rubber is a complex substance, which its properties changes (physically and mechanically) during the drying process and after the drying process are not always understood completely. Review of literatures showed that there is not much information on public domain that is related to both moisture dependent physical properties and mechanical behaviour of crumb rubber. The initial state of crumb rubber is uniformly dispersed white granules. Upon prolonged exposure to heat, light or oxygen, both the oxidative damage and non-oxidative thermal changes lead to browning of dried rubber. A typical good piece of dried rubber should be golden yellow or light brown colour. The colour change was part of the thermal degradation that lead to denaturation of protein (Park *et al.*, 1928). The thermal degradation through oxidative damage could be observed immediately not only from the colour change, but also the hardness and stickiness of the dried products. These properties changes are linked to moisture loss during drying. Aside from moisture loss, some volatile components in the crumb rubber were constantly evaporated off during drying, along with oxidative degradation. The loss of volatiles and moisture turned the dried samples into highly rubbery state (Alves-Filho, 2006). Thus, it is important to understand the effect of drying to visual attributes and textural attributes prior the systematic approach of experimental works. The changes in the product properties (physically and mechanically) for both standard drying and stepwise drying were investigated, to have a better grasp on how each drying parameter (vacuum duration and heating temperature) affect the product quality. Degradation and discolouration tends to occur during the drying process as the protein and fatty acid esters were hydrolysed by heat (Park *et al.*, 1928). The physical appearance, including colour and shape of the rubber products, such as browning, hardness and stickiness will change upon heat treatment.

2.3.1 Colour analysis

The colour change of rubber is one of the common technique to determine rubber quality (Suchonpanit *et al.*, 2011) and complete dryness of rubber particles (Rahman, 1985). Immediately after the drying process, the machine operator in a rubber processing plant will selectively pick and cut the selected dried rubber for visual inspection. This quality assurance procedure aims to check the presence of wet rubber particles, generally named as white-spots (Rahman, 1985). Thus, the white-spots problem need to be resolved by understanding the effect of drying parameters to colour change characteristic. Figure 2.7 below shows the white-spots that is often found during the drying process:

Figure 2.7: A visual defect (white-spots)

In this study, visual observation of white spots and colour measurement of browning activity was measured to determine the effect of drying methods to crumb rubber quality. A Lovibond colorimeter (Hunter Lab, USA) is utilized to quantify the results and study the effect of various drying methods to colour changes in dried rubber. The repeating occurrence of white-spots in the rubber indicated that the heat transfer to the rubber samples was insufficient, hence, prolonged drying was required to allow diffusion of moisture from centre to surface. However, an additional period of drying time will cause oxidative degradation and excessive browning of crumb rubber. Some manufacturers use chemicals to inhibit the enzymatic discolouration for prolonged drying (Nadarajah *et al.*, 1971). The main contributing factor of the browning or discolouration of rubber is due to the presence of amines (Morris and Sekhar, 1959). The studies on phenolic discolouration in natural rubber discovered that high thiols contents could suppress the discolouration due to tyrosine and indole (Yapa, 1976). However, thiols contents differ from clones to clones. As the crumb rubber used in this study is a mixture of various tree clones, it can be considered as a representative sample for colour change determination.

The white, discrete crumb rubber solid (Suchonpanit *et al.*, 2011; Rahman, 1985) became softened and dark yellow or brown in colour (Siriwardena *et al.*, 2010; Suchonpanit *et al.*, 2011) upon exposure to sunlight or high temperature drying. Suchonpanit *et al.* (2011) reported that the rubber turned darker and stickier after drying at higher temperature (44°C) for 111 hr, compared to samples that was dried 12.6% longer drying time at lower temperature (39.2°C). Siriwardena *et al.* (2010) measured the colour changes at various drying conditions and found that the rubber’s colour turned from light honey brown to brown colour after drying for 120 hours. Natural rubber, if unvulcanized (at raw state) and not added with antioxidants, is particularly prone to oxidative deterioration and browning due to reaction with oxygen in the air. Nevertheless, the inherent antioxidants in natural rubber are high enough to withstand high temperature drying procedure (Nadarajah *et al.*, 1971).

The ISO standard (ISO 4660:1999) for technical specified rubber (TSR) – Latex grades (L) based on the Lovibond Chart below (Figure 2.8) is limit to a maximum of 6.0 Lovibond Units (Suchonpanit *et al.*, 2011). The rubber should remain yellow to gold colour throughout the drying process. However, the lightness, L* of rubber became lower when the drying temperature is higher than 54.2°C, which the rubber colour quality was degraded to 4.0 (Suchonpanit *et al.*, 2011). The standard reference method for rubber colour measurement is based on the Lovibond chart in Figure 2.8 and Table 2.1 below:

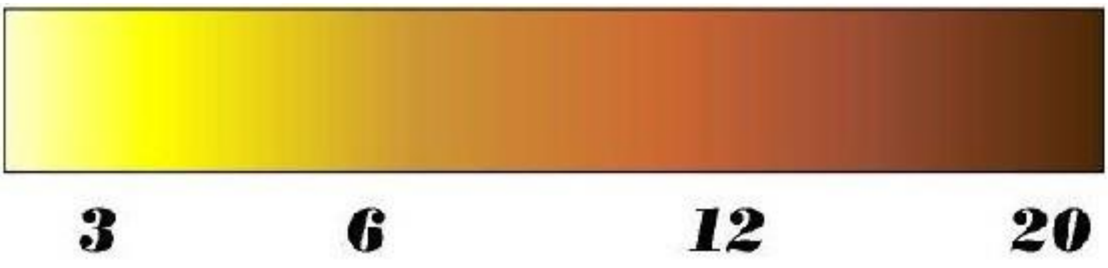


Figure 2.8: Standard reference method of Lovibond Chart (Lovibond, 1908)

Table 2.1: Lovibond units and basic colour

Basic Colour	Hue	Lovibond Units
Yellow	Light	2.0 – 3.0
	Medium	3.0 – 4.5
	Gold	4.5 – 6.0
	Deep Gold	6.0 – 7.5
Amber	Light	7.5 – 9.0
	Copper	9.0 – 11
	Red/ Brown	11 – 14
Brown	Light	14 – 17
	Medium	17 – 20
	Dark – Light Black	20 – 25
Black	Start of Full Black	>25

Despite that, the standard rubber colour that was used as referencing samples in this study is brown in colour (14 – 20 Lovibond Units). This is because TSR 20 tested in this study is not of latex grades. The inherent dirt and ash content in the crumb rubber lead to a poorer colour quality. Hence, the browning of crumb rubber in this study was assessed based on the standard rubber colour via colour space CIELAB, instead of the Lovibond units stated in Table 2.1. According to ASTM D3157, dried rubber samples can also be tested with Lovibond colorimeter or a HunterLab Spectrophotometer (Siriwong *et al.*, 2015).

Most of the research on rubber colour quality was based on Lovibond colour scale to determine the intensity of rubber colour (Subramaniam, 1987). A study on producing light-colour natural rubber used thin layer chromatography (TLC) method to determine the colour change (Boonyang and Sakdapipanich, 2014). However, the extraction of colour through TLC method is not commonly practice due to the time-consuming procedure. The extraction procedure also included heating of the extracts up to 180°C, which further enzymatic browning might be introduced during the heating process. Thus, colour measuring with TLC method was not the best option.

Typically, the colour changes depend on the drying temperature and the drying time. The browning of rubber could indicate the suitability of the selected drying techniques and the rate of moisture evaporation. Many studies have been done on the effect of heating temperature and drying time to colour change (İlter *et al.*, 2018; Doymaz, 2017; Aral and Beşe, 2016; Łechtańska *et al.*, 2015; Wojdyło *et al.*, 2014). Generally, the reduction of drying temperature and drying time would give a better visual characteristic. For all the processing of rubber with grade number (TSR-5, TSR 10 or TSR-20), which originated from cup lump, colour is not a mandatory attribute in the rubber grading standard. However, the measurement of the colour change is important in this study as it helps to determine the right drying conditions in a quantitative manner. Colour parameters is controllable through a comprehensive study on the onset browning time for rubber drying at different drying temperatures. Therefore, a study on the manipulation of drying temperature would be able to identify the timing for optimum colour change, which the moisture content and browning time are determined for the change point of temperature during stepwise drying.

2.3.2 Texture analysis

Texture analysis of the rubber material is correlated to two important properties, which is stickiness and hardness. Both textural attributes (hardness and stickiness) are physical-mechanical properties that is important for crumb rubber. Rubber is well known for its high tackiness or stickiness at its raw state. When the rubber was in its crude state, it turned tacky or sticky when stored at hot conditions or under sunlight due to the oxidation process or depolymerization. As mentioned by Bussemaker (1964), tackiness is one of the important properties that was desired by the rubber industry. Typically, a high molecular weight crumb rubber would develop good tack and high cohesive strength compared to most synthetic rubber due to the ease of rubber molecules diffusion across the interface at certain contact time (Bryan, 2001). Even though texture is not a mandatory attribute in rubber grading standard, but texture properties of rubber material insight into how samples behave during the mastication process. The texture properties of rubber, namely, hardness and stickiness, can be characterized through texture analysis. This method can quantify multiple textural parameters with a single sample, which is a useful method to measure the textural changes during rubber processing.

Hardness is one of the most widely measured properties in rubber materials, which represents the elasticity (Pornprasit *et al.*, 2016). Crumb rubber is an unvulcanized rubber, with a hardness value typically around 20 to 30, typically tested by using a bench hardness tester (Durometer) for its shore hardness value. The hardness of rubber varies depending on its clone origin (Pusca *et al.*, 2010). A low hardness level indicated that the rubber is very elastic and soft, which has a high level of adaptability during mastication. The degradation of the insoluble fraction, macrogel to microgel during drying process also affected the textural attribute of rubber, such as hardness and stickiness (Ehabe and Bonfils, 2011). The increase in rubber gel content and average molecular weight leads to hardening of rubber sample (Rolere *et al.*, 2016; Li *et al.*, 1998; Gan and Ting, 1993). In short, the hardness level of the dried crumb rubber sets the limit to its practical applications.

However, a rubber without filler materials does not have much application. A reinforcement of the rubber through vulcanization process gives increased higher hardness and tear strength, but decreased resilience and flex-fatigue life due to the increase in cross link density of the vulcanized rubber. During drying, the rubber experience progressive hardening, due to the reaction between carbonyl group and rubber chain under the effect of amino acid catalyst that presence naturally in rubber (Bengtsson and Stenberg, 1996). This hardening effect is named as inter-chain crosslink formation, can be accelerated by heating or oxidative process (Andrew *et al.*, 1946). There is a large increase in hardness when the samples are heated up to glass transition (G_T) temperature (Fakirov, 1999). Below the G_T temperature, the elastomer becomes a "glassy" solid that will fracture upon impact (Pearson and Yee, 1986). Thus, the hardness of the elastomer may change, depending on the drying temperature. Due to the complexity of natural rubber, there is no existing literature on the public domain that able to explain the observed characteristic of texture changes during drying. It is speculated that the heating temperature and drying time affected the hardness and stickiness of the rubber, which the changes reduce the quality of the final dried product.

For rubber stickiness, it measured the surface adhering to other materials (tack) and itself upon contact pressure. This criterion indicated how quick the component can stick to the rubber (surface adhesion) and how long the components can be hold together (cohesive strength), which is important for the tyre manufacturers. The measurement of physical-mechanical properties is done in relation to the processing conditions, for both continuous and intermittent basis. Determination of stickiness and hardness of the crumb rubber would increase our understanding to the processability of rubber products from another point of view. Hence, the measurement of texture properties changes during drying would be useful to predict the degradation of natural rubber.

CHAPTER 3

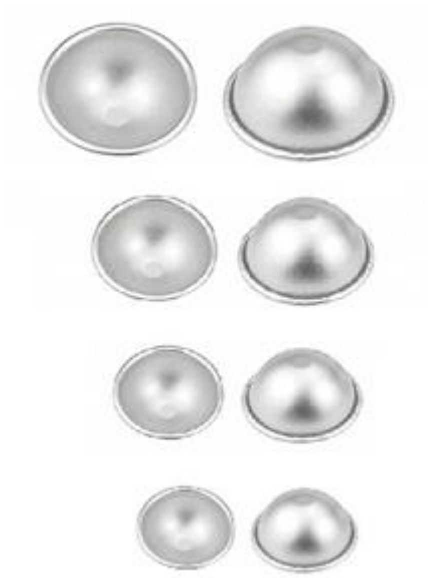
METHODOLOGY

3.1 Material

3.1.1 Sample preparation

For each experimental trial, crumb rubber (Hevea *Brasiliensis*) were purchased from local rubber processing factory, Kuala Pilah Rubber Factory Sdn. Bhd., located at Negeri Sembilan, Malaysia (Latitude, 3°07'02.1"N and Longitude, 102°13'27.6"E). All rubber samples were weighed and packed in a freezing bag prior storage at −18°C (0 °F). The quick freezing was to ensure the sample purchased could last for a longer period, as well as improve the consistency in testing by preserving most of the moisture originally exist in the rubber and avoid microbial growth.

All the crumb rubber samples had been washed before moulded to a similar crumb rubber size. Prior to and following the thawing process, the rubber samples was weighed to check for any significant mass loss. Thawing was performed by leaving the rubber sample at room condition of 25°C and relative humidity (RH) percentage of 55-60% for at least 30 min. The surface temperature of rubber samples was measured to determine the completion of thawing process. Then, the rubber samples were moulded into desired diameter using metal ball mould and each of the rubber ball was weighed to check its compactness. The rubber balls for each of the sample diameter of 10, 15, 20 and 50mm weighed about 2, 4, 6 and 30g. The metal ball mould used is as Figure 3.1 below, from the largest diameter to smallest diameter.



50mm

20mm

15mm

10mm

Figure 3.1: Rubber ball mould

By preparing samples in spherical shape using metal ball mould, a better comparison on the crumb rubber's drying characteristics at four diameters of the sphere rubber balls (10mm, 15mm, 20mm and 50mm) could be analysed. All different size of the rubber balls was weighed to a total of 30g for each trial of the drying experiments.

3.1.2 Equipment

The following equipment were used in this study.

Table 3.1: The list of testing instruments

Instrument Name (Model)	Functions	Supplier (Country)
Infrared thermometer (Fluke 62 Max)	To determine the rubber temperature	Fluke APAC (Canada)
Analytical Balance (AB204-S)	To measure weight up to 0.0001g	Mettler Toledo (United State)
Thermogravimetric Analysis (TGA)	To crosscheck the thermal decomposition temperature of sample	Mettler Toledo (United State)
Differential Scanning Calorimetry (DSC)	To determine the mechanical behaviour, plasticity of rubber	Mettler Toledo (United State)
Lovibond (LC 100)	To determine the colour changes in rubber drying process	Lovibond (United Kingdom)
Texture Analyser (TA.XT)	To determine the hardness and stickiness for dried rubber products	Stable Micro Systems (United Kingdom)

The mass loss of samples was recorded periodically by using analytical balance (Mettler Toledo, ME204, Malaysia, Mettler-Toledo (M) Sdn. Bhd.) with accuracy up to ±0.0001 g. The measurement of sample weight throughout the drying process was determined with increasing interval from 5 to 60 min with reference to the drying rate, until the moisture content decreased to 0.05 gH_2O gDM^{-1}. As the measurement of sample weight would relatively took less than 30 seconds, the steady-state drying condition would not be affected. In the intermittent drying experiments, the crumb rubber samples were rapidly weighed following same drying interval and returned to the dryer.

3.2 Drying techniques

A series of experiment were designed and split into three stages to improve the rubber drying process. In the first stage, three drying techniques: hot air drying (HAD), vacuum drying (VD), and two-stage vacuum-convective drying (VCD) were compared to determine suitability of two-stage VCD. Three different vacuum durations was compared to determine the effect of VCD to the drying kinetic of rubber. At the second stage, a suitable intermittent strategy was evaluated for the chosen two-stage VCD method (vacuum pre-drying for 30 min, followed by hot air convective drying), based on the drying kinetic results. A series of experiments were carried out by varying the combination of intermittent ratio (0.25, 0.50, 0.75 and 1.0) with intermittent duration (15, 30, and 45 min). Then, the selected intermittent VCD method was tested on various hot air convective drying temperatures (90, 110, 130 and 150°C) and various sample diameters (10, 15, 20 and 50mm). Lastly, a time-varying stepwise profile was introduced to the intermittent VCD drying method.

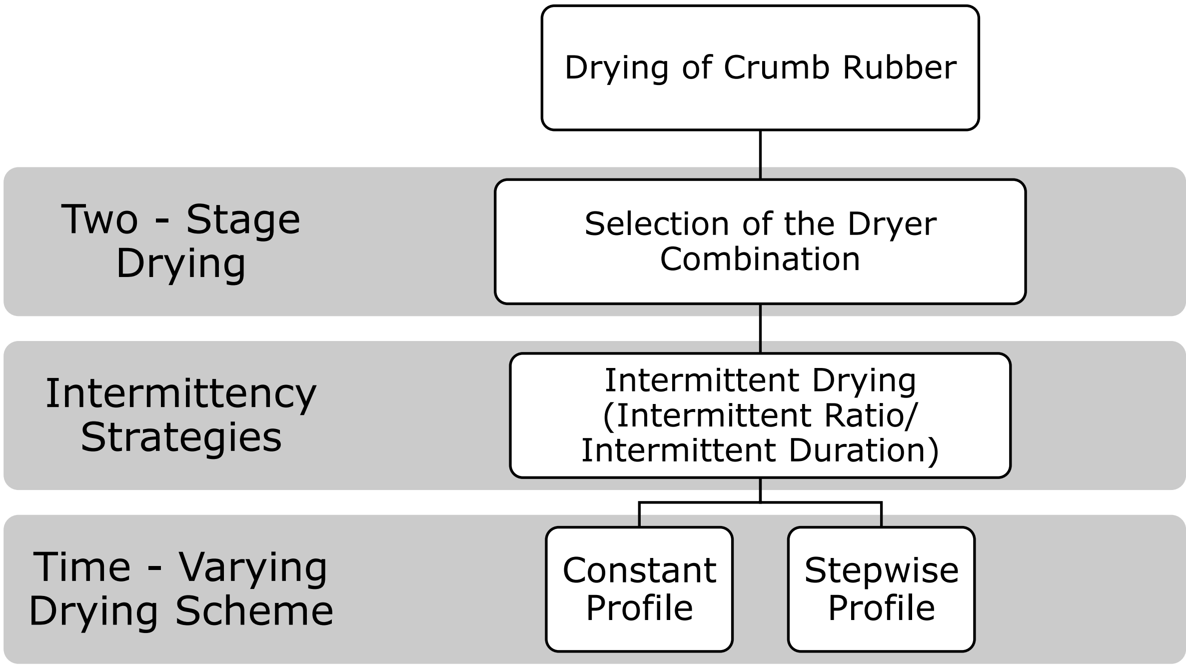


Figure 3.2: Experimental Design and Process

3.2.1 Comparison of three drying techniques

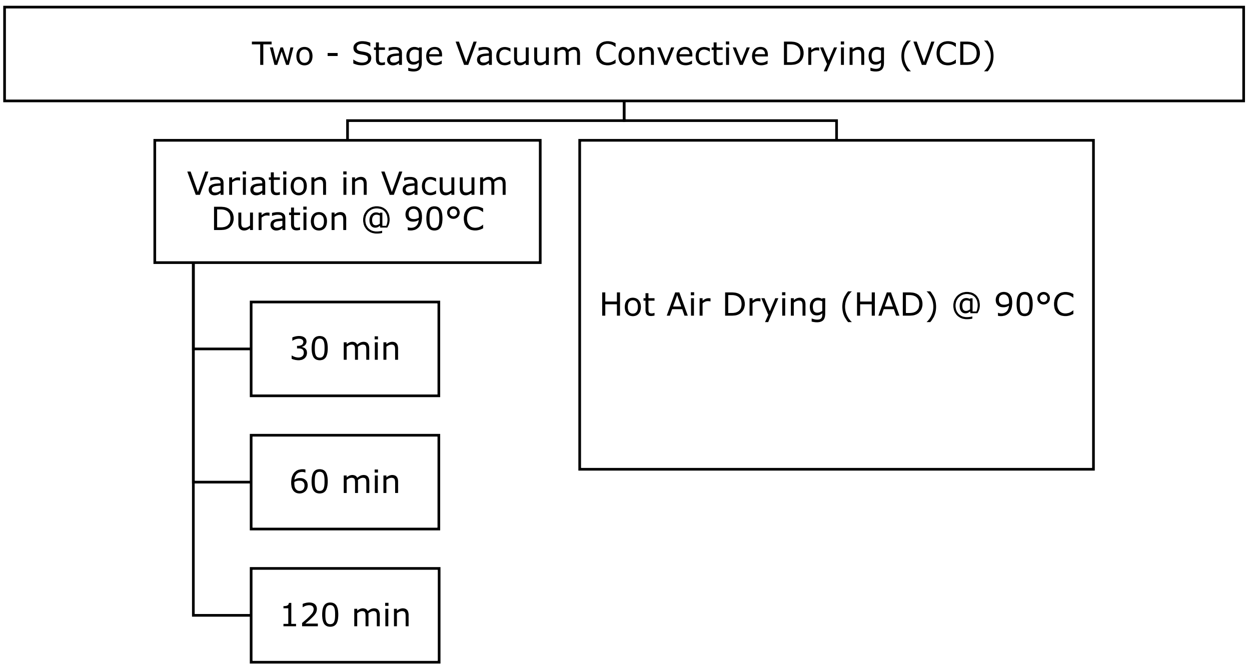


Figure 3.3: Determination the vacuum duration for two - stage VCD

The drying experiments were carried out with two types of drying equipment, which included a hot air convective dryer and a vacuum dryer. Once the desired temperatures (steady-state conditions) in the dryers were achieved, the rubber balls were distributed uniformly on a sheet of aluminium foil before placing onto the drying racks. As illustrated in Figure 3.2, rubber samples were processed via three stages with a thorough evaluation of the effect of vacuum duration, heating temperature and sample diameter to the drying kinetic and moisture diffusivities. The first stage of the experiment compared the hot air convective drying (HAD), vacuum drying (VD), and two- stage vacuum convective hot air drying (VCD). VD showed a promising quick drying of crumb rubber during the initial evaluation and was suitable to be used as pre-drying step for hot air drying (HAD). Two-stage vacuum convective drying (VCD) of crumb rubber at varying conditions of vacuum duration (30, 60 and 120 min), as shown in Figure 3.3 was made to determine the effect of vacuum duration to drying kinetics. After the selection of vacuum duration based on the least properties changes in crumb rubber, the experiment progress to second stage of the experiment.

3.2.2 Intermittent drying procedure

For intermittent drying, the heat was supplied intermittently (on/off) based on the beginning of falling rate period during the second stage of vacuum convective drying (VCD). Upon completion of the vacuum stage, the rubber samples were immediately transferred to the hot air convective dryer. The drying and tempering period of the hot air drying (HAD) was determined based on evaluation of intermittent ratio, ∝ and intermittent duration, while the intermittent cycle was based on the total time taken to reach the second falling rate region. The intermittency ratio is defined by the equation 2.1 as follows, where: t_{on} is the total time with heat input and t_{off} is the total time without heat input.

$$\text{Intermittent Ratio, IR} = \frac{t_{on}}{t_{on}+t_{off}} \qquad (3.1)$$

For the intermittent drying scheme, three variable levels for each independent variable (intermittent ratio and intermittent duration) were determined. For intermittency ratio, three variables, 0.25, 0.50 and 0.75 of heating duration over one intermittent cycle were considered; while, for intermittent duration, the three variables tested was 15, 30 and 45 min. Thus, a total of 9 intermittent drying scheme were undertaken for the study. Three replications of each experimental works were carried out in a random order. Figure 3.4 below shows the design of experiments for intermittent drying strategies, based on two independent variables.

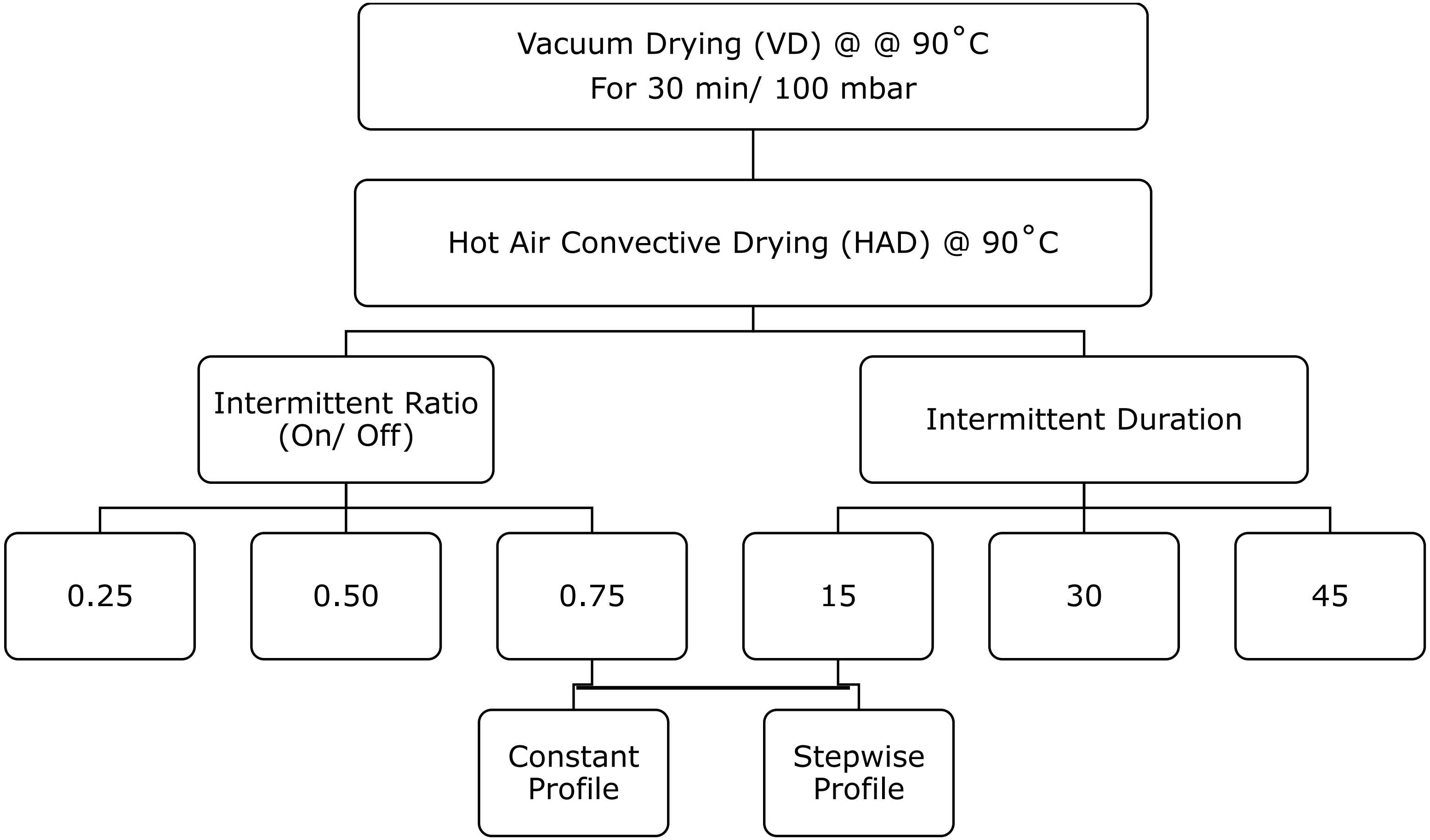


Figure 3.4: Intermittent Vacuum Convective Drying (VCD) Procedure

Drying of the intermittent drying scheme was carried out at 90°C by using two-stage vacuum convective drying VCD with a fixed vacuum duration of 30 min (V30). The dryer was 'on' during drying period while it was kept 'off' during tempering period. A calculation of effective drying time was based on the 'on' period, until the samples reach 5% (d.b.) moisture content. The selected intermittent VCD scheme was tested on the various hot air drying temperature and sample size. Details setting for drying of rubber is shown in Figure 3.5 below.

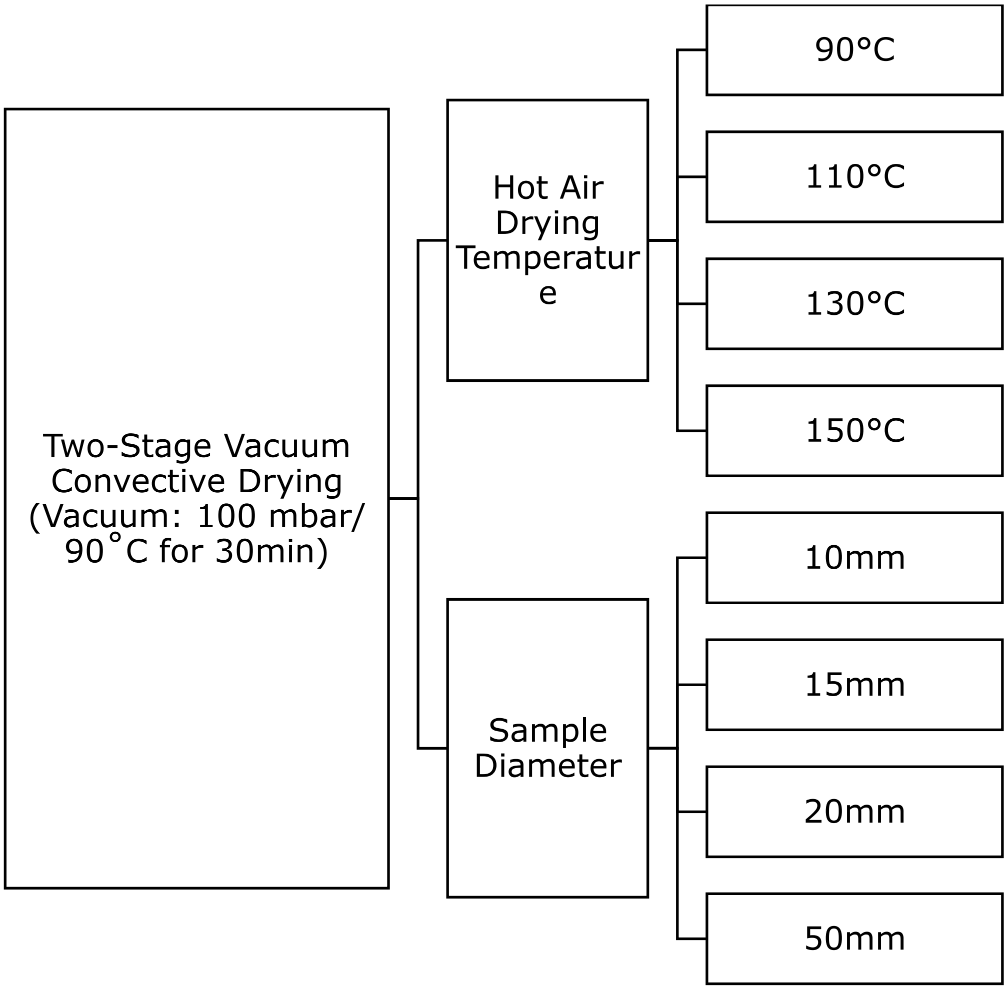


Figure 3.5: Evaluation of sample size and HAD temperature

A preliminary evaluation of rubber drying characteristic showed that prolonged drying of crumb rubber at low temperature lead to poor product quality, hence only high temperature drying (>90˚C) was considered. The drying conditions (drying airflow and relative humidity) used throughout each intermittent drying experiment were fixed at 1- 5 m s^{-1} and 0 – 5%. A comparison of the heating temperature of hot air convective dryer at 90°C (VAD90), 110°C (VAD110), 130°C (VAD130) and 150°C (VAD150) and sample diameter (10, 15, 20 and 50mm) were tested following the intermittent drying procedure. A comparison of sample diameter was done to determine how the thick rubber layer behave to the same drying conditions as thin rubber layer. The drying kinetics and colour changes at each drying temperature were determined. The evaluation of various hot air drying temperature and sample diameter on the selected intermittent drying scheme gave a better understanding of rubber drying characteristic. Browning development was determined through colorimeter and recorded as a function of time and moisture content for the next stage of experiment. Based on the L* value, when there was a drastic reduction in L*, the rubber weight and time were recorded for determination of stepwise profile.

3.2.3 Stepwise profile with intermittent VCD

By using optimizer tool in Minitab 18 to determine the most optimum process conditions for rubber drying, a chosen intermittent strategy, via on-off heating method was coupled with stepwise profile to improve the crumb rubber quality. Through careful regulation of the rubber temperature and browning time during the drying process, the minimization of total colour changes and hardness level with the maximization of stickiness level should therefore be possible. In this study, the effect of stepwise change of medium temperature, i.e., vacuum drying followed by hot air drying, on the evolution of colour and textural changes on rubber surface was investigated; the results were compared with those in the cases of continuous drying medium temperature.

The determination of browning time based on the monitoring of colour change in relation to the moisture content showed that the rubber undergone vacuum drying at 90 °C for 30 min and then hot air drying at high drying temperature of 150°C, followed by 130 °C and 110°C exhibit the most optimum drying pattern, with minimal quality changes. By regulating the duration of rubber at one specific drying temperature, based on the browning time, the rubber can be dried in a shorter duration without quality deterioration. The effects of the temperature changes at each step and the intermittent drying strategies on both drying kinetics and product quality were studied. The determination of the drying kinetic (drying rate and moisture diffusivity) of crumb rubber conducted with intermittent drying scheme for four heating temperature (90°C to 150°C) with four sample diameters (10mm to 50mm), allow development of a framework of the drying procedure. Figure 3.6 below shows the determination of engineering properties and rubber quality.

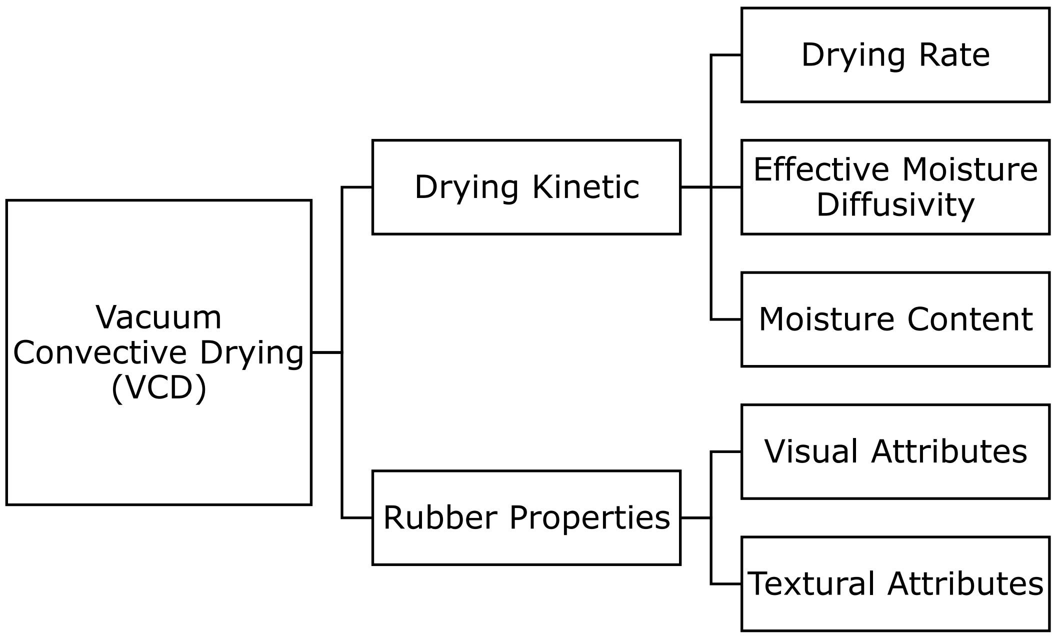


Figure 3.6: Fundamental Drying Characteristic of Rubber and Its Properties

Then, rubber quality (colour, hardness and stickiness) was determined to select the suitable drying temperature for the optimized drying period. Crumb rubber was heated sequentially at various process condition to find out the most optimum drying condition by the on-off heating method and stepwise drying method. Determination of the drying procedure was based on the effect of drying condition on the final moisture content, the drying kinetic, the visual attribute and the textural attribute of crumb rubber.

3.3 Drying equipment and specification

3.3.1 Hot Air Drying (HAD)

A 630 x 938 x 650 mm hot air convective dryer (Memmert, HCP 153, Germany), which composes of humidity chamber with natural convective ventilation system, primary heating assembly, drying compartment with a temperature controlling unit, and a heater of 1500 watt was used. The temperature setting was maintained at desired temperature and the amount of fresh air entering the humidity chamber would be regulated before experiment started. The sample was placed parallel to the air flow direction in the drying compartment with the width of 480mm, the height of 640mm and the depth of 500mm. During the drying process, the colour and sample weight were measured; but, after drying, the rubber samples were tested for physical changes (visual attributes and textural attributes) to ensure that the drying process did not affect the products quality. Figure 3.7 below shows the hot air convective dryer that was used for crumb rubber drying.

Figure 3.7: Laboratory scale hot air convective dryer

Hot air was channelled to the drying compartment where crumb rubber was placed three in a row with 20mm gap by large area all-round heating technique. The air temperature in the drying chamber was measured by using two separate Pt 100 temperature sensors and the drying temperature was varied from 90°C to 150°C. The humidity control was adjustable from 20% RH to 95% RH, but not functional at high drying temperature (>90°C). Both air velocity and humidity was fixed at 1 m s^{-1} and 0% RH during the drying experiments.

3.3.2 Vacuum Drying (VD)

Rubber drying was carried out by using a vacuum dryer (Memmert, VO 200, Germany). This vacuum dryer (550 x 400 x 600 mm) basically consists of a temperature controlling unit, a vacuum pump with pump capacity (at atmospheric pressure) up to 2.04 $m^3 hr^{-1}$, and an electrical heater of 1200 watt. The thermo-shelf is equipped with a Pt100 DIN class A temperature sensor with convenient integral 3-point temperature and vacuum setting. The temperature and air pressure of the vacuum dryer were recorded as reference once the dryer was in vacuum condition, which the minimum pressure limit and maximum temperature limit are 100 mbar and 90°C, respectively. Hence, the maximum heating temperature and minimum vacuum pressure was selected to investigate the maximum drying capacity of the vacuum dryer. A similar setting was practised by several researchers for drying heat sensitive material in vacuum condition (Lee, 2009, Keey, 1991).

The main drying mechanism in a vacuum drying is usually based on heat conduction and pressure differences. The dryer was heated up to desire setting temperature in advance before rubber sample was placed on the drying trays. Sufficient spacing of the sample was given to ensure proper air circulation inside the chamber. The door is sealed with silicone rubber without any leakage of air during vacuum condition.

Figure 3.8: Laboratory scale vacuum dryer

3.4 Determination of engineering properties

3.4.1 Determination of dry basis moisture content

The initial moisture content of rubber was determined for every testing batches to minimize the variance in crumb rubber sample. The moisture content was determined based on Association of Analytical Communities (AOAC) method. Triplicate samples of rubber (1g each) were weighed and dried in hot air convective dryer at 105°C for 24 hours. After a constant weight was obtained, the moisture loss was expressed in terms of percentage dry basis (d.b.) moisture content using equation 3.2:

$$\text{Moisture Content, } X\ (gH_2O\ gDM^{-1}) = \frac{\text{Weight of moisture}}{\text{Weight of dry solid present}} \qquad (3.2)$$

3.4.2 Determination of Moisture Ratio (MR)

During the drying process, all temperature data were measured with an infrared thermometer, with an accuracy of ±2 °C. The drying rate was determined from the changes of mass with time, while drying constant, k was obtained from the slope of the negative natural log of the moisture ratio, ln (MR) versus time. The moisture ratio can be calculated from the results obtained by following Fick's second law (Crank, 1975) in the theoretical model of drying, as shows in equation 3.3.

$$MR = \frac{M - M_e}{M_0 - M_e} \qquad (3.3)$$

Where: MR is the dimensionless moisture ratio; M_0, M_e and M is the initial moisture content, equilibrium moisture content, and moisture content at time t, respectively.

3.4.3 Determination of drying kinetic

After the initial period of adjustment in rubber drying process, the dry basis moisture content, X, would decreases linearly with time, t, beginning with evaporation of moisture from surface. The drying process was followed by non-linear decreasing of X until equilibrium moisture content. The drying rate of rubber could be calculated in functions of decreasing X in term of time, t with references to the weight of dry matter, DM and surface area of the solid, A as in equation 3.4.

$$N = -\frac{DM}{A}\frac{dX}{dt} \tag{3.4}$$

3.4.4 Determination of effective moisture diffusivity (D_{eff})

Fick's second law of diffusion is normally used to model the drying process to estimate the diffusivity value of moisture. For spherical object, the equation used should be as follows:

Sphere: $$\boldsymbol{MR} = \frac{M_t - M_e}{M_{cr} - M_e} = \frac{6}{\pi^2}\sum_{n=1}^{\infty}\frac{1}{n^2}\exp\left(-n^2 \frac{D_{eff}t}{{r_k}^2}\right) \tag{3.5}$$

Where: MR is the dimensionless moisture ratio; M_t, M_e and M_{cr} is the moisture content at time t, equilibrium moisture content, and critical moisture content, respectively; D_{eff} is the effective diffusivity; L, r_s and r_k are the 50% thickness of slab, radius of cylinder and radius of sphere, respectively; n and b_n are the positive integer and characteristic root of zero order functions.

After simplification, the temperature dependence of effective diffusivity (D_{eff}) can be expressed in terms of activation energy and temperature using the Arrhenius equation as given by the following equation (Henderson, 1961):

$$D_{eff} = Do\exp\left(-\frac{E_a}{RT_a}\right) \tag{3.6}$$

Where: D_{eff} is the diffusivity value; D_o is the diffusion coefficient; E_a is the activation energy; and T_a is the absolute temperature for rubber, respectively.

3.5 Characterization of Rubber Quality

The visual and textural attributes, such as colour index measurement, hardness and stickiness changes are the main critical physical analyses in this research.

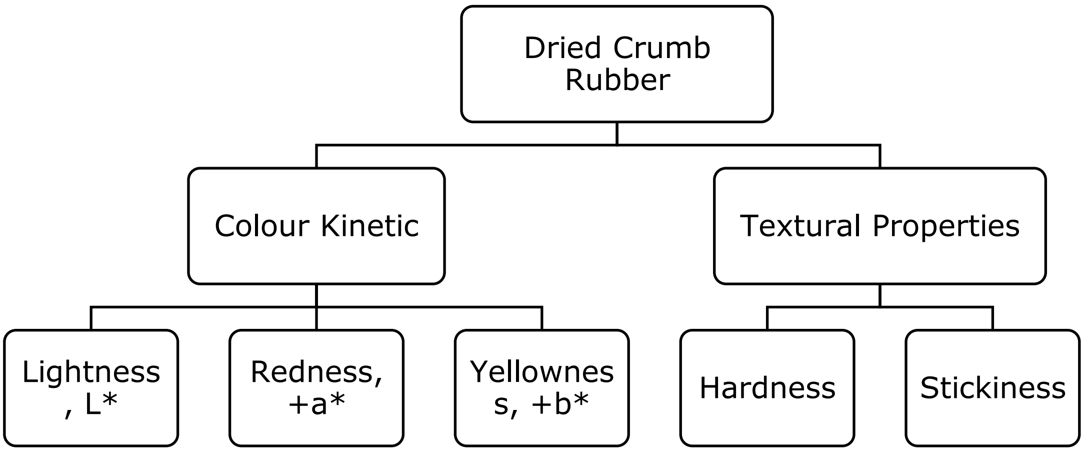


Figure 3.9: Quality Analysis of Dried Crumb Rubber

The dried rubber quality (both colour and textural properties) was compared to the rubber samples obtained from rubber processing factory. The L*a*b* colour space of standard referencing sample was measured and used for calculation of total colour change, while the hardness and stickiness value of standard referencing sample was measured for direct comparison with other experiment data.

3.5.1 Colour analysis

A colour measurement system (Hunter Lab, USA) was used to measure the three main CIE parameters, L*a*b* colour space as its perception of colour is closest to human eyes (Ivana, 2013). By using the colorimeter to examine three different spots on the rubber surface, the value of total colour difference (**Δ**E) was calculated automatically in comparison with the colour of standard referencing sample (Ivana, 2013).

The results were obtained as L*, a*, b* values which determine the different colour range, including brightness and darkness of the sample. The total colour difference was governed by the equation 3.7 as follows:

$$\Delta E = \sqrt{\Delta L^2 + \Delta a^2 + \Delta b^2} \qquad (3.7)$$

Where L* value determines the brightness (value is 100) and darkness (value is 0), a* measures green (negative) and red (positive), while b* represents blue (negative) and yellow (positive).

Calibration was done using a white tile prior to the sample analysis in triplicates. Figure 3.10 below shows the Hunter colorimeter used in this research:

Figure 3.10: Lovibond LC100 for colour measurement

3.5.2 Texture analysis

Texture analysis was performed by using TA-XTPlus Texture Analyser (Stable Micro Systems, Co., UK) through compression test. Figure 3.11 below shows the TA-XTPlus Texture Analyser used for the analysis of rubber hardness and stickiness:

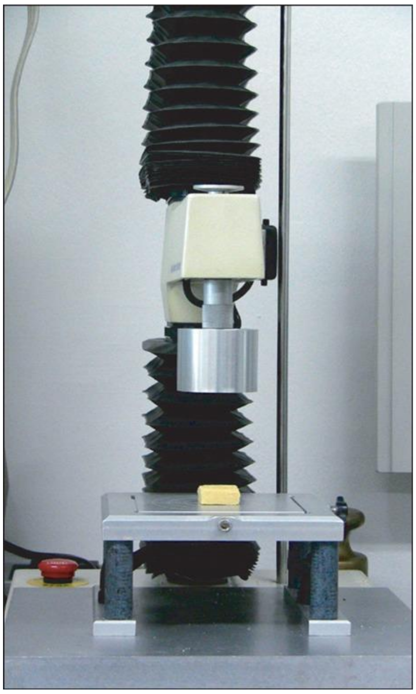


Figure 3.11: Texture analyser for texture measurement

Texture analyser was commonly used by researchers on cooked rice to determine the textural attributions of rice. Li (2017), Cheon (2016), Juliano (2016) and Han (2016) had studied the physio-chemical properties of cooked rice through evaluation of the hardness and stickiness value. Starch leaching of rice during cooking affects the cooked rice properties. Likewise, protein degradation of rubber during drying affects the hardness and stickiness of dried rubber. The dried rubber texture was dependent on the drying rate and moisture diffusivity. The rubber tends to return to its natural tackiness behaviours when dried under high drying temperature (Rahman, 1985). The mechanisms causing deterioration in rubber texture, such as lower stickiness and higher hardness was not reported elsewhere hence, poorly understand by many. By understanding the relation of the textural attributions of rubber with drying, rubber processors would be able to specifically design the products into required softness and stickiness to ease the subsequent processing stage.

The results are presented in chart form, with two compression cycles, which resulting in two maximum forces (F1 and F2) and two areas under the curve (A1 and A2). The two maximum force values on the graph are a measure of sample hardness in the first and second compression (hardness 1 and 2)/firmness. These values correlate with the amount of force required by the machine to process the rubber; the higher the value, the harder the rubber. The analysis of the degree of hardness of all rubber samples was carried out using a flat-ended cylinder stainless steel probe (75 mm diameter) with the deformation speed of 2 mm s^{-1}. Compression tests were performed to evaluate changes in the sample hardness and stickiness simultaneously. Once a trigger force of 5g had been detected on the surface of the sample the probe then proceeds to penetrate to a depth of 2mm. A maximum force reading was taken and used as an indication of hardness, at this distance. The probe then withdraws at maximum speed. The resistance to withdrawal from the sample was shown as the negative peak force and used as an indication of stickiness. The comparison of hardness and stickiness of samples dried with various drying condition was correlated to the rubber quality. Softer and high tackiness samples are preferable, hence, a dried crumb rubber with such characteristic would be considered as quality products.

3.6 Statistical analysis

The research was conducted using a single factor randomized design experiment. The results were analysed using Minitab Version 18 statistical software (Minitab, Inc., United State) and expressed as mean ± standard deviation of three replications. Statistical analysis, analysis of variance (ANOVA) and 95% confidence interval (95% CI) were performed with the Minitab 18 software to identify if there exists any significant difference for the rubber quality dried at various drying conditions. General Linear Model (GLM) was applied on the preliminary studies, intermittent studies and other comparison tests to determine the significant factors in rubber drying process.

CHAPTER 4

Results and Discussion

4.1 Comparison of drying methods

Preliminary study based on three drying techniques, including vacuum drying (VD), hot air drying (HAD) and two-stage vacuum convective drying (VCD) were conducted to evaluate the drying kinetics and rubber quality. The two-stage VCD experiments were investigated by drying crumb rubber via vacuum dryer (Memmert, VO 200, Germany) and a single display hot air convective dryer (Memmert, HCP 153, Germany, Evergreen Engineering & Resources). Control samples were dried using HAD at 90°C/ 1000mbar and VD at 90°C/ 100 mbar. A series of experiments were run on 50mm crumb rubber, with an initial moisture content of 47.26 – 68.44% to determine the suitability of two-stage drying method. The comparison and selection of the most suitable drying method from VD, HAD and two-stage VCD might be of interest in rubber processing manufacturers. The quality indices evaluated in the dried crumb rubber were: colour and plasticity retention index (PRI).

4.1.1 Drying kinetics of VD, HAD and two-stage VCD drying

Figure 4.1 shows the drying curves of crumb rubber dried by VD, HAD and two-stage VCD in this study, while a summary of the drying kinetics study of this preliminary study can be found in Table 4.1. Figure 4.1 reveals that the drying curves of single-stage HAD, VD and two-stage VCD are given by an exponential function. Comparison between the results in Figure 4.1 indicates that the moisture ratio reduction of VCD was faster than HAD and VD. Even though low-pressure condition resulted in sub-atmospheric pressure in the drying chamber, the moisture removal via vacuum drying slowed down when the rubber material was left with 70% moisture. This is due to the moisture transport rate from centre of rubber material to the surface, which govern the total drying time. The drying rates of rubber during the first stage of VCD (vacuum drying) had the exact same drying profile as VD, as shown in region 1 of Figure 4.1.

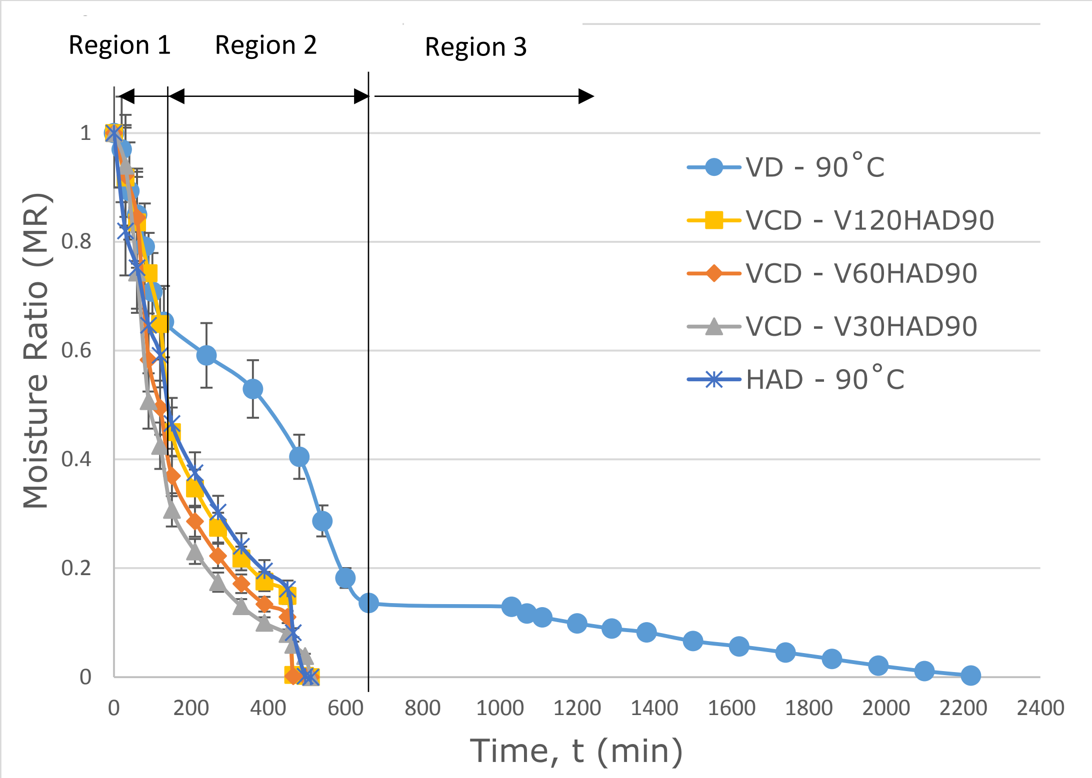


Figure 4.1: Moisture ratio versus time for vacuum drying (VD), vacuum – convective drying (VCD) and hot air drying (HAD)

Figure 4.1 shows the decreasing of moisture ratio with increasing drying time under various drying conditions (HAD, VD, and two-stage VCD). The drying kinetics pattern observed for both HAD and two-stage VCD was similar, as shown in region 2. This is due to the short vacuum drying duration and long hot air drying duration. The drying time, drying rate and final moisture content of the dried crumb rubber were measured, as tabulated in Table 4.1. The drying rate can be calculated from the gradient of the drying curves in Figure 4.1.

Table 4.1: Drying time, drying rate and final moisture content of different drying techniques on crumb rubber

Drying techniques	Drying time (min) to reach 0.40 (d.b.)	Highest drying rate ($gH_2O\ min^{-1}$)	Final moisture content ($gH_2O\ gDM^{-1}$)
HAD: Hot air drying at 90°C	330 [a]	0.00224 [c]	0.0909 [d]
V30HAD90: Vacuum (30min) - hot air drying at 90°C until EMC	270 [a]	0.00296 [c]	0.0737 [d]
V60HAD90: Vacuum (60min) - hot air drying at 90°C until EMC	330 [a]	0.00216 [c]	0.0649 [d]
V120HAD90: Vacuum (120min) - hot air drying at 90°C until EMC	390 [a]	0.00134 [c]	0.0731 [d]
VD: Vacuum drying (400mbars) at 90°C	600 [b]	0.000309 [c]	0.2618 [d]

Note: Means in the same column that do not share subscripts differ at $p < 0.05$ probability level.

From the results provided in Table 4.1, 10 hours was required to remove the moisture from crumb rubber to a 26.18% (d.b.) via vacuum drying. Vacuum drying (VD) took 81.8% longer time to reach the final moisture content of 0.2618 gH_2O gDM^{-1} compared to hot air drying (HAD), while vacuum – convective drying (VCD) resulted in 35 - 55% decrease in the drying time compared to vacuum drying. The drying rate for VD was slower after 130 min of drying time, when the moisture ratio of the rubber samples dropped below 0.65. This falling rate period for VD is due to the development of a non-porous surface skin on rubber when rubber was dried at low humidity condition for prolonged period (Rahman, 1985). The formation of hard, oxidised skin restricted the moisture diffusion, hence lowered the drying rate. In overall, VD's drying rate was 86.2% lower than HAD's drying rate. However, a combination of vacuum drying and hot air drying into two-stage vacuum convective drying (VCD) gave a synergistic effect, which the drying rate increased. For example, first stage vacuum drying for 30 min, coupled with second stage hot air drying at 90°C (V30HAD90) gave 89.6% and 32.14% higher drying rate than both VD and HAD, respectively. The two-stage vacuum convective drying increased the rubber's drying rate and reduced the total drying time taken to reach a final moisture content of 0.074% (d.b.). This might be due to higher diffusion rate of moisture and less degradation in rubber porosity (Rahman, 1985). During drying of rubber, some of the entrapped moisture are liberated and pores are developed. The degree of porosity depends on the conditions prevailing during the first stage of VCD. High drying temperature tends to increase rubber porosity and the collapse of rubber structure lead to the formation of oxidised skin. A drying rate versus moisture ratio chart was plotted in Figure 4.2 below.

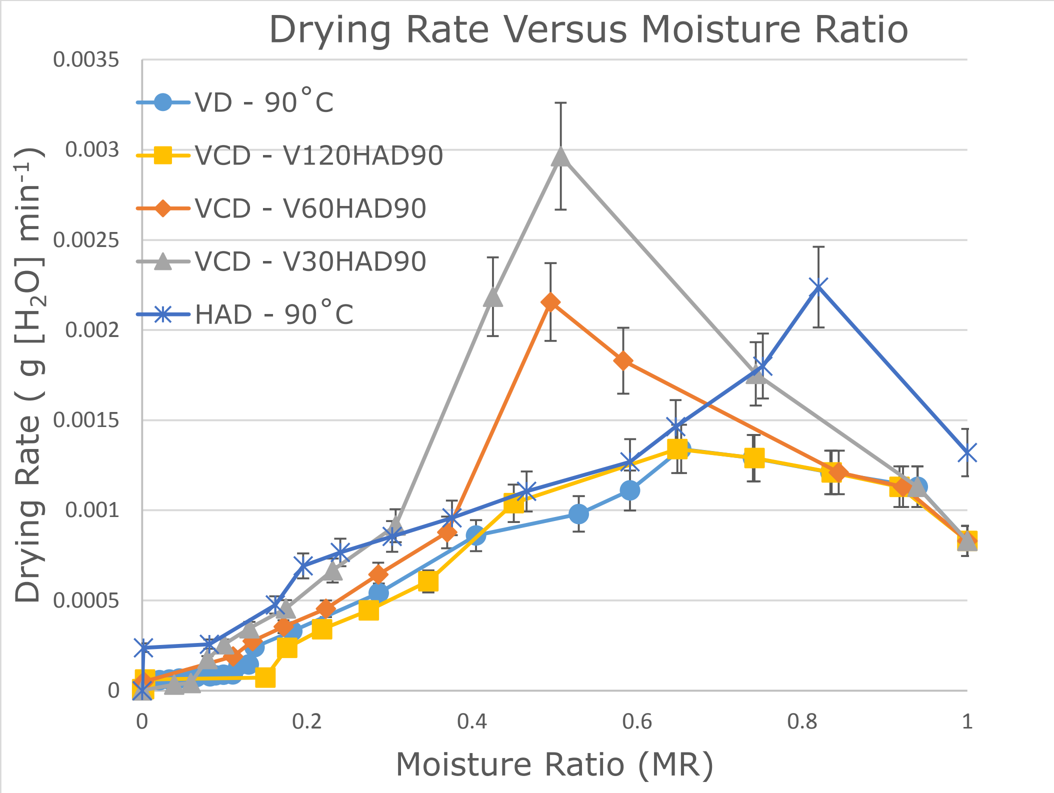


Figure 4.2 Comparative study on the effect of drying technique to drying rate of 50mm rubber samples dried with vacuum drying (VD), vacuum convective drying (VCD) and hot air drying (HAD) at 90ºC

For single stage continuous drying, such as vacuum drying (VD) and hot air drying (HAD), the drying rate curves exhibit as a smooth linear line, which consist of initial transient period, followed by falling rate period. The drying only occurred in falling rate period, without any constant rate period for both VD and HAD as the surface moisture was removed quickly. This finding is consistent with the study reported by Lee and Kim (2009). The drying rate curve for vacuum drying of radish slices also consisted of initial transient period and falling rate period, without constant drying rate period (Lee, 2009). This phenomenon is common for materials with diffusion – dominant drying characteristic, such as lemon slices (Torki-Harchegani et al., 2016), grape (Adiletta et al., 2016), garlic (Demiray and Tulek, 2014), okra (Doymaz, 2005), apricot (Toğrul and Pehlivan, 2003), and white bean (Adu and Otten, 1996).

For two-stage vacuum convective drying (VCD) in continuous basis, the drying rate curves consisted of several drying periods. For instance, V60HAD90 had an initial transient period and multiple falling rate period. Chong (2014) stated that multiple distinctive drying periods was common for multi-stage drying technique. Those inflection points only occurred for two-stage drying method and differs for varying vacuum drying duration. From Figure 4.2, a longer vacuum drying duration lead to a lower drying rate. The main reason was because of the formation of impermeable skin on rubber surface when rubber was dried at vacuum condition for prolonged period. The rubber structure collapsed when there is no air to fill up the moisture gap, lead to the difficulties for the remaining moisture to diffuse to rubber surface.

From the drying curve, the critical moisture content (CMC) of V30HAD90 appears at 30.80% (d.b.) and the second CMC when the moisture content dropped below 17.68% (d.b.). The evaporation at this stage was slower as there was no free moisture, which the drying rate in this second falling rate period decreased faster than the drying rate in first falling rate period (Brennan and Champhell-Platt, 1994). Similar findings were discussed in other multi-stage drying study and hybrid drying (Chong et al., 2014; Chua et al., 2005). In the step-up temperature study, the drying curve of apple cubes had a similar drying chart like vacuum – convective combined drying of rubber, with an increasing drying rate in the early stages of drying (Chong et al., 2014). This showed that the first stage is dominated by surface moisture evaporation, while the second stage is dominated by diffusion, which it depends on the moisture diffusivity from centre of material to the surface. The drying rate continues to decrease with a reduction in moisture content. The estimation of effective moisture diffusivity was calculated from the slopes derived from the linear regression of the ln (MR) against time, as shows in Table 4.2 below.

Table 4.2 Values of effective diffusivity attained at various drying techniques

Drying Techniques	Individual Effective diffusivity ($m^2 s^{-1}$) x 10^{-09}		Total Effective diffusivity ($m^2 s^{-1}$) x 10^{-09}
	HAD	VD	VCD
HAD - 90°C	2.8497 [a]	nil	nil
VCD - V30HAD90	2.4984 [a]	1.2378 [c]	3.7362 [d]
VCD - V60HAD90	2.0399 [a]	1.1897 [c]	3.2296 [e]
VCD - V120HAD90	1.7562 [b]	1.1568 [c]	2.9130 [d]
VD - 90°C	1.1399 [b]	nil	nil

Note: Means in the same column that do not share subscripts differ at $p < 0.05$ probability level.

From the effective moisture diffusivity results, vacuum drying (VD) gave the lowest diffusivity value, 1.1399 x 10^{-09} m^2 s^{-1}, whereas two-stage vacuum convective drying (VCD) at V30HAD90 gave the highest rate of 3.7362 x 10^{-09} m^2 s^{-1}. Vacuum drying rely on the contact surface area to provide heat energy to the sample, which lead to the long drying time (Woodroof and Luh, 1986). By using multiple dryers for drying with the use of pressure regulating drying technique, drying condition could be more versatile (Jin et al., 2017; Chou et al., 2001). Vacuum drying as a pre-drying stage were found to be statistically significant parameters ($p<0.05$) affecting the rubber drying process. Thus, the proposed method of two-stage vacuum convective drying is suitable for crumb rubber drying.

4.1.2 Rubber quality of VD, HAD and two-stage VCD drying

Table 4.3 illustrates the colour parameters L*, a* and b* results and plasticity retention index (PRI) of rubber samples undergoing various drying methods. The colour values were measured using a Lovibond colorimeter (Hunter Lab, USA), while the PRI values were tested using Wallace Plastimeter, in accordance with international standard ISO 2930:1995 or ASTM D3194-04. PRI quantified the oxidative resistance of the dried crumb rubber. There is no significant differences on the total colour change and PRI value for all drying techniques ($p>0.05$).

Based on Table 4.3, the rubber dried with two-stage vacuum convective drying (VCD) method had a lower lightness, L* value, with V30HAD90 had the lowest value of L* parameter among all. The drying of crumb rubber with vacuum setting had a positive influence on the total colour change. Hot air-dried rubber had a high total colour change value, 12.75, with a darker appearance, which the ΔE value was 28.4% higher than vacuum-dried rubber. The redness, a* and yellowness, b* value for rubber undergone hot air drying (HAD) were higher than VD and two-stage VCD methods. A high total colour change is typically due to higher oxidative degradation, which can be reflected in the plasticity retention index (PRI). At an oxygen – free drying condition, the oxidative degradation of natural rubber is minimal (Shashoua and Scott, 1993). Van-Amerongen (1955) reported that rubber do not degrade or experience lower physical changes when undergoes oxygen-free heating at 90°C. However, the PRI results for HAD was found to be the highest, 70%, similar PRI value with the control samples obtained from the factory (67.29%).

Table 4.3 Colour parameters L*, a* and b* and PRI of rubber samples

Drying Techniques	Continuous Drying			Total Colour Change	Plasticity Retention Index
	Colour parameter				
	L*	a*	b*	ΔE	PRI
HAD - 90°C	27.07 ± 0.60 ^{a}	6.33 ± 1.46 ^{c}	13.67 ± 1.76 ^{b}	12.75 ^{b}	70 ^{e}
VD - 90°C	25.50 ± 2.95 ^{a}	3.07 ± 1.53 ^{c}	9.53 ± 2.67 ^{b}	9.93 ^{b}	67 ^{e}
VCD - V120HAD90	11.00 ± 3.59 ^{b}	3.70 ± 1.99 ^{c}	6.70 ± 5.42 ^{c}	9.01 ^{b}	67 ^{e}
VCD - V60HAD90	8.40 ± 2.33 ^{b}	5.30 ± 4.10 ^{c}	8.60 ± 2.19 ^{b}	11.60 ^{b}	61 ^{e}
VCD - V30HAD90	5.80 ± 3.61 ^{c}	7.40 ± 1.37 ^{c}	6.60 ± 0.95 ^{c}	13.36 ^{b}	60 ^{e}
Fresh Rubber	50.53 ± 6.51 ^{d}	1.65 ± 2.55 ^{c}	13.35 ± 7.82 ^{b}	-	-
Standard Rubber	18.9 ± 3.96 ^{a}	7.10 ± 1.70 ^{c}	4.00 ± 0.35 ^{c}	-	67.29 ^{e}

Note: Means in the same column that do not share subscripts differ at $p < 0.05$ probability level.

The lower PRI value for VD might be due to the longer drying duration to achieve equilibrium moisture content, and shorter drying duration for HAD. Nevertheless, PRI value with a minimum 60% is suitable for tyre manufacturing industries to obtain consistent good quality tyres (Chou, 2001). Based on the statistical analysis (ANOVA), there is no statistically significant of the drying techniques to PRI values. Hence, any value in between 60-70% is considered acceptable. In short, rubber dried in the laboratory with various drying techniques were able to achieve the target PRI value. Based on the drying kinetic results, two-stage vacuum convective drying (VCD) was found suitable for crumb rubber drying as it dried faster. Therefore, two-stage vacuum convective drying (VCD) was chosen to be investigated as a potential drying technique for crumb rubber with high drying efficiency and low-quality degradation.

4.2 Intermittent drying results

In this study, the feasibility of applying intermittent on the second stage of the vacuum convective drying was carried out. The first stage was the vacuum pre-drying (V) of 30 min carried out under low pressure condition (100 mbar), which was found feasible during the preliminary study. The second stage was the hot air drying (HAD) at temperatures of 90ºC to finish the drying process. Three intermittent duration, ID (15, 30 and

45 min) and three intermittent ratios, IR (0.25, 0.50 and 0.75) were compared to determine the optimal period for reducing the effective drying time and avoiding possible quality changes (colour and texture). Intermittent ratio, IR = 1 is equal to continuous drying that are used as a benchmark drying method. In the subsequent section, the label as below will be used for discussion purposes. Intermittent duration, ID for 15, 30 and 45 min, will be labelled as ID15, ID 30 and ID45; intermittent ratio, IR of 0.25, 0.50, 0.75 and 1, will be labelled as IR025, IR050, IR075 and IR1.

4.2.1 Drying kinetics of two-stage VCD intermittent drying

Results for the effect of tempering period (intermittent ratio, IR and intermittent durations, ID) are shown in the form of drying curves and drying rates in Figure 4.3a, b, c and 4.5a, b, c, respectively, while Figure 4.4 shows the comparison of total drying time and effective drying time for varying intermittency.

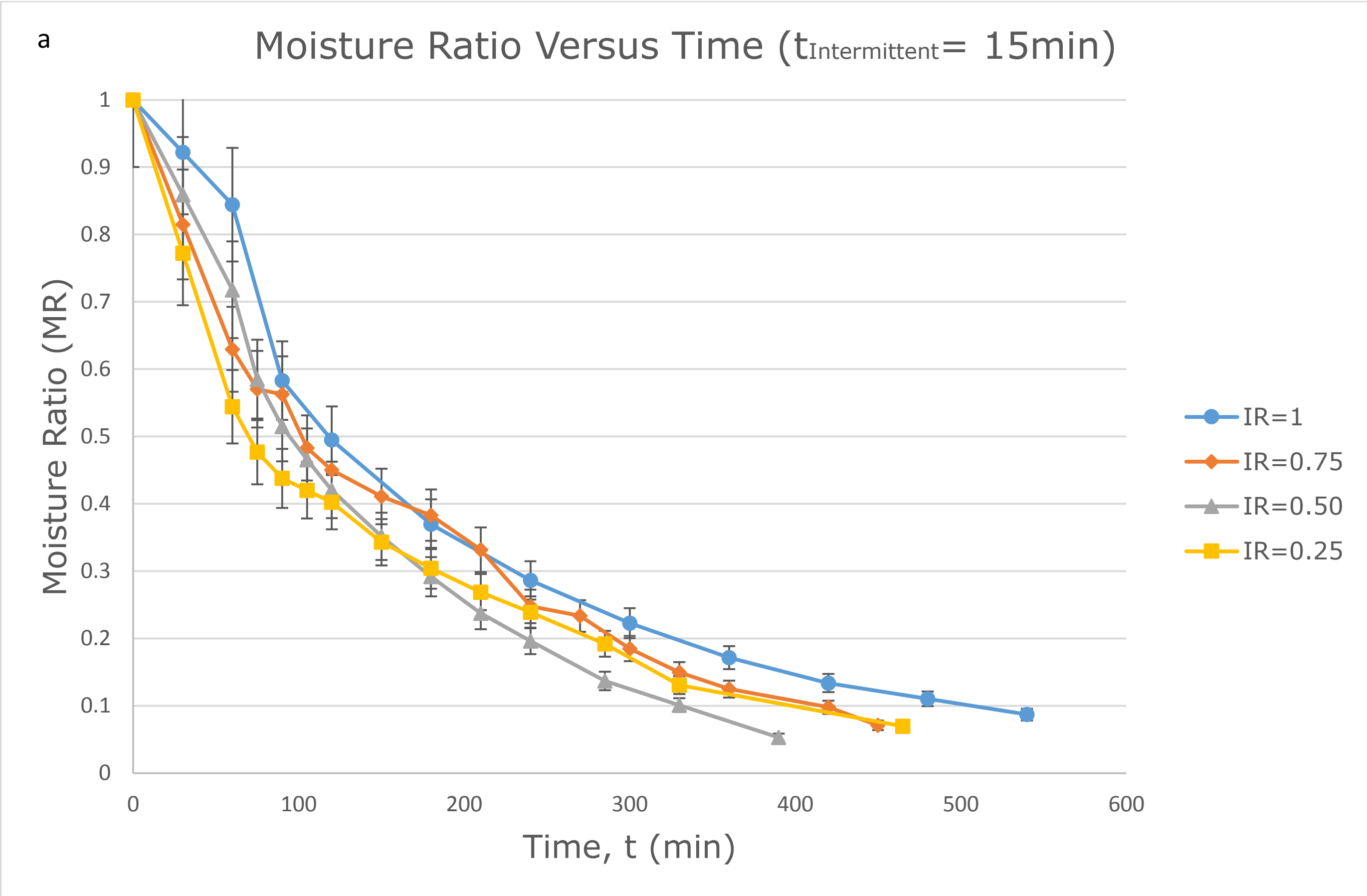
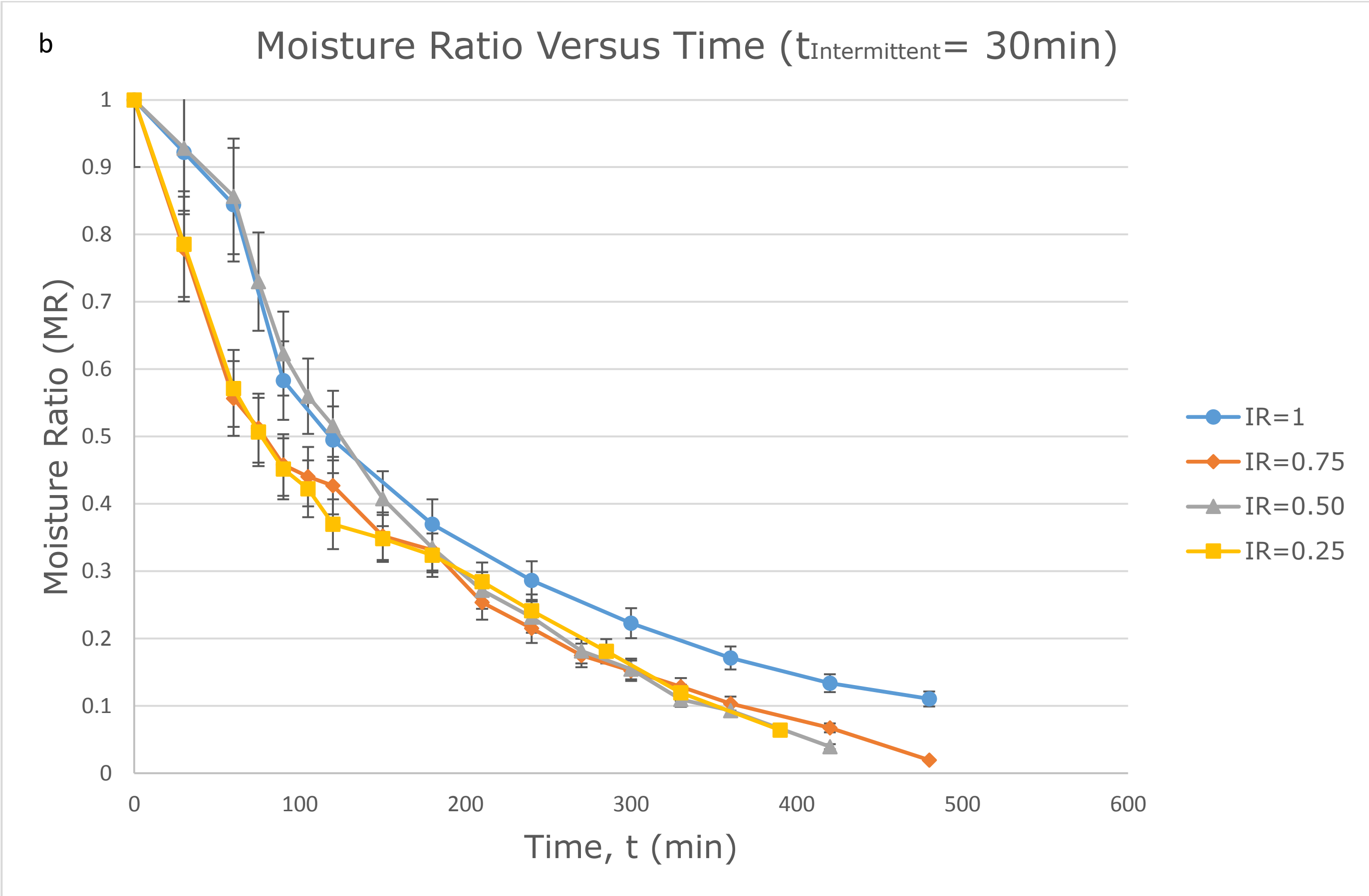


Figure 4.3a, b, c: Drying curve of rubber samples subjected to two-stage vacuum - convective drying (VCD) under various intermittent duration (15, 30, 45 min) and various intermittent ratio (0.25, 0.50, 0.75 and 1) at fixed temperature of 90˚C



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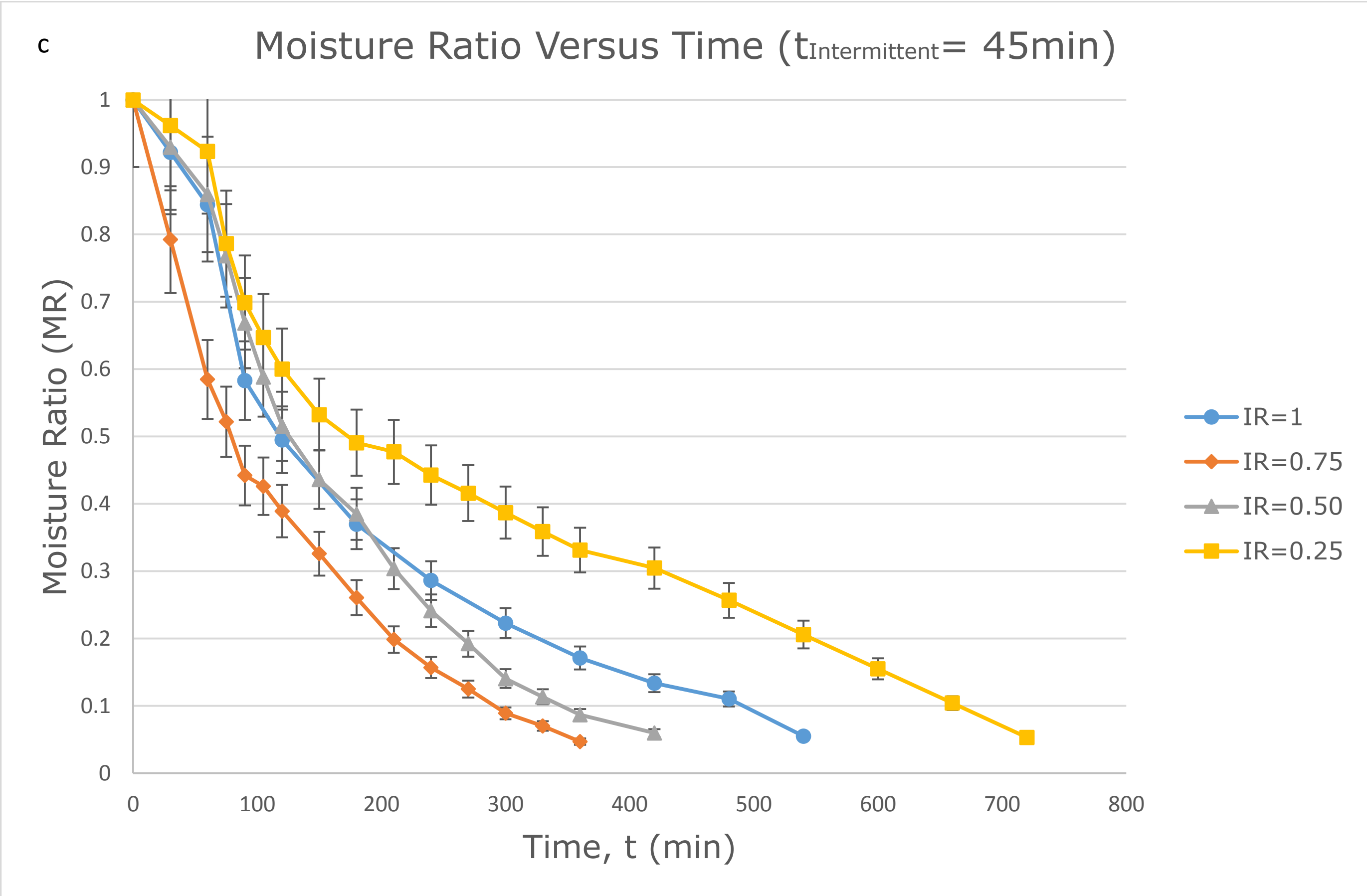


Figure 4.3a, b, c: Drying curve of rubber samples subjected to two-stage vacuum - convective drying (VCD) under various intermittent duration (15, 30, 45 min) and various intermittent ratio (0.25, 0.50, 0.75 and 1) at fixed temperature of 90˚C

Figure 4.3a, b, c shows the decreasing of moisture ratio (MR) with increasing drying time, t for varying intermittence. The drying curves were indifferent for all tests ran. From Figure 4.3a, drying of the rubber by vacuum convective drying (VCD) with ID15 with IR025 required the shortest drying duration while ID45 with IR075 required the longest drying duration in this study. The rubber sample took merely 60 min to dry to a 0.54 moisture ratio for intermittency of 15 min on, 5 min off (ID15- IR075) compared to the continuous drying, IR1 that required 120 min. However, the slowest drying was observed for ID45-IR075, which required 180 min to remove 50% of the rubber moisture. A summary of the total drying time and effective drying time of the dried crumb rubber were shown in Figure 4.4.

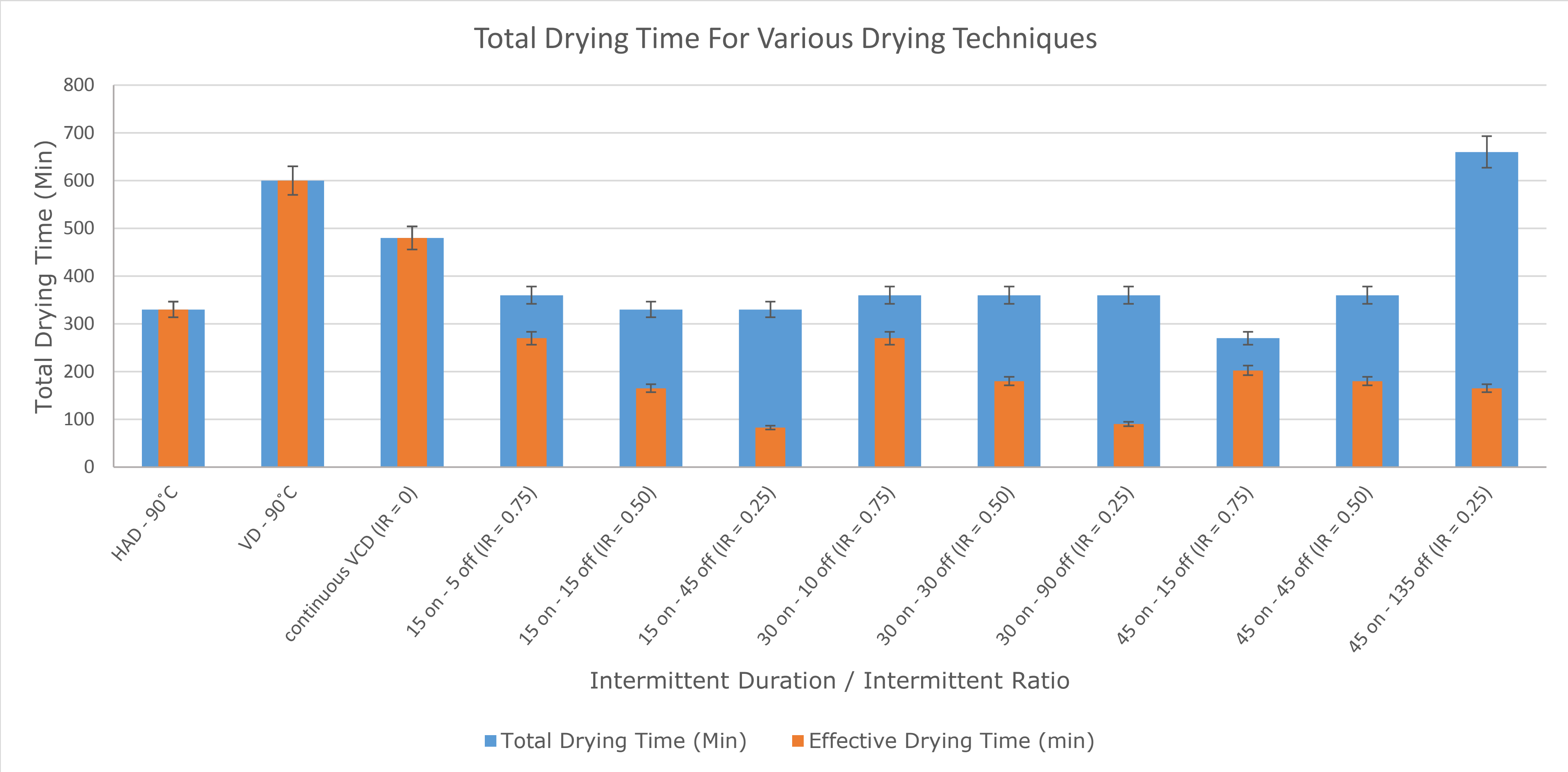
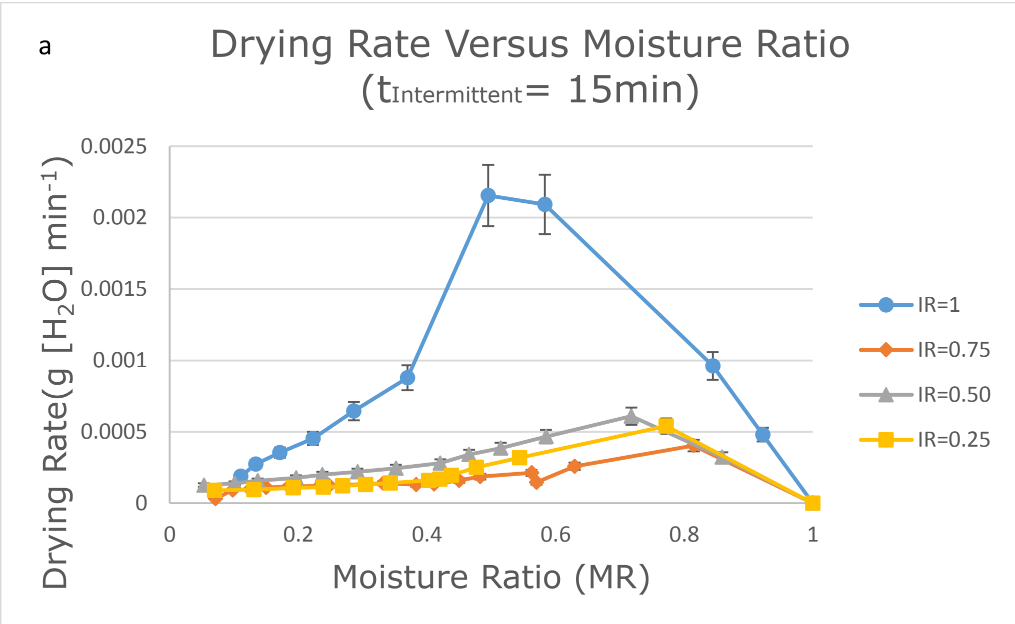
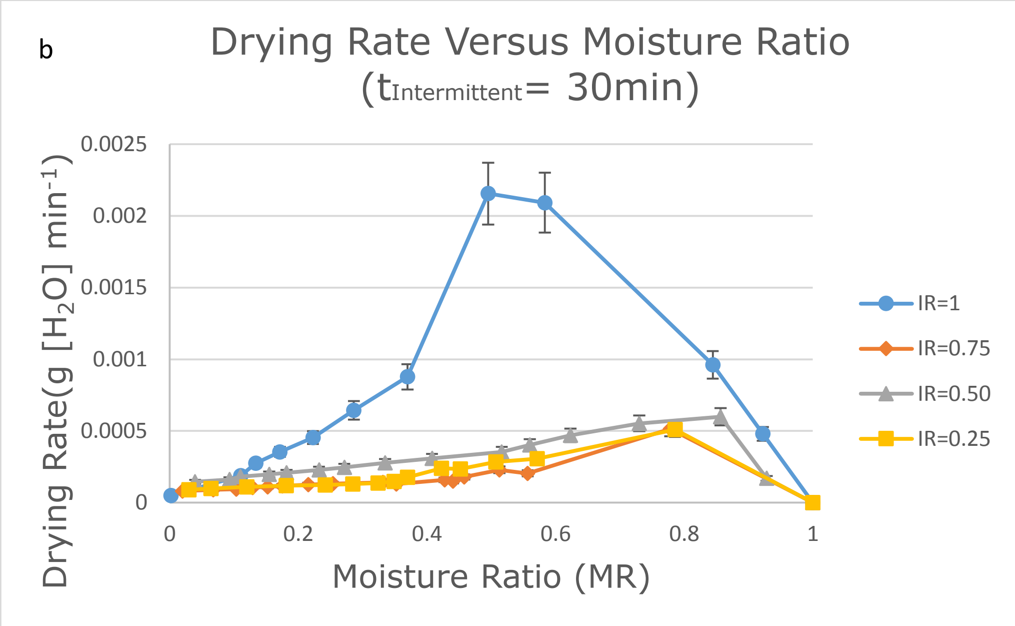


Figure 4.4: Drying time and effective drying time of different intermittent drying strategy

From Figure 4.4, the most efficient drying was found for the combination of IR025 and ID15, which only required an effective drying time of 82.5 min to reach final moisture content. Another interesting aspect of rubber drying is that the shorter ID15 dried faster than ID45. For example, the effective drying time was 50% longer for ID45 compared to ID15, at the IR025. The vacuum convective drying (VCD) with ID45 and IR025 required a total drying time of 660 min to reach 0.225 $gH_2O\ gDM^{-1}$, with a percentage difference of 31.6% and 83.9% compared to continuous VCD and intermittent VCD with ID45 and IR075.

It was found that the final moisture contents of samples dried by continuous VCD (ID = 0 min; IR = 1), intermittent VCD (ID = 15 min; IR050), intermittent VCD (ID = 30 min; IR050) and intermittent VCD (ID = 45 min; IR050) were 0.051, 0.229, 0.227 and 0.228 $gH_2O\ gDM^{-1}$, respectively. The final moisture content for the continuous VCD was lower as there is a higher heat stress. This is due to the continuous dried samples received more heat inputs to reach the latent heat of evaporation compared to intermittently dried sample. After pre-drying by vacuum, about 15.8% of moisture evaporated from the sample. The remaining moisture in the samples was dried with hot air drying (HAD) in the second stage of VCD. When the intermittency begins, the rubber continues to dry with the remaining heat retained inside the rubber material. However, the heat intensity is not as strong as the heat inputs from dryer, thus the final moisture content was four times higher than continuously dried samples. By increasing the drying temperature of hot air drying, the final moisture content for intermittent drying can be reduced to desire moisture content of 5% (d.b.). For better understanding of rubber drying characteristics during intermittency, a drying rate versus moisture ratio curve is plotted in Figure 4.5a, b, c.





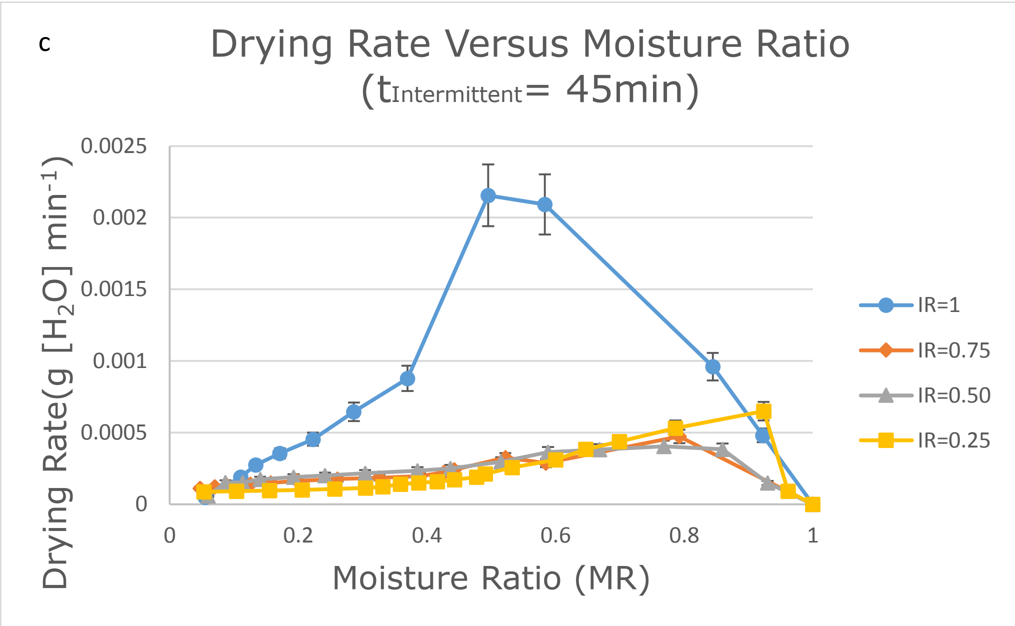


Figure 4.5a, b, c: Comparative study on the effect of intermittent ratio (0.25, 0.50, 0.75 and 1) and intermittent duration (15, 30 and 45 min) based on the variation of drying rate in respect of moisture ratio

According to Figure 4.5, a, b, c, all drying rate curves had two distinct drying rate periods, with initial transient period and falling rate period after the critical moisture content of rubber. The drying rate curves of continuous vacuum convective drying

(VCD), IR1, exhibits a significant rise in drying rate during the transition from hot air drying (HAD) to vacuum drying (VD). For rubber dried with various intermittency (IR= 0.25, 0.50 and 0.75), the increment of drying rate was at least 3.5 times lesser.

As shown on Figure 4.5c, the comparison of intermittent drying at various intermittent ratio, IR (0.25, 0.5 and 0.75), showed that the drying efficiency was low when the tempering period was long. Thus, the usage of long intermittent duration at high temperature drying condition was not suitable for crumb rubber. The rubber was unable to dry up to satisfactory level within a short time frame (< 240 min). The total effective drying time was prolonged for 50% when the IR was changed from 0.75 to 0.25, at ID45. From Figure 4.5a, the rubber dried under IR of 0.75 was 69.4% faster than 0.25, at intermittent duration of 15 min, ID15. This shows that a different intermittency strategy gave different drying rate and speed to remove internal moisture from rubber.

The two-stage VCD drying, IR 1, increased the drying rate of crumb rubber and dried the rubber to a 50% moisture before the falling rate began. On contrary, two-stage intermittent VCD drying (IR= 0.25, 0.50 and 0.75) had a lower drying rate and the falling rate period began when the rubber moisture was lesser than 90%. The off pulsating of heat input negatively affected the crumb rubber drying rate. This might be due to the low drying temperature at 90°C. The heat transferred to the samples was insufficient, lead to a low overall drying rate value. Thus, it was important to study tempering at higher drying temperature to improve the drying rate. The drying kinetic curves of two-stage intermittent VCD with drying temperature of 110, 130 and 150˚C were shown in Figure 4.6 and 4.7.

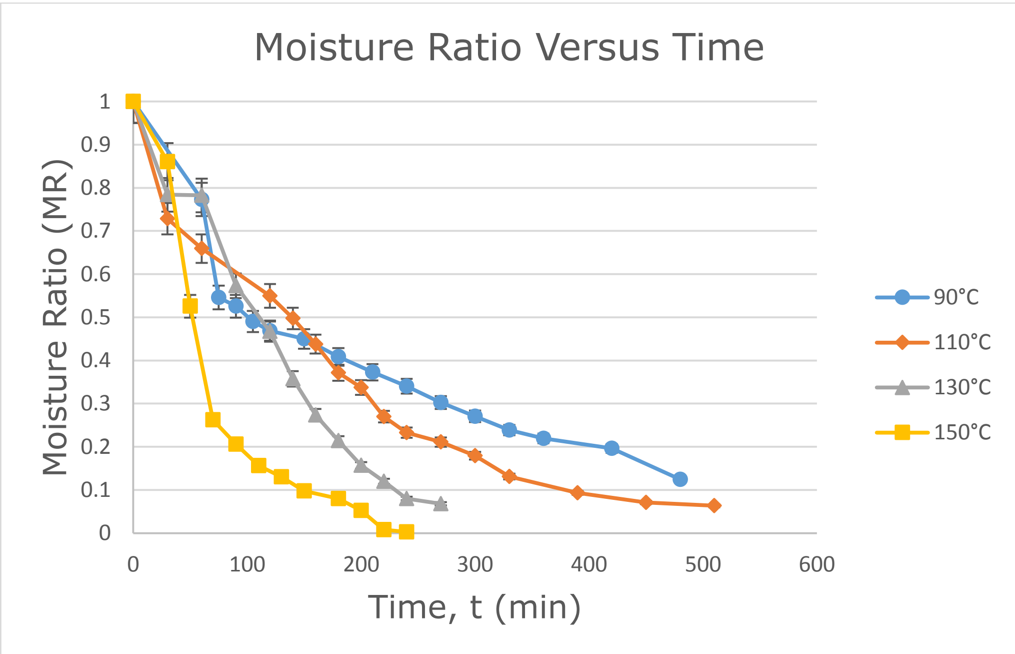


Figure 4.6: The variation of moisture ratio in respect of time for rubber dried via intermittent duration of 15 min and intermittent ratio of 0.25

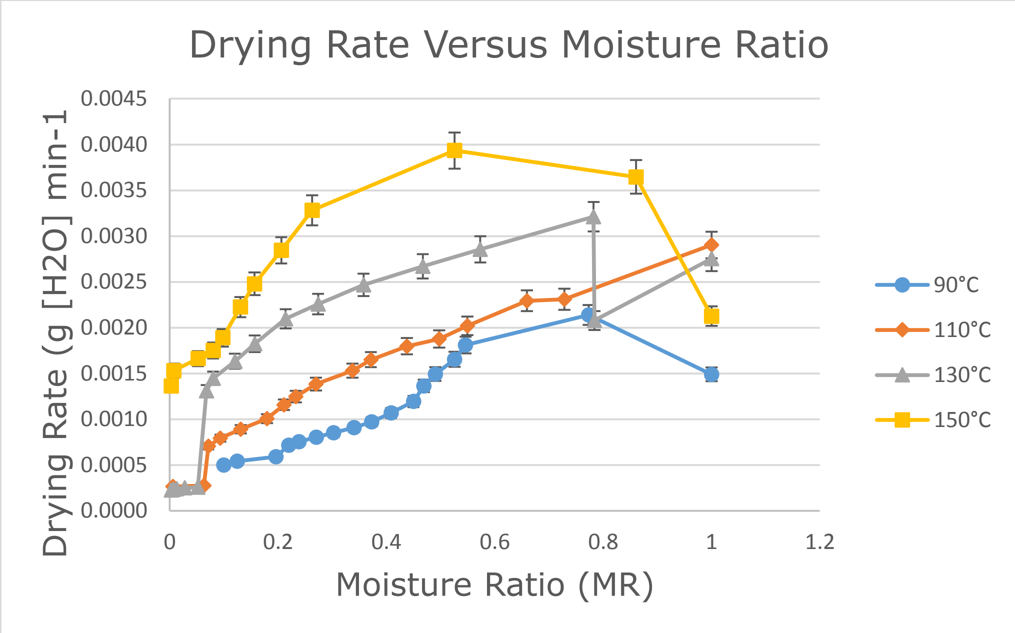


Figure 4.7: The variation of drying rate in respect of moisture ratio for rubber dried via intermittent duration of 15 min and intermittent ratio of 0.25

Figure 4.6 and 4.7 for varying hot air drying (HAD) temperature of the intermittent drying at ID15- IR025 was as shown graphically above. Figure 4.6 shows that a high temperature intermittent drying could reduce the moisture ratio rapidly. As the rubber exposure to high temperature heated dry air, the moisture that attained latent heat of evaporation will evaporated quickly. Thus, the high temperature drying at 150°C can dry the rubber at least 4 times faster than at 90°C.

Figure 4.7 showed that the drying rates were higher under intermittence of heating temperature in comparison with results obtained under conventional operation with constant heating temperature. There was a significant higher drying rate and shorter drying time when rubber was dried at 150°C for the intermittent drying carried out at ID15- IR025. The higher the drying temperature, the higher the drying rate. Comparison of the four drying temperatures (90, 110, 130 and 150°C) used in the second stage of vacuum convective drying showed that high temperature drying was preferable for rubber drying, based on the drying rate value obtained. The comparison of various intermittent duration, ID (15, 30, and 45 min) and intermittent ratio, IR (0.25, 0.50 and 0.75) showed that a short and frequent intermittency strategy is most suitable for rubber drying to overcome quality deterioration, which is ID15-IR075.

4.2.2 Rubber quality of two-stage VCD intermittent drying

Others than drying kinetics, the effects of tempering during the vacuum convective drying (VCD) on colour measurement (L*a*b*), plasticity retention index (PRI) and texture can be seen from Table 4.4, Figure 4.8 and 4.9, respectively. The colour differences and textural changes were confirmed by measuring the colour change using a colorimeter and a texture analyser.

Table 4.4 Effect of Varying Intermittence on Vacuum Convective Drying (VCD) to Visual Attributes of Dried Crumb Rubber

Intermittent Duration (ID)	Intermittent Ratio (IR)	L*	a*	b*	ΔE	PRI
t = 15 min	α= 0.25	9.43 ± 3.44 [a]	2.70 ± 1.08 [b]	5.23 ± 2.64 [b]	10.86 ± 2.26 [e]	>70 [g]
	α= 0.50	14.70 ± 7.61 [a]	4.20 ± 1.68 [b]	7.03 ± 4.73 [b]	10.32 ± 3.86 [e]	>70 [g]
	α= 0.75	13.33 ± 0.90 [a]	3.73 ± 1.65 [b]	6.97 ± 2.36 [b]	7.45 ± 0.48 [f]	>70 [g]
t = 30 min	α= 0.25	12.05 ± 2.10 [a]	3.27 ± 0.61 [b]	5.42 ± 0.26 [b]	8.00 ± 2.04 [e]	>70 [g]
	α= 0.50	10.57 ± 5.81 [a]	2.83 ± 3.11 [b]	5.23 ± 1.90 [b]	9.44 ± 2.85 [e]	>70 [g]
	α= 0.75	15.03 ± 7.17 [a]	1.23 ± 1.37 [b]	3.58 ± 1.67 [b]	8.67 ± 2.19 [e]	>70 [g]
t = 45 min	α= 0.25	11.00 ± 3.30 [a]	3.18 ± 4.55 [b]	6.70 ± 0.40 [b]	10.03 ± 0.71 [e]	>70 [g]
	α= 0.50	13.33 ± 2.71 [a]	2.63 ± 2.92 [b]	6.40 ± 2.15 [b]	9.53 ± 2.57 [e]	>70 [g]
	α= 0.75	11.70 ± 2.55 [a]	0.87 ± 2.78 [b]	4.25 ± 3.18 [b]	9.83 ± 3.55 [e]	>70 [g]
Continuous	α= 1.00	27.07 ± 0.60 [a]	6.33 ± 1.46 [c]	13.67± 1.76 [d]	12.75 ± 1.65 [e]	>70 [g]

Note: Means in the same column that do not share subscripts differ at $p < 0.05$ probability level.

Table 4.4 shows the colour parameters results of brightness, L*, redness, a* and yellowness, b* for intermittently dried rubber had a lower value than the constant dried rubber. The industrial standard rubber colour, which is golden yellow or golden light brown colours, preferred higher L* and b* coupled with lower a* and ΔE. The statistical analysis showed that only a* and b* parameters were affected by the intermittent ratio and intermittent duration. Out of all the combinations, only intermittent drying of 15 min on 5 min off, ID15- IR075 was statistically significant compared to continuous drying results.

In this study, the L* value was amounting to 13.33 ± 0.90 for ID15- IR075 which was 50.76% darker compared to continuous drying (IR1). The low colour brightness can be due to the protein denaturation of rubber or enzymatic browning reaction, as reported by Krokida (1999). From the results, the b* value for intermittently dried samples were 16.04% - 29.25% lower than continuous drying, which indicated that the samples had a lighter brown colour. A less severe browning was found for all intermittently dried samples, with no incorporation of wet rubber particles in the dried rubber. This showed that intermittent drying helped to improve the colour quality of rubber, referring to the results with a closer colour characteristic to the standard rubber sample. Any total colour change, ΔE value below 10 was acceptable, based on industrial practices.

Rubber quality tends to deteriorate upon continuous drying for a prolonged drying period. For this, an optimal intermittent drying strategy was determined based on the rubber quality. The rubber samples dried by intermittent drying ID15- IR075 looked more alike to standard rubber colour compared to continuous drying. In term of PRI values, there was no significant effect of intermittency strategies to the rubber plasticity index. A statistical significant lower total colour change value for crumb rubber subjected to the two-stage intermittent vacuum convective drying (VCD) ID15- IR075 indicated that this intermittent strategy is a better drying scheme compared to two-stage continuous VCD drying. Thus, this intermittent strategy could be adopted in subsequent two-stage intermittent vacuum convective drying.

Statistical analysis (analysis of variance, ANOVA) with the use of response optimizer showed that the change of intermittent ratio and duration had statistically significant influence on only one parameter, a* values. The regression equation 3.1 found for ΔE in relation to the intermittent duration (ID) and intermittent ratio (IR) is as below:

$$\Delta E = 9.348 + 0.196\ (ID15) - 0.644\ (ID30) + 0.449\ (ID45) + 0.282\ (IR025) + 0.416\ (IR050) - 0.698\ (IR075) \quad (3.1)$$

Figure 4.8 shows the optimization plot for the c change due to both the intermittent duration (ID) and intermittent ratio (IR).

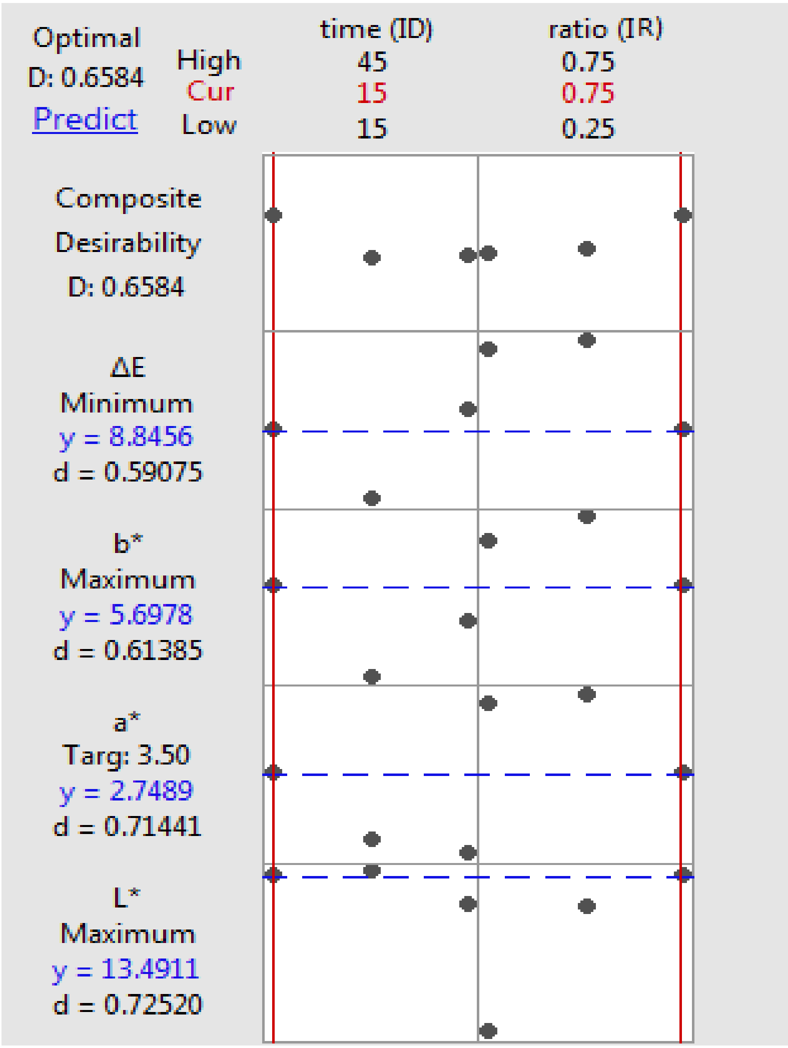


Figure 4.8: The optimization plot for total colour change, ΔE, lightness, L*, redness, a* and yellowness, b* values

Based on the results, the process conditions were optimized using response optimizer tool in Minitab 18. The intermittent duration (ID15) and intermittent ratio (IR075), ID15- IR075 were obtained as optimum condition leading to the lowest total colour change values. In contrast, when the intermittency duration (ID) was long, e.g. 45 min with low intermittent ratio, e.g. IR025, it took a relatively longer duration to reach the desired colour change and sufficient dryness. The L* value was amounting to 11.00 ± 3.30 for the ID45-IR075, which was 59.36% darker compared to continuous drying (IR1). As the temperature was fixed to study the intermittency strategies, only a minor colour change for sample undergone intermittent drying 45 min on, 135 min off was observed. Considering the time taken to dry the samples were too long, the long intermittency duration would not be a good choice for rubber drying. Hence, the chosen intermittency strategy based on the optimization plot of total colour change, confirmed to be ID15- IR075.

On top of the effect on colour changes, the effect of two-stage intermittent vacuum convective drying (VCD) on texture changes (hardness and stickiness) is as shown in Figure 4.9. All the quality results were considered for the determination of intermittency drying strategy.

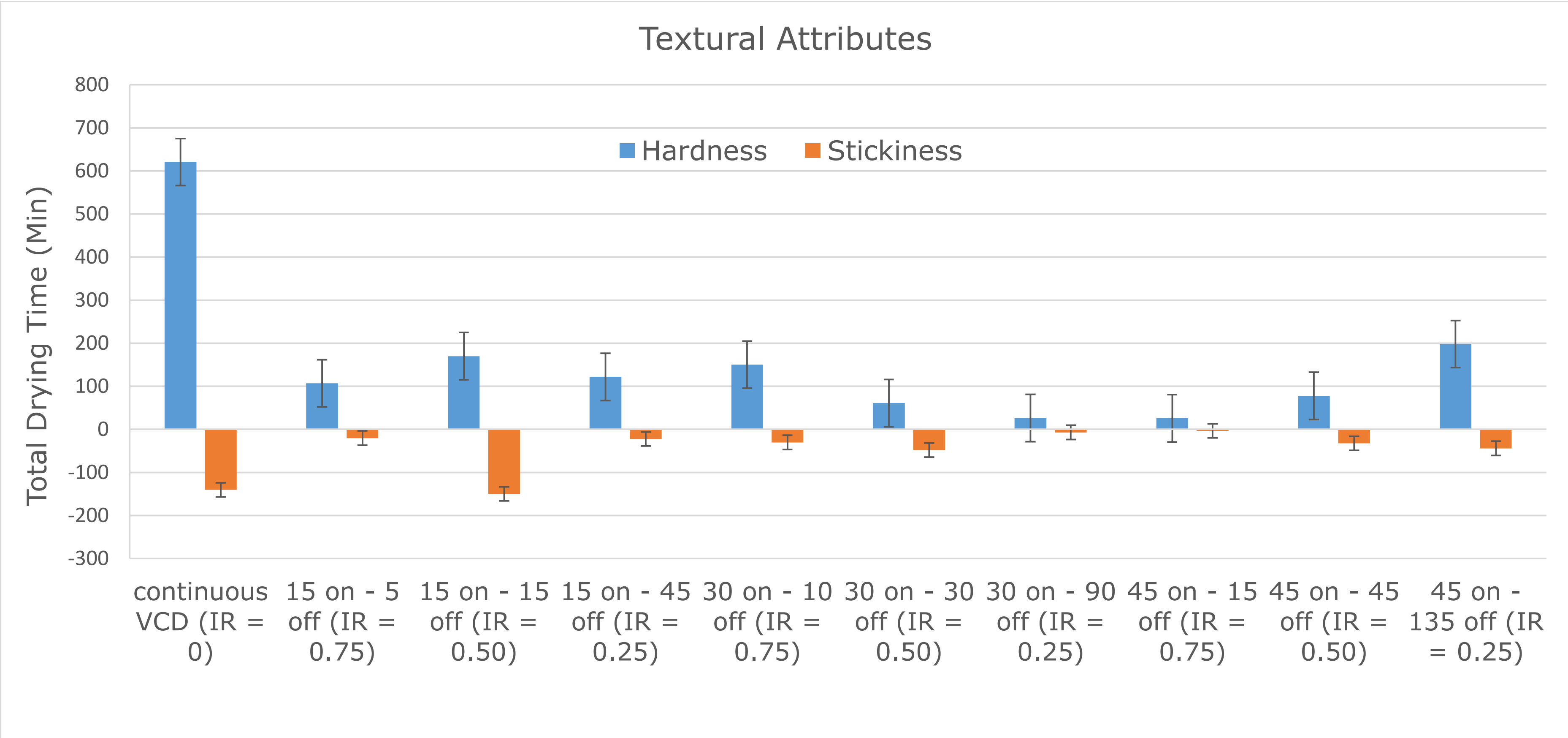


Figure 4.9: The textural attributes for comparison of continuous and intermittent drying scheme

The results in Figure 4.9 showed that intermittent ratio IR=0.25 (30 min on, 90min off) and IR=0.75 (45 min on, 15 min off) had a high degree of softness, 26.29 g and 25.82 g, respectively, while IR=0.50 (15 min on, 15 min off) had a similar softness with continuous VCD. The stickiness results of IR=0.50 (15 min on, 15 min off) was the highest, -149.73 g too. This results indicated that the rubber dried with IR=0.50 (ID = 15 min) had the best textural attributes. However, based on statistical analysis (ANOVA), there was no significant differences on the textural attribute for all intermittent drying strategies ($p>0.05$).The regression equation found for hardness and stickiness in relation to the intermittent duration (ID) and intermittent ratio (IR) is as below:

$$\text{Hardness} = 104.2 + 28.7\,(\text{ID15}) - 25\,(\text{ID30}) - 3.7\,(\text{ID45}) + 6.2\,(\text{IR025}) - 1.3\,(\text{IR050}) - 4.9\,(\text{IR075}) \quad (3.2)$$

$$\text{Stickiness} = -39.8 - 24.4\,(\text{ID15}) + 11.3\,(\text{ID30}) + 13\,(\text{ID45}) + 16\,(\text{IR025}) - 37.1\,(\text{IR050}) + 21.1\,(\text{IR075}) \quad (3.3)$$

Figure 4.10 shows the optimization plot for the texture changes due to both the intermittent duration (ID) and intermittent ratio (IR).

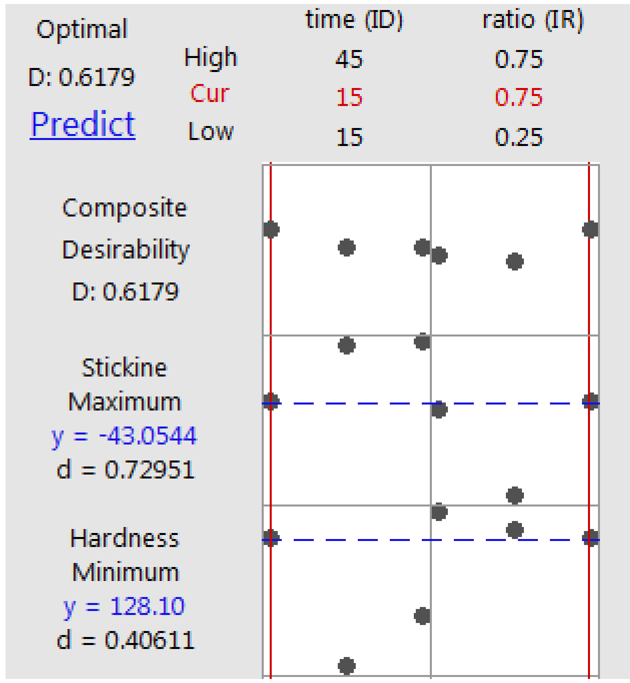


Figure 4.10: The optimization plot for texture changes

From the optimization plot, the intermittent duration (ID15) at 15 min and intermittent ratio (IR075) were also selected as the most optimum intermittent drying parameters to obtain maximum stickiness and minimum hardness for better rubber quality. The hardness and stickiness value were 41.7% and 47.3% lower than standard rubber. The change in sample colour and textural attributes, which was highly dependent on moisture content and heating temperature, was therefore expected to improve by the intermittent strategy of ID15- IR075. The effective diffusivities, Deff for this intermittent drying strategy was 38.149 x 10-9 /m^2 s^{-1}.

4.3 Two-stage Vacuum Convective Drying (VCD) results

A series of two - stage vacuum convective drying (VCD) experimental works were carried out to determine the effect of continuous drying and intermittent drying on the drying kinetics and rubber quality. A total of twenty - four experiments were carried out on the three drying parameters (vacuum duration, heating temperature and sample diameter) for the selected intermittent drying VCD. The effect of varying parameters to the moisture content, moisture diffusivities and colour changes of the sample was determined. Rubber samples with four different size (10 - 50mm) were dried at three vacuum duration (30, 60 and 120min) and four heating temperatures (90 - 150 ºC) during the second stage of intermittent VCD. All intermittent drying processes were performed with a fixed intermittency ratio (IR075) and intermittency duration (t = 15 min), ID15- IR075.

For each drying experiment, rubber samples with individual weight of 30g (diameter = 50mm), 10g (diameter = 20mm), 2 - 4 g (diameter = 15mm) and 1- 2g (diameter = 10mm) were cut into pieces and spread in a single layer to a total weight of 30g on an perforated aluminium tray, before placing into the dryer. The initial colour, texture and weight of the rubber samples were measured at the beginning and at the end of all experiments. Sample was withdrawn from the drying chamber for weighing in an increasing interval (from five-minute intervals to thirty-minute interval, followed by hourly interval) until the sample reached the equilibrium moisture content. Each experiment was repeated thrice and dried until their moisture level fall below 0.05 gH_2O gDM^{-1}. The effect of heating temperature with varying sample diameter on the drying kinetic of the rubber, dried with both two - stage continuous and intermittent vacuum convective drying (VCD) method were discussed in subsequent section. The reporting of VCD method was important as the preliminary studies showed promising effective diffusivities results and higher drying rate.

4.3.1 Drying time

A compilation on the total drying time taken for the rubber samples to reach below the final moisture content of 5% (d.b.) is shown in Table 4.5 and 4.6 for varying drying conditions, including vacuum duration, drying temperatures and sample diameter for the two-stage vacuum convective drying (VCD) in both continuous and intermittent basis.

Table 4.5: Drying time and final moisture content values for two-stage vacuum convective drying of rubber, by continuous drying

Sample Diameter/ Heating Temperature		Continuous Drying: Vacuum Duration (120 Min) Final Moisture Content (d.b.)	Vacuum Duration (120 Min) Total Drying Time (Min)	Vacuum Duration (60 Min) Final Moisture Content (d.b.)	Vacuum Duration (60 Min) Total Drying Time (Min)	Vacuum Duration (30 Min) Final Moisture Content (d.b.)	Vacuum Duration (30 Min) Total Drying Time (Min)
10mm	90°C	0.04495 [a]	480 [c]	0.04495 [a]	330 [c]	0.06600 [a]	450 [c]
	110°C	0.04772 [a]	300 [c]	0.03502 [a]	150 [d]	0.04745 [a]	240 [c]
	130°C	0.04132 [a]	150 [d]	0.01508 [b]	120 [d]	0.03270 [a]	90 [e]
	150°C	0.02117 [b]	150 [d]	0.02046 [b]	120 [d]	0.04746 [a]	60 [e]
15mm	90°C	0.04532 [a]	420 [c]	0.04237 [a]	450 [c]	0.03800 [a]	450 [c]
	110°C	0.03942 [a]	300 [c]	0.03942 [a]	240 [c]	0.04611 [a]	180 [d]
	130°C	0.04385 [a]	150 [d]	0.02765 [b]	120 [d]	0.05534 [a]	90 [e]
	150°C	0.03708 [a]	150 [d]	0.02776 [b]	90 [d]	0.02350 [b]	60 [e]
20mm	90°C	0.04206 [a]	480 [c]	0.04765 [a]	390 [c]	0.04859 [a]	330 [c]
	110°C	0.05345 [a]	420 [c]	0.04067 [a]	240 [c]	0.04791 [a]	240 [c]
	130°C	0.05028 [a]	180 [d]	0.04517 [a]	150 [d]	0.02817 [b]	120 [d]
	150°C	0.04043 [a]	150 [d]	0.04245 [a]	90 [d]	0.04519 [a]	60 [e]
50mm	90°C	0.05100 [a]	540 [c]	0.04229 [a]	480 [c]	0.04286 [a]	420 [c]
	110°C	0.04218 [a]	300 [c]	0.03612 [a]	300 [c]	0.04485 [a]	300 [c]
	130°C	0.04111 [a]	180 [d]	0.03408 [a]	180 [d]	0.04488 [a]	150 [d]
	150°C	0.03768 [a]	120 [d]	0.03373 [a]	120 [d]	0.05476 [a]	90 [e]

Note: Means in the same column that do not share subscripts differ at $p < 0.05$ probability level.

Table 4.6: Drying time and final moisture content values for two-stage vacuum convective drying of rubber, by intermittent drying

		Intermittent Drying					
		Vacuum Duration (120 Min)		Vacuum Duration (60 Min)		Vacuum Duration (30 Min)	
Sample Diameter/	Heating Temperature	Final Moisture Content (d.b.)	Total Drying Time (Min)	Final Moisture Content (d.b.)	Total Drying Time (Min)	Final Moisture Content (d.b.)	Total Drying Time (Min)
10mm	90°C	0.04779[a]	270[b]	0.05329[a]	270[b]	0.04964[a]	390[b]
	110°C	0.06309[a]	270[b]	0.05597[a]	270[b]	0.05756[a]	180[c]
	130°C	0.07077[a]	240[b]	0.05793[a]	100[d]	0.05387[a]	90[d]
	150°C	0.07958[a]	120[c]	0.04398[a]	100[d]	0.04213[a]	50[d]
15mm	90°C	0.05117[a]	390[b]	0.05916[a]	330[b]	0.05020[a]	450[b]
	110°C	0.04687[a]	270[b]	0.05417[a]	240[b]	0.05387[a]	180[c]
	130°C	0.05961[a]	160[c]	0.04218[a]	120[c]	0.04500[a]	90[d]
	150°C	0.03016[a]	140[c]	0.04463[a]	100[d]	0.06520[a]	50[d]
20mm	90°C	0.05445[a]	330[b]	0.05464[a]	390[b]	0.04745[a]	390[b]
	110°C	0.05102[a]	330[b]	0.05777[a]	180[c]	0.05240[a]	180[c]
	130°C	0.05842[a]	180[c]	0.05072[a]	180[c]	0.05158[a]	90[d]
	150°C	0.03534[a]	160[c]	0.05044[a]	100[d]	0.03428[a]	70[d]
50mm	90°C	0.05132[a]	540[b]	0.05095[a]	420[b]	0.05542[a]	360[b]
	110°C	0.04218[a]	300[b]	0.05878[a]	240[b]	0.04485[a]	300[b]
	130°C	0.04111[a]	210[b]	0.05929[a]	150[c]	0.04488[a]	150[c]
	150°C	0.03768[a]	120[c]	0.06379[a]	90[d]	0.05476[a]	90[d]

Note: Means in the same column that do not share subscripts differ at $p < 0.05$ probability level.

Drying time is a property dependent on heating temperature, hence, a higher increment in heating temperature resulting in shorter drying time. Rahman (1985) reported that a 33.3% increase of heating temperature to 80ºC and a 78.6% reduction of relative humidity (RH) to 15%RH reduced the rubber drying time by a factor of 2.67 and 2.7, respectively. For continuous drying of 10mm rubber samples at 90, 110, 130 and 150°C, the shortest total drying time found was 330, 150, 90 and 60, respectively while intermittent drying of 10mm rubber samples at 90, 110, 130 and 150°C, the shortest total drying time found was 270, 180, 90 and 50 min, respectively.

From Table 4.5 and 4.6, the highest drying time recorded is 540 min for the drying of 50mm rubber via two-stage VCD at V120-HAD90, in both continuous and intermittent basis; whereas, the lowest drying time recorded was 50 min for the drying of 10mm and 15mm rubber via two-stage intermittent VCD with 30 min vacuum pre-drying and hot air drying temperature at 150°C (V30-HAD150). Drying with V30-HAD150 compared to V120-HAD90 resulted in a 90.74% reduction in total drying time. As the total drying time of intermittent drying included tempering period, the effective drying time for two-stage intermittent V30-HAD150 of 10mm and 15mm were only 45 min. This showed that VCD carried out intermittently at high temperature was more efficient. The drying of rough rice also found that higher intermittent temperature would allow faster moisture removal and time reduction up to 38% (Aquerreta, 2006). Correia (2015) reported on the improvement in drying time by using higher heating temperature and lower vacuum pressure as well. In short, there was no significant effect of varying vacuum duration (30, 60 and 120 min) to drying time reduction while the most significant differences was through varying heating temperature (90, 110, 130 and 150°C).

From the results, the effect of varying vacuum duration (30, 60 and 120 min) to drying time was highly dependent on the heating temperature of hot air drying (HAD) during the second stage of vacuum convective drying (VCD). As the incremental temperature step change during the transition from vacuum drying (VD) to HAD become higher, the effect of vacuum duration to drying time may change. For example, the drying of 10mm crumb rubber via intermittent VCD with 30 min vacuum pre-drying and hot air drying temperature at 90°C (V30-HAD90), took 44.44% and 44.44% longer drying time compared to vacuum pre-drying for 60 min (V60-HAD90) and vacuum pre-drying for 120 min (V120-HAD90), respectively; but the drying of 10mm crumb rubber via intermittent VCD at with 30 min vacuum pre-drying and hot air drying temperature at 150°C (V30-HAD150), required 50% and 58.33% shorter drying time compared to vacuum pre-drying for 60 min (V60-HAD150) and vacuum pre-drying for 120 min (V120-HAD150), respectively. This inverse trend found when the heating temperature for HAD was step-up for 20, 40 and 60°C to 110, 130 and 150°C, respectively. This was because vacuum drying (VD) supplies the heat by conduction. The slow heat transfer rate to the materials lead to a longer drying time (Duan, 2012), especially when the hot air drying (HAD) stage was carried out at high drying temperature. The temperature increment boosted the drying rate and hence reduced the drying time. Similar trend was also found for intermittent VCD of 15 and 20mm, but not seen for 50mm sample. This showed that the effect of vacuum drying was more significant for thin layer rubber dried via intermittent mode. However, there was not much different in term of final moisture content, regardless of the vacuum duration.

Both continuous drying and intermittent drying showed a similar decreasing trend in drying time when the heating temperature was increased. However, the drying time reduction percentage for incremental temperature step change was different for continuous VCD drying and intermittent VCD drying.

For example, the drying of 50mm crumb rubber via continuous VCD with vacuum pre-drying for 30 min and hot air drying temperature at 150°C (V30-HAD150) had 78.57%, 70% and 40% shorter drying time than 90°C (V30-HAD90), 110°C (V30-HAD110) and 130°C (V30-HAD130), respectively while the drying of 10mm crumb rubber via intermittent VCD at V30-HAD150 was 87.18%, 72.22% and 44.44% shorter drying time than V30-HAD90, V30-HAD110 and V30-HAD130, respectively. This showed that the increment in heating temperature for intermittent VCD had a higher effect to drying time. Similarly, the drying of 10, 15 and 20mm crumb rubber via continuous VCD at V30-HAD150 had 86.67%, 86.67% and 81.81% shorter drying time than V30-HAD90, respectively. A thinner rubber sample dried faster compared to a thicker sample, especially when the heating temperature was higher. Thus, reduction percentage of drying time was more significant for high temperature drying of thin samples compared to 50mm thick sample.

Another interesting observation in this experiment was the linear correlation of the critical drying time with the sample diameter. Table 4.5 and 4.6 showed that the rubber was dried 90.74% faster when it was in smaller sizing (10mm versus 50mm), lower vacuum duration (V30 versus V120) and higher hot air drying temperature (150°C versus 90°C). Drying temperature was the only key variable that affect the time required to reach the final moisture content of 5% (d.b.). The results were consistent with the findings reported in the literature where heating temperature are the most important factor affecting the drying characteristics of bio-product (Singh et al., 2014; Correia et al., 2015).

It was clear from Table 4.5 and 4.6 that the total drying time for two-stage intermittent VCD was always lower than continuous VCD while lower vacuum duration, higher drying temperature and low sample diameter always gave shorter drying time. All samples eventually reached a moisture content level which was lower than 0.005 gH_2O gDM^{-1} when exposed to the corresponding drying temperature for long periods of time. The final moisture values (d.b.) varies from 0.000493% to 0.05132%. Moisture content is a property dependent on heating temperature and relative humidity, hence, the value was expected to be lower with higher heating temperature and higher with higher relative humidity. The pulsating of heat input during intermittent drying caused a slight increment in the relative humidity. Thus, the moisture content of dried rubber at equilibrium for intermittent drying was slight higher than continuous drying.

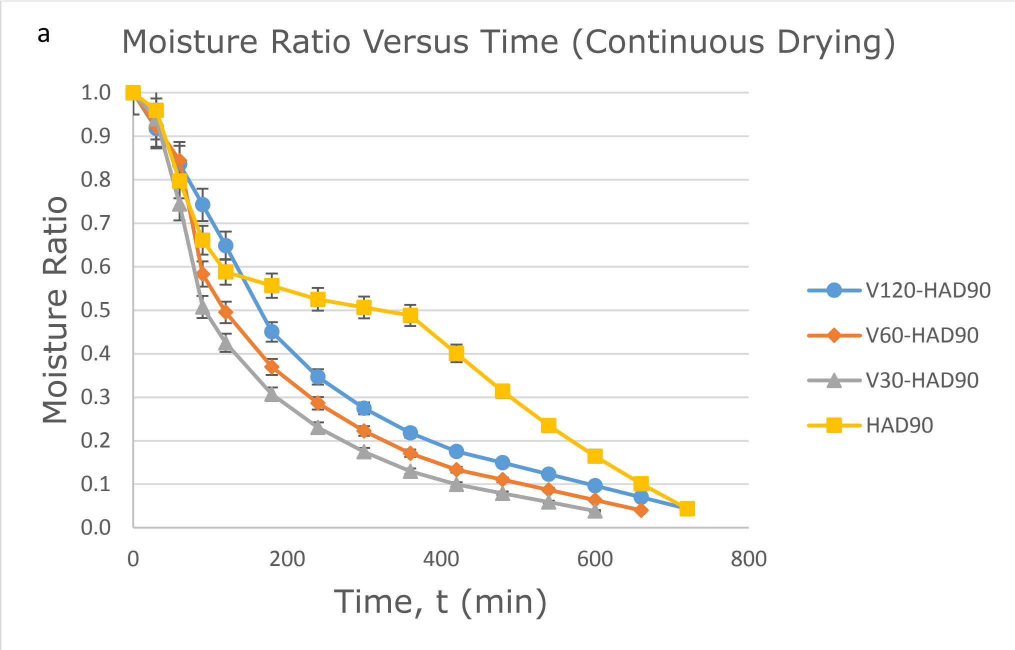
4.3.2 Drying characteristic curve of two-stage intermittent VCD drying

The moisture ratio was plotted against drying time for varying vacuum duration, heating temperatures and sample diameter. The effect of these variables on the drying characteristics of rubber was plotted in Figure 4.11a, b, 4.12a, b and 4.13a, b, respectively for two-stage vacuum convective drying (VCD) in continuous basis and intermittent basis.

4.3.2.1 Effect of vacuum duration

Drying curves (moisture ratio versus drying time) for 50mm rubber dried under various vacuum duration (30, 60 and 120 min) in vacuum pre-drying stage during the two-stage vacuum convective drying (VCD) were plotted in Figure 4.11 a, b. Both graph show that the drying curves for rubber dried with two-stage vacuum convective drying (VCD) typically given by an exponential function. There was a higher reduction in moisture content for two-stage VCD compared to the rubber dried with hot air drying at 90°C (HAD90). HAD90 took 600 min and 460 min to lose at least 80% of its moisture content, for continuous drying and intermittent drying, respectively. Whereas, two-stage VCD with vacuum pre-drying for 30 min coupled with hot air drying at 90°C (V30-HAD90) took a total drying time of 300 min and 330 min to remove at least 80%

moisture from rubber, for continuous drying and intermittent drying, respectively.



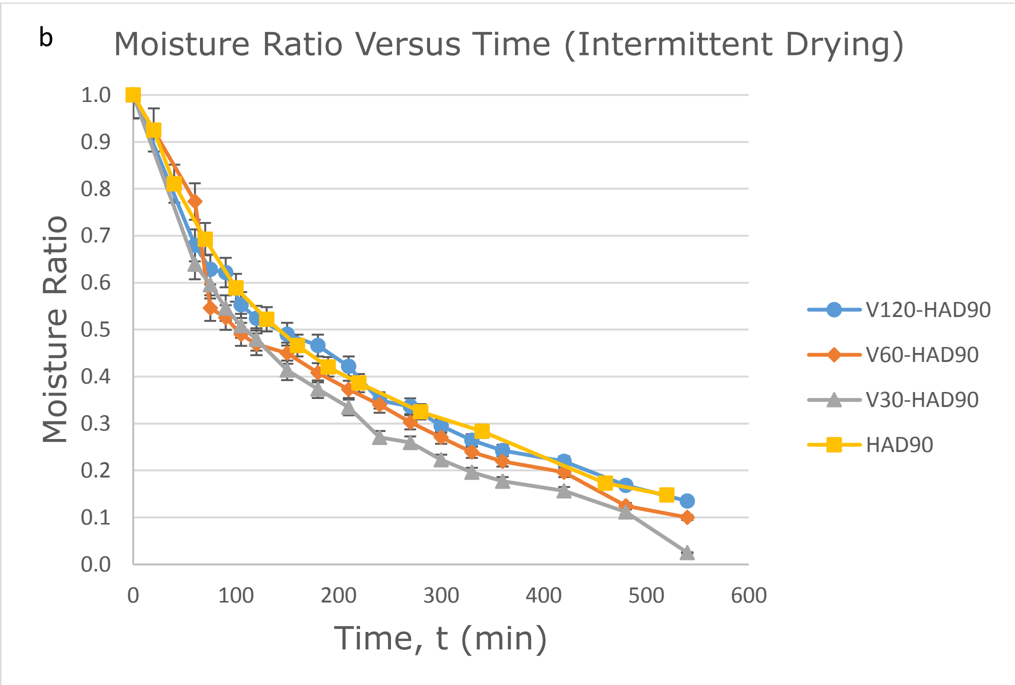


Figure 4.11a, b: Drying curve of 50mm thick rubber subjected to two-stage vacuum convective drying (VCD) at various vacuum pre-drying duration, followed by hot air drying at 90˚C

The use of vacuum pre-drying seems was able to dry the materials at low oxygen condition and reduce the total drying time. The moisture evaporation for all set of experiments seem to slow down after the moisture ratio fall below 0.2. As the dominant drying mechanism in rubber drying is diffusion, the time taken for moisture diffusion increased when the moisture content reduced. The study of internal moisture movement in rubber also showed that the diffusion was largely affected by moisture concentration (Gale, 1962). When a drying pressure was reduced, drying time could be reduced as the pressure gradient drives a better moisture diffusion (Correia, 2015).

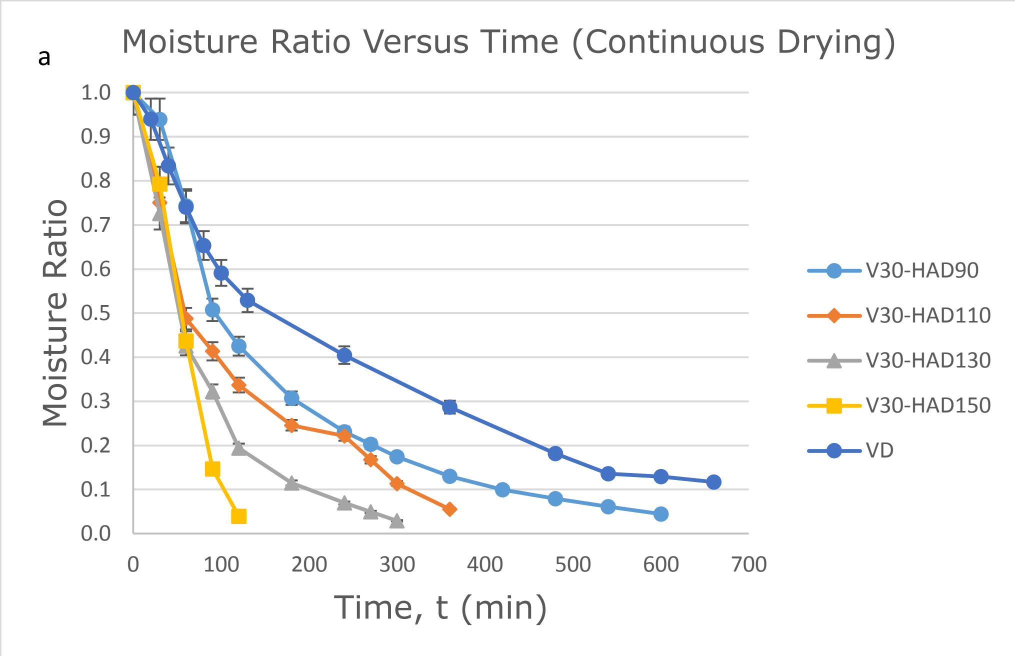
From the results, the effective drying time for intermittent HAD90 and intermittent V30-HAD90 were 390 min and 255 min, respectively. Compared to continuous V30-HAD90, continuous HAD90 and intermittent HAD90 were 100% and 30% slower, but intermittent V30-HAD90 was 75% faster. The moisture evaporation for rubber dried via continuous HAD90 slowed down when the moisture ratio had fallen below 0.66 after 90 min of drying. This was due to the collapse of rubber structure when rubber was exposed to hot, dry air for a prolonged period (Rahman, 1985). This phenomenon was not seen for rubber dried via intermittent HAD90 or any of the two-stage VCD, which showed that the tempering period and vacuum pre-drying allowed sufficient moisture diffusion from the internal to the surface. This concluded that the use of intermittent drying or two-stage vacuum convective drying improved the moisture reduction and drying time. Hence, energy might be saved through the intermittent VCD drying (Thomkapanich et al., 2007; Pan et al., 1999; Lijuan et al., 2005).

The continuous drying of two-stage vacuum convective drying (VCD) in varying vacuum duration, 30 min (V30-HAD90), 60 min (V60-HAD90) and 120 min (V120-HAD90) required a drying time of 300, 360 and 420 min, respectively. For intermittent drying, V30-HAD90, V60-HAD90 and V120-HAD90 showed that a total drying time of 330, 420 and 480 min was required to dry rubber to 20% moisture content, which the effective drying time were 255, 330 and 390min, respectively. Based on the effective drying time, shorter time was required by using two-stage intermittent VCD. The time saved for each of the two-stage VCD, i.e., V30-HAD90, V60-HAD90 and V120-HAD90, was 15%, 16.67% and 14.29%, respectively. The vacuum pre-drying duration up to 60 min gave the maximum benefit for reduction of drying time. However, the final moisture content values for varying vacuum duration (30, 60 and 120 min) were not significantly different from each other, for both continuous and intermittent drying. Nevertheless, two-stage vacuum convective drying is a suitable rubber drying method as the continuous hot air drying lead to undesirable colour and textural changes.

4.3.2.2 Effect of heating temperature

Drying curves (moisture ratio versus drying time) for 50mm rubber subjected to two-stage vacuum convective drying (VCD) under different heating temperatures (90, 110, 130 and 150°C) were plotted and shown in Figure 4.12 a, b. From the chart, there was significant differences in the drying curves for rubber drying at various heating temperature. However, not all final moisture content values for rubber dried at higher temperature was lower. This might be due to the uncontrollable of relative humidity in the present study. The variability of relative humidity (0 – 5%) probably masked the influence of heating temperature.

The drying curves for rubber dried with two-stage VCD at all temperatures were given by an exponential function. It was apparent that all experiments showed a decreasing moisture ratio with time. Two-stage VCD with 30 min vacuum pre-drying duration and hot air drying temperature at 150°C (V30-HAD150), in continuous basis, required the least drying time, 120 min to reach the desired moisture content of 5% (d.b.), compared to hot air drying temperature at 90°C (V30-HAD90) that required 600 and 660 min of total drying time, in continuous and intermittent basis, respectively.



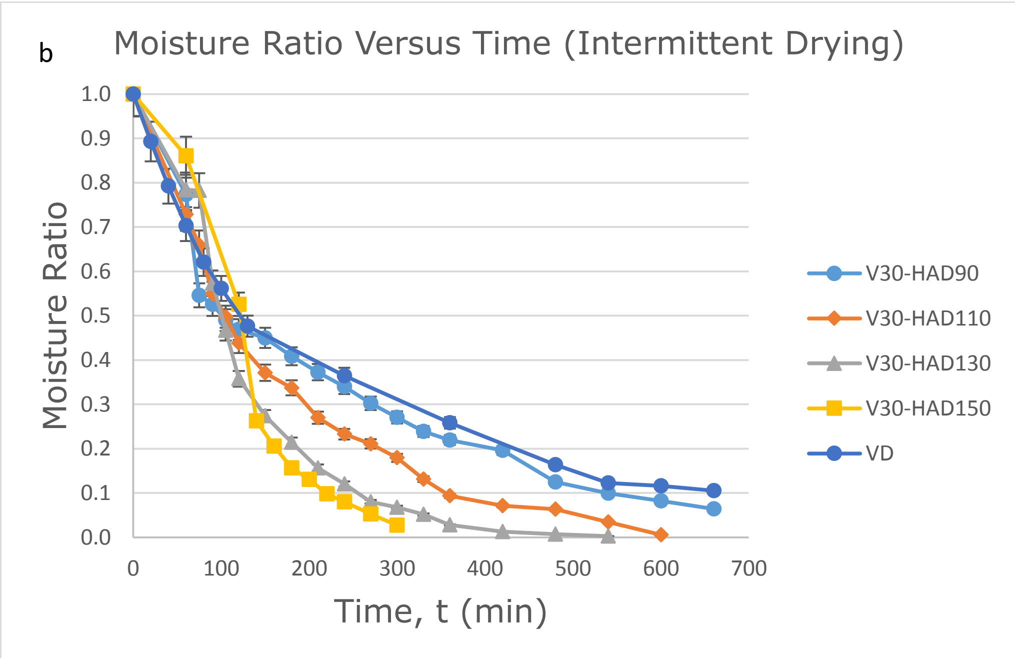


Figure 4.12a, b: Drying curve of 50mm thick rubber subjected to two-stage vacuum convective drying (VCD) at 30 min vacuum pre-drying, followed by hot air drying at different heating temperatures

Based on Fig. 4.12 a, b, both continuous V30-HAD150 and intermittent V30-HAD150 had the shortest total drying time, followed by drying temperature of hot air drying at 130°C (V30-HAD130), 110°C (V30-HAD110) and 90°C (V30-HAD90). The total drying time for V30-HAD150, V30-HAD130, V30-HAD110 and V30-HAD90 in continuous basis were 120, 270, 360 and 600 min, respectively to reach the desired moisture content. However, it was obvious from the chart that the final 20% of total moisture content in rubber took majority of the drying time. For example, V30-HAD150, V30-HAD130, V30-HAD110 and V30-HAD90 in continuous basis only required 90, 120, 270 and 300 min to remove 80% of the moisture, but an additional 33.33%, 125%, 33.33%, 100% of the drying time was required to further reduce from 20% residual moisture to 5%.

On the other hand, the total drying time for V30-HAD150, V30-HAD130, V30-HAD110 and V30-HAD90 in intermittent basis were 270, 300, 420 and 660 min, with an effective drying time of 210, 232.5, 322.5 and 502.5 min, respectively. Similarly, the final 20% of the rubber moisture for V30-HAD150, V30-HAD130, V30-HAD110 and V30-HAD90 in intermittent basis, took additional 50%, 42.86%, 60% and

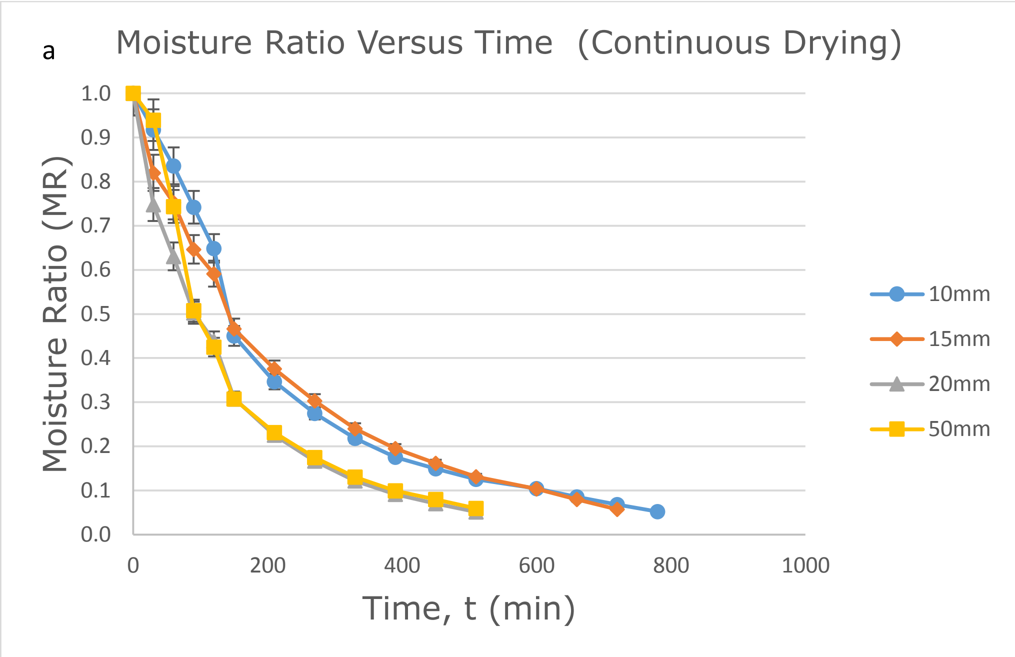
57.14% of the drying time to reach the desired moisture content of 5% (d.b.). Piddlesden (1937), who was the pioneer in rubber drying study, also stated that the moisture diffusion for the final 10% moisture of rubber took a long drying time with low drying rate. Thus, it was practical to consider whether the drying of rubber to moisture content of such extend was necessary for the industry.

When the sample experienced a step-up temperature profile of 90/150°C for two-stage vacuum convective drying (VCD), the increment in the temperature gradient of rubber surface and heated air promoted the moisture removal. The heated air supplied sufficient latent heat of evaporation to remove the moisture from rubber surface. As the intermittent drying began, the evaporation of moisture during the tempering period rely on the potential heat, which was the sensible heat accumulated when heat was applied on rubber. The sensible heat provided sufficient latent heat of evaporation to remove moisture from rubber during the tempering period, as shown by the smooth drying curve in Figure 4.12a, b. The highest reduction of the effective drying time for two-stage intermittent VCD, such as V30-HAD90 was 16.25% compared to continuous VCD. The improvement in effective drying time showed that possible energy saving could be achieve with the application of two-stage intermittent VCD process. Thus, intermittent V30-HAD150 with the shortest total drying time could be a good choice for rubber drying process.

4.3.2.3 Effect of sample diameter

Drying curves (moisture ratio versus drying time) for rubber samples of various diameter subjected to two-stage vacuum convective drying (VCD) with 30 min vacuum pre-drying and heating temperatures (90°C) were plotted and shown in Figure 4.13 a, b. Drying of various samples diameters (10, 15, 20 and 50mm) subjected to two-stage vacuum convective drying in both continuous and intermittent basis were given by an exponential drying curve.

Figure 4.13a shows a 10mm sample required 52.94% longer drying time compared to 50mm sample, to reach a moisture ratio below 0.05, when the sample was subjected to two-stage vacuum convective drying (VCD) in continuous basis. Similar findings were reported by other researchers who had done substantial studies on crumb rubber drying process (Berthomieu et al., 1997; Rahman, 1985). In Berthomieu's study, the drying time was reduced by 27% when the crumb rubber thickness was increased by 66.67% of its original thickness. This was due to the high porosity of the granulated rubber, which the larger surface area allows more heat transfer and hence, improved the moisture diffusion and drying rate (Rahman, 1985). The high rubber porosity in thick rubber samples helped to improve the moisture diffusion.



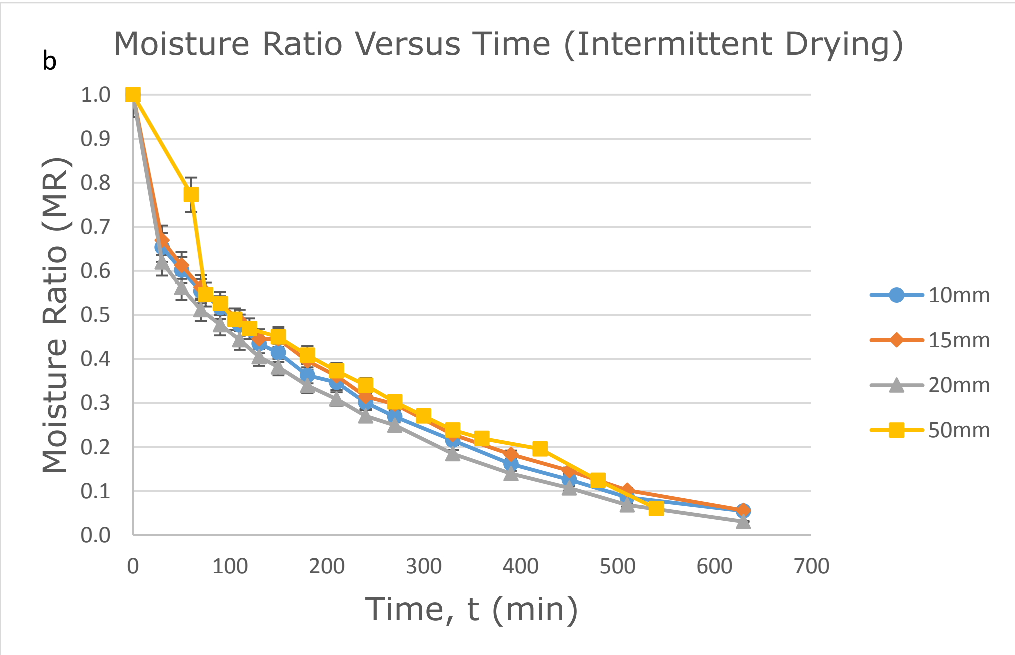


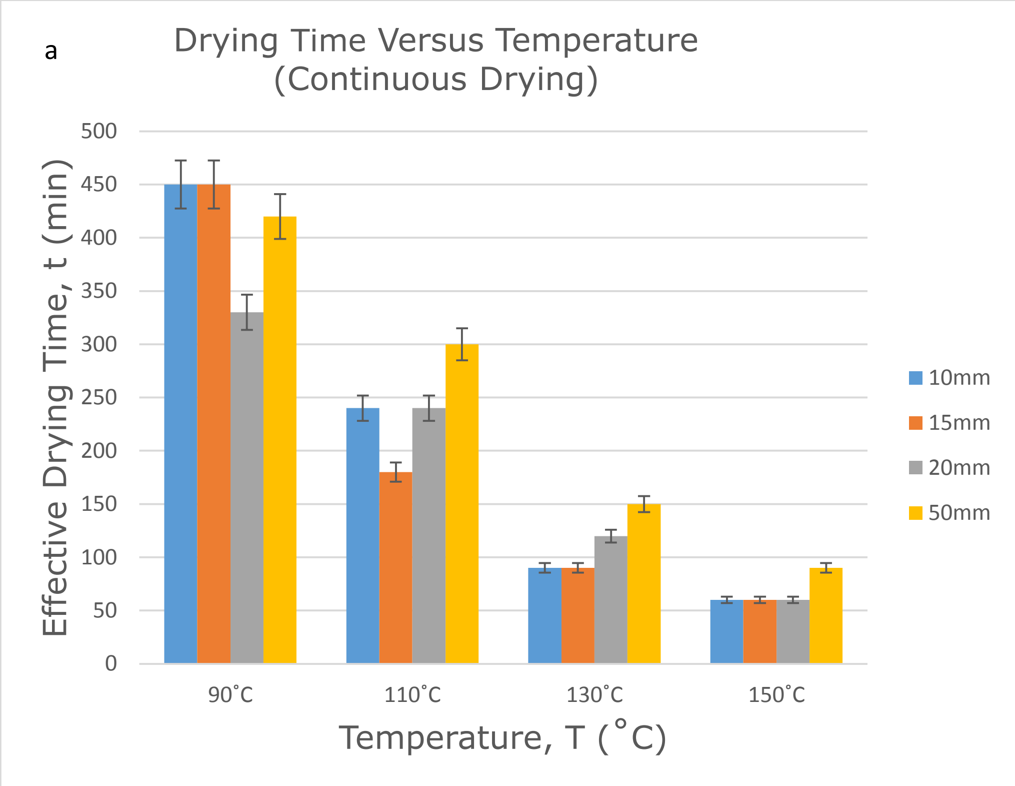
Figure 4.13a, b: Drying curve of rubber samples of various diameter dried with two-stage vacuum convective drying (VCD) at 30 min vacuum pre-drying, followed by hot air drying at 90˚C

From Figure 4.13a, b, the moisture content of rubber cut into various sample diameter (10, 15, 20 and 50mm) decreases continually with drying time. As seen in Figure 4.13a, there was a gap between the drying curves of thin sample (10 and 15mm) and thick sample (20 and 50mm). By employing intermittent drying, the drying curves in Figure 4.13b overlaps among each other. This showed that intermittent drying allows the moisture diffusion to the rubber surface consistently. When the sample was dried continuously, the moisture trapped in the thicker sample required more time to diffuse to the surface as the structure collapsed. With the help of intermittent drying strategy, the moisture removal from samples was reduced gradually and avoided the collapse of structure. Intermittent allow rewetting of rubber to have moisture accumulated on the rubber surface before resuming the drying process.

The varying of sample diameters does not affect final moisture content as much as relative humidity and drying temperature. The only exception found was during the drying of 50mm crumb rubber via two-stage vacuum convective drying (VCD) at intermittent mode. The higher sample diameter of 50mm gave a higher final moisture content values as the moisture need to travel through a larger cross-sectional area to reach the surface. Due to the low moisture gradient at the final drying period, the moisture diffusion might be too slow and hence, higher EMC values were obtained when the partial vapour pressure of the rubber and air become equal (Rizvi, 1986). In addition, Piddlesden (1937) stated that the drying rate of rubber became very slow to remove the remaining 10% of moisture. In the preliminary study discussed earlier, similar findings were found for the vacuum drying (VD) of rubber. The removal of the remaining 12.55% of moisture via VD process took an additional of 1440 min, which accounts 68.57% of total drying time. The time taken by hot air convective dryer to remove similar amount of moisture was 99.64% shorter than VD. Despite that, it was preferable to dry rubber to a final moisture content 0f 5% (d.b.) to prevent unnecessary heat stress and oxidative degradation to rubber. This final moisture content was also used by Tasara (2009) for the drying of same material.

For two-stage vacuum convective drying (VCD) in continuous basis, the 10mm rubber sample took additional 450 min (57.69% of the total drying time) to remove 16.56% of the residual moisture content while 50mm sample took 300 min (58.82% of total drying time) to remove the residual 22.5% of moisture. The results were in accordance with earlier findings, as more than 50% of the total drying time was spent on removing the last 20% of moisture in rubber. Two-stage intermittent VCD resemblance the same trend, which 10, 15, 20 and 50 mm of the rubber samples, each spent 47.62%, 42.85%, 41.18% and 22.22% of the total drying time to lose 20% of moisture. As the sample diameter increase, the moisture removal took lesser time. Therefore, it was safe to decide that larger sample with higher diameter had a higher drying rate and should be chosen for stepwise drying.

Figure 4.14a, b for both continuous drying and intermittent drying of two-stage vacuum convective drying (VCD) with vacuum pre-drying of 30 min shows the effect of sample diameter to effective drying temperature across heating temperature. Based on Figure 4.14a, b, it is very noteworthy that temperature dependency is more significant than the size dependency for both continuous drying and intermittent drying of the two-stage vacuum convective drying (VCD). An increase in the heating temperature, leads to an increase of the moisture evaporation rate. Drying time was typically shorter with increasing heating temperature of hot air drying (HAD) stage and sample with lower diameters.



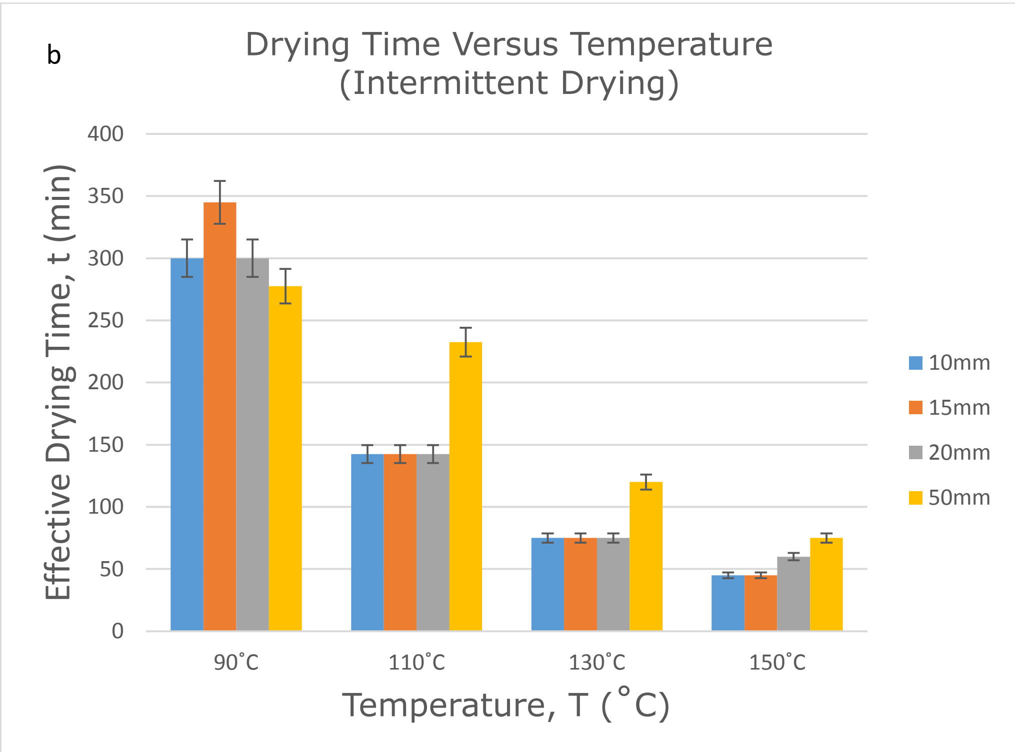


Figure 4.14a, b: Histogram of rubber samples of various diameter subjected to two-stage vacuum convective drying (VCD) at 30 min vacuum pre-drying, followed by hot air drying at four drying temperatures (90, 110, 130, 150˚C)

A comparison between the effective drying time shows that the intermittent drying process gave a reduction of 0 – 40.63% of drying time. For example, intermittent drying of 50mm rubber samples at 90, 110, 130 and 150°C decreased the drying time by 33.93%, 22.50%, 20% and 16.67%. There was a decreasing trend in reducing the drying time when intermittent drying was applied at high heating temperature. This was due to the higher driving force of moisture diffusivity, which lead to a lesser time taken for the rubber to dried in the continuous drying process. When the water vapour concentration was low in the drying atmosphere, especially for high heating temperature, it encourages the diffusion of moisture. Given that heat transfer was required for drying to occur, it was not surprised that temperature affected the drying results.

It was found that the final moisture content of 50mm samples subjected to two-stage vacuum convective drying (VCD) at 30 min vacuum pre-drying, followed by hot air drying (HAD) at 90, 110, 130 and 150°C, in continuous basis, were 0.043, 0.045, 0.045 and 0.055 $gH_2O\ gDM^{-1}$ while the intermittent drying gave final moisture content values of 0.055, 0.045, 0.045 and 0.055 $gH_2O\ gDM^{-1}$. There were no different in term of the final moisture content, even though the samples were dried at different drying temperature. Thus, the study on the effect of drying temperature or sample diameter to product quality is necessary to determine the suitability of heating temperature.

4.3.3 Drying rates

The time taken to reach EMC and final moisture content (< 5% d.b.) was defined, while the drying rate behaviour of crumb rubber under varying parameters was discussed. The drying rate (DR) was plotted versus moisture ratio (MR) in Figure 4.15a, b, Figure 4.16a, b and Figure 4.17a, b. for the two-stage vacuum convective drying (VCD). The effects of vacuum duration, heating temperature and sample diameter on the drying rate were investigated.

4.3.3.1 Effect of vacuum duration

Figure 4.15a, b shows the DR versus MR curves for two-stage vacuum convective drying (VCD) of rubber at vacuum duration of 30 min (V30), 60 min (V60) and 120 min (V120) coupled with hot air drying temperature of 150°C (HAD150), in continuous and intermittent basis.

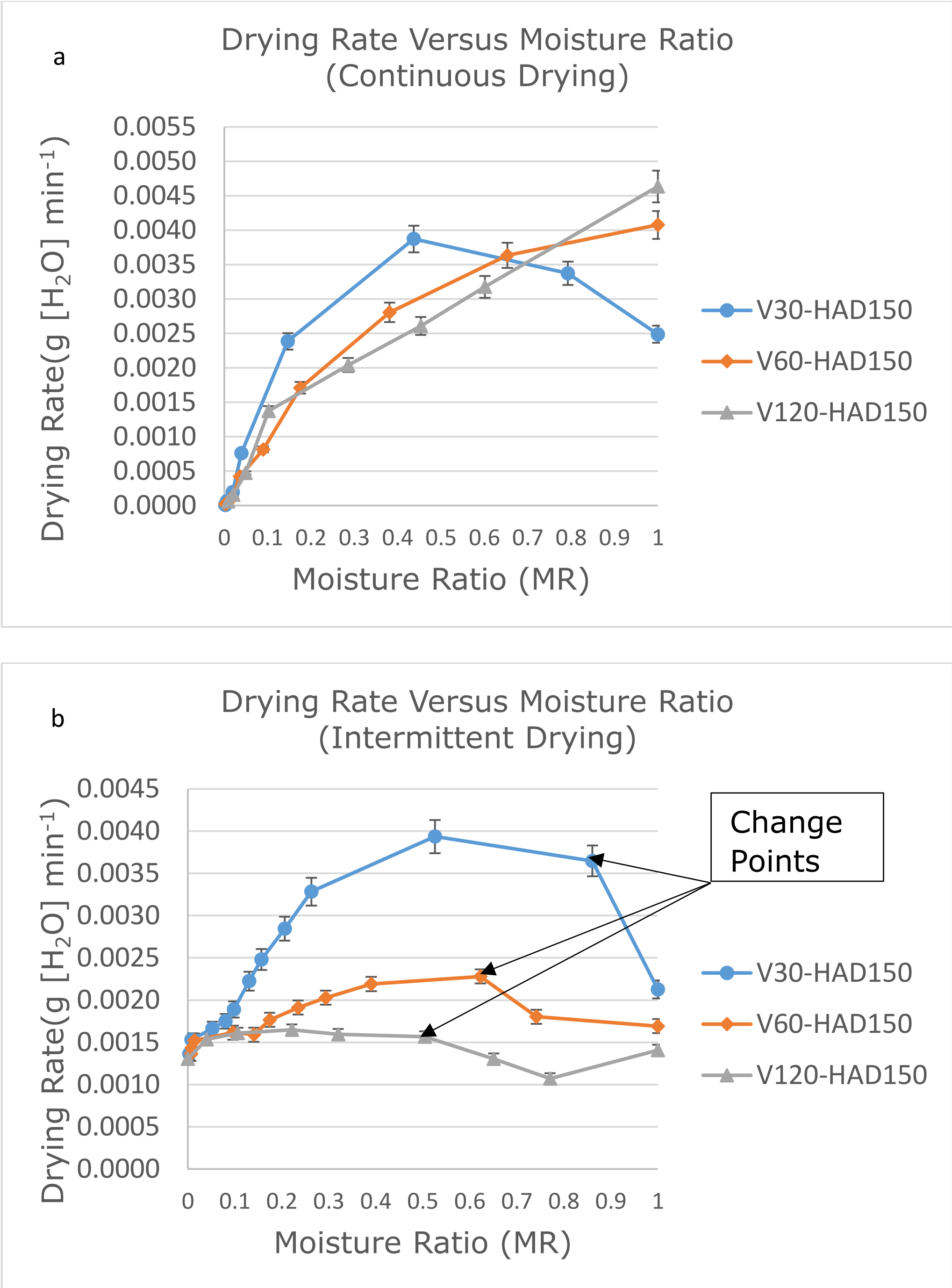


Figure 4.15a, b: Drying rate versus moisture ratio for 50mm rubber subjected to two-stage vacuum convective drying (VCD) with various vacuum duration (30, 60 and 120 min) at hot air drying temperature of 150°C (HAD150)

For the long pre-drying vacuum duration, V60 and V120 exhibits two different drying curves for continuous and intermittent drying. Continuous V60-HAD150 and V120-HAD150 had a steep falling rate line compared to V30-HAD150 while the increasing rate period for intermittent V60-HAD150 and V120-HAD150 were not as high as V30-HAD150. There was an existence of initial transient period, increasing rate period and falling rate period for V60-HAD150 and V120-HAD150 during intermittent drying period.

For both continuous and intermittent V30-HAD150, the drying curves remained steady at a drying rate above 0.0025 g H_2O min^{-1} after the initial transient period and until the moisture ratio was lowered than 0.2. Vacuum pre-drying of 30 min gave a higher drying rate values. In the first stage of VCD, the air and water vapour in drying products were expanded, which promoted the moisture diffusion. After 30 min vacuum pre-drying, the excess moisture on rubber skin was readily evaporated. Thus, when the samples were transferred to hot air drying (HAD), the drying rate increased immediately.

The drying curves for intermittent V60-HAD150 and V120-HAD150 shows a first falling rate period before an increasing rate period. This was due to the loss of moisture equilibrium on rubber surface at the first stage of VCD, after drying inside vacuum dryer for prolonged period. At the second stage of VCD, when the samples were intermittently dried at HAD150, with step-up temperature profile of 60°C, the drying rate climbed to the peak, then continued with second falling rate period. The increasing rate period for V30-HAD150, V60-HAD150 and V120-HAD150 began at the point of change of 30, 60 and 120 min, respectively.

Based on Figure 4.15a, both V60-HAD150 and V120-HAD150 began the drying with falling rate period and dried gradually after the transition to hot air convective dryer (150°C). There was no increment in drying rate as the moisture content at the transition stage for both V60-HAD150 and V120-HAD150 was 13.6% d.b. and 3.8% d.b., respectively while the drying rate of V60-HAD150 and V120-HAD150 is 0.00281 and 000138 g H_2O min^{-1}. A prolonged vacuum pre-drying duration decrease the moisture content in rubber to a low level, which resulting in less increment in drying rate for the step-up of drying temperature to 150°C at second stage of hot air drying.

On the other hand, drying rate curves for a 30 min vacuum pre-drying duration, V30-HAD150 exhibits an increase in drying rate when HAD150 was applied in the second stage of VCD, for both continuous and intermittent drying basis. There was a 36.9% and 41.6% increment of drying rate, which maintained for 90 min and 180 min, for both continuous and intermittent V30-HAD150 drying, respectively. The high drying rate was maintained for a longer period during intermittent drying because of the rewetting cycle. The critical moisture content for continuous and intermittent V30-HAD150 was 15.9% d.b. and 18.8% d.b., respectively. The falling rate began shortly after the moisture content dropped below the critical moisture content. V30-HAD150 took a total of 90 min and 180 min to reach below 5% d.b., which was 33.3% and 25% faster than V120-HAD150 or 33.3% and 18.18% faster than V60-HAD150, in term of continuous drying and intermittent drying, respectively. Thus, the sample dried via vacuum drying for 30 min will be selected for next stage of test plan as it best suits the rubber drying characteristic.

4.3.3.2 Effect of heating temperature

An analysis of drying rate at different air temperatures were used to determine the suitable drying techniques for sample size of 50mm. Figure 4.16a, b shows the drying rate versus moisture ratio for 50mm rubber subjected to two-stage vacuum convective drying (VCD) at vacuum pre-drying duration of 30 min (V30) at various hot air drying temperatures (90, 110, 130, 150ºC), in continuous and intermittent basis.

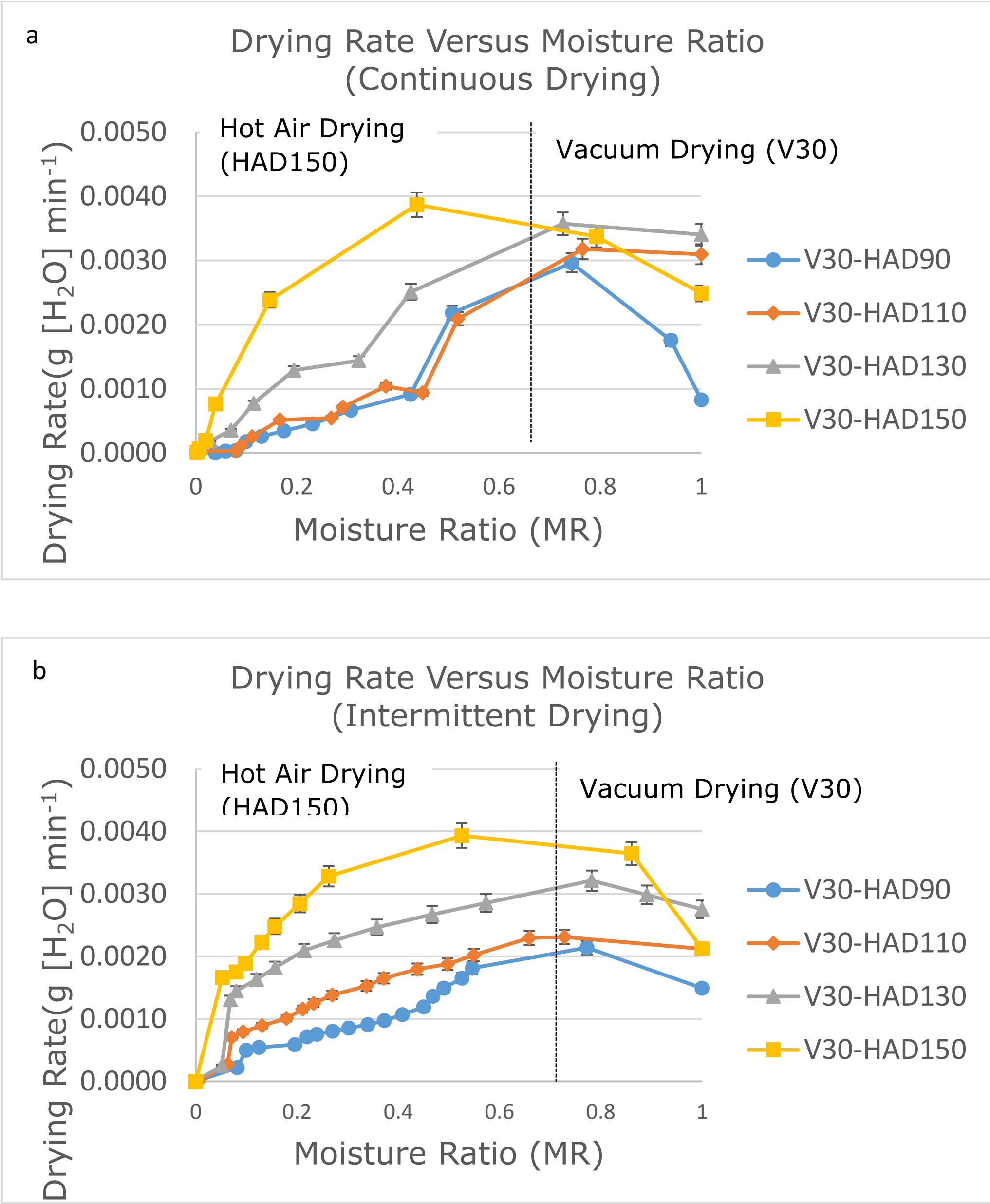


Figure 4.16a, b: Drying rate versus moisture ratio for 50mm rubber subjected to two-stage vacuum convective drying (VCD) with vacuum pre-drying (30 min) at various hot air drying temperature

For intermittent drying, the pulsating of heating gave a smoother drying rate curve, which is a desirable phenomenon. All drying rate curves had an initial transient period, an increasing rate period and a falling rate period. The drying rate increases to a maximum value when the samples were transferred from vacuum dryer to hot air convective dryer, as indicated as vacuum pre-drying period and hot air drying (HAD) in Figure 4.16a, b. The drying rate of intermittent V30-HAD150 increased to the peak at 0.003934 g H_2O min^{-1} during the transition from vacuum dryer to hot air convective dryer. Similarly, the continuous V30-HAD150 have a peak drying rate at 0.003873 g H_2O min^{-1}. The step-up temperature profile of 60°C lead to a larger temperature gradient between sample temperature and heated air, hence, the drying rate for rubber increased.

Figure 4.16a, b indicates that the drying time shortens as the drying temperature increases. The drying rate curves for continuous drying and intermittent drying show similar drying trend. Based on two-stage vacuum convective drying (VCD) at vacuum pre-drying duration of 30 min (V30) at hot air drying temperatures, 150°C (HAD150), V30-HAD150 had the highest drying rate for both continuous drying and intermittent drying, as predicted. It was because drying rate of rubber increase proportionally with heating temperature. The drying rate value for V30-HAD150 was 9.16% - 199.82% and 42.67% - 117.18% higher than continuous drying and intermittent drying of V30-HAD90. The drying rate for V30-HAD150 during initial transient period ranged from 0.00213 - 0.00249 g H_2O min^{-1}, for both continuous and intermittent drying basis. Due to the high heat input after the vacuum drying, the drying rate of continuous V30-HAD150 was boost up to 0.003873 g H_2O min^{-1}. The increment of drying rate for heating temperature at 150°C reach its peak after drying for 120 min. Then the drying rate dropped drastically to 0.000065 g H_2O min^{-1} as the moisture ratio of crumb rubber was lowered to 0.006. This showed that V30-HAD150 in continuous drying basis had a high fluctuation of drying rate compared to V30-HAD150 in intermittent drying basis.

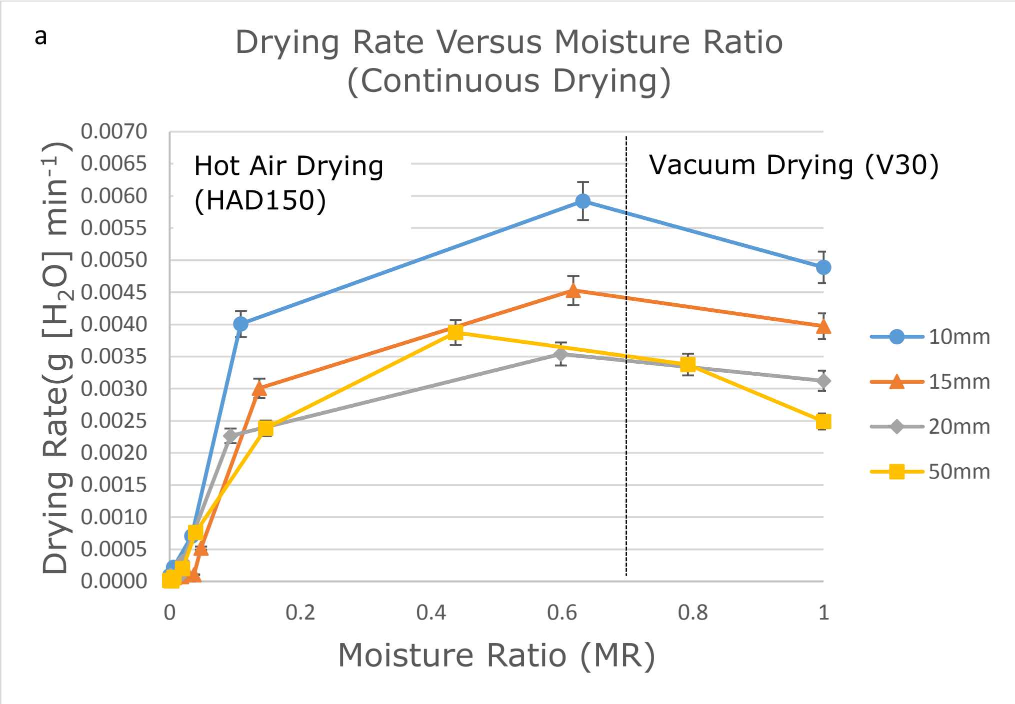
After that, the drying rate values took a considerable longer time to reach the lowest drying rate of 0.001662 g H_2O min^{-1}. This drying rate changes was a result of moisture content changes and rewetting cycles (Jayas, 1991). For the continuous drying, there was no rewetting as heat was constantly supply to rubber material. When more time was given for the rewetting of rubber surface, the problems of wet particles trapped in dried rubber can be eliminated as the heat distribution was more homogeneous.

Figure 4.16a, b shows that the critical moisture content for each drying temperature, 90°C (V30-HAD90), 110°C (V30-HAD110), 130°C (V30-HAD130) and 150°C(V30-HAD150) was 30.8% d.b., 30.4% d.b., 27.3% d.b. and 15.9% d.b., respectively for the continuous two-stage vacuum convective drying (VCD) and 28.7% d.b., 41.2% d.b., 29.2% d.b. and 18.8% d.b., respectively for the intermittent two-stage vacuum convective drying (VCD). The critical moisture content indicates the end of moisture equilibrium at rubber skin (Hall, 1980). The lower critical moisture content values shows that the falling rate begin slower, which was a desirable phenomenon. As the falling rate period begins, a diffusion predominant mechanism take places. Typically, there was two stages of the falling rate period for crumb rubber, similar to other agricultural crops. The remaining surface moisture was removed during the first falling rate period, while the second falling rate period was heavily depending on the moisture diffusion rate. Due to insufficient moisture on the surface, the drying conditions did not affect the drying rate as much at this stage. In short, higher heating temperature should be used for the intermittent period, so that the rubber drying can be maintained at higher drying rate.

4.3.3.3 Effect of sample diameter

Figure 4.17a, b shows the drying rate versus moisture ratio for rubber subjected to two-stage vacuum convective drying (VCD), V30–HAD150 with vacuum pre-drying duration of 30 min (V30) and hot air drying temperature of 150°C (HAD150), continuously and intermittently at varying sample diameter of 10, 15, 20 and 50mm.

The drying curves of crumb rubber consisted of three distinct phases: an initial transient period (during vacuum drying), an increasing rate period (during stage change) and a falling rate period (during hot air drying). At the initial transient period, the rubber was vacuum pre-dried for 30 min at 90°C. Then, a step-up temperature profile from 90°C to 150°C during equipment change lead to the increasing drying rate period. As the temperature gradient increased, the higher heat transfer to rubber promote the moisture diffusion and rewetting of the surface. However, there was no constant rate drying period available as the moisture evaporated from the surface quickly and reach critical moisture content. At the critical moisture content, the surface moisture was in equilibrium with the drying air before the drying rate fall drastically at the moisture ratio of 0.65.



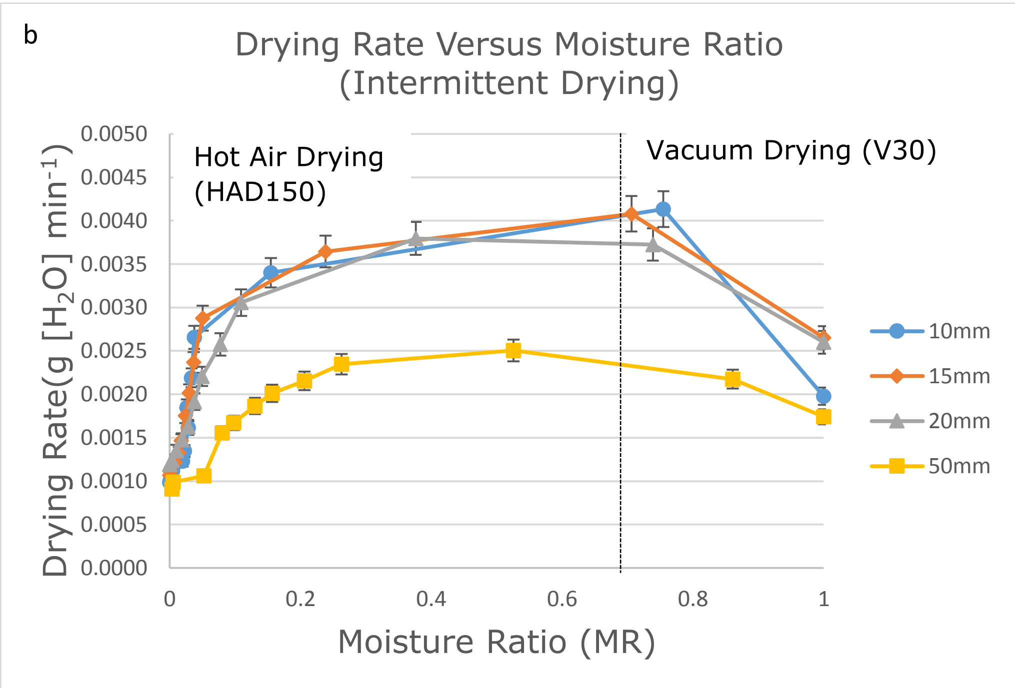
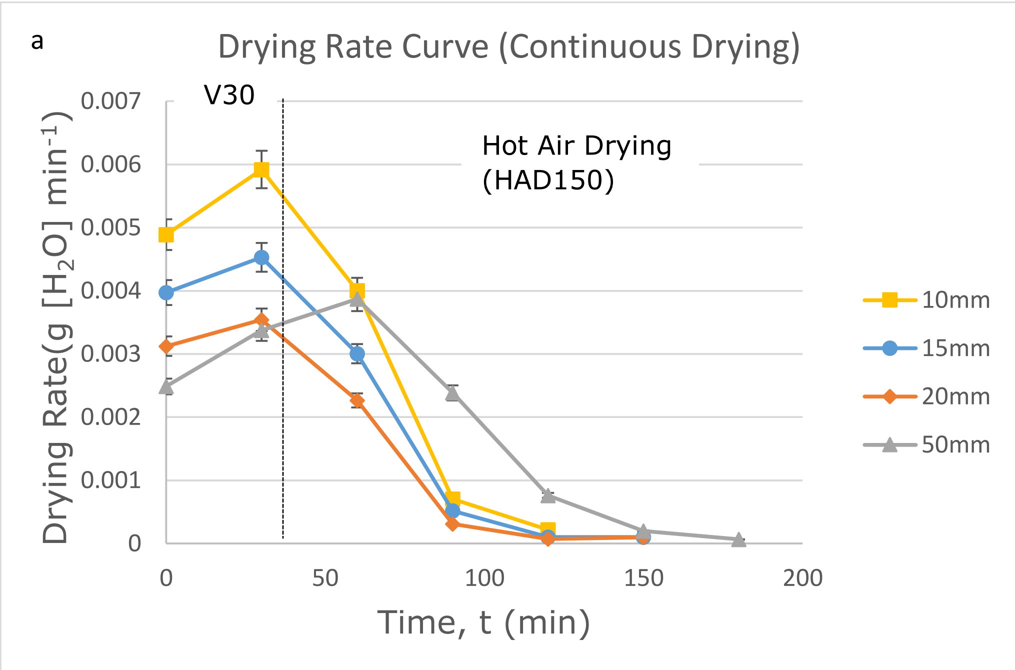


Figure 4.17a, b: Drying rate versus moisture ratio for rubber of varying diameter subjected to two-stage vacuum convective drying (VCD) with vacuum pre-drying (30 min) at hot air drying temperature of 150˚C

The drying rate of thinner (10,15 and 20mm) sample reach its peak at the point of change while the drying rate of 50mm samples reach its peak at moisture ratio of 0.44 and 0.53 for continuous and intermittent VCD, respectively. The diffusion dominant mechanism began with the falling rate period, but not all samples had the same critical moisture content. The critical moisture content for each sample diameter, 10, 15, 20 and 50mm was 25.6% d.b., 19.8% d.b., 14.2% d.b. and 15.9% d.b., respectively for the continuous two-stage vacuum convective drying (VCD) and 18.9% d.b., 18.9% d.b., 11.4% d.b. and 18.8% d.b., respectively for the intermittent two-stage vacuum convective drying (VCD). In overall, the drying rate for thinner samples was higher than thicker samples.

Drying rate curve of rubber with various diameter and various hot air drying temperature subjected to two-stage vacuum convective drying (VCD) with 30 min vacuum pre-drying (VD), followed by hot air drying (HAD), as shown in Figure 4.18a, b, c, d. All drying curves exhibit an initial transient period and a falling rate period. For continuous VCD in Figure 4.18a, the sample diameter that have the highest drying rate of 0.00592 g H_2O min^{-1} was 10mm samples; whereas, the lowest drying rate, 9.22 x 10^{-6} g H_2O min^{-1} fall on 50mm samples. For intermittent VCD in Figure 4.18b, the sample diameter that have the highest drying rate of 0.00413 g H_2O min^{-1} was 10mm samples; whereas, the lowest drying rate, 9.88 x 10^{-4} g H_2O min^{-1} fall on 50mm samples.



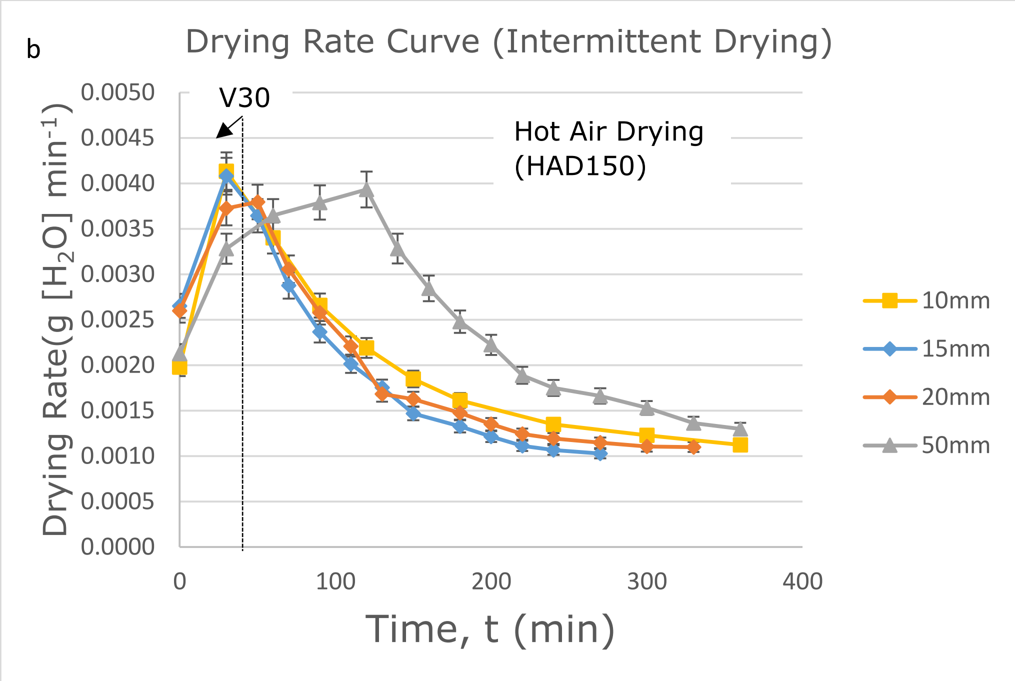
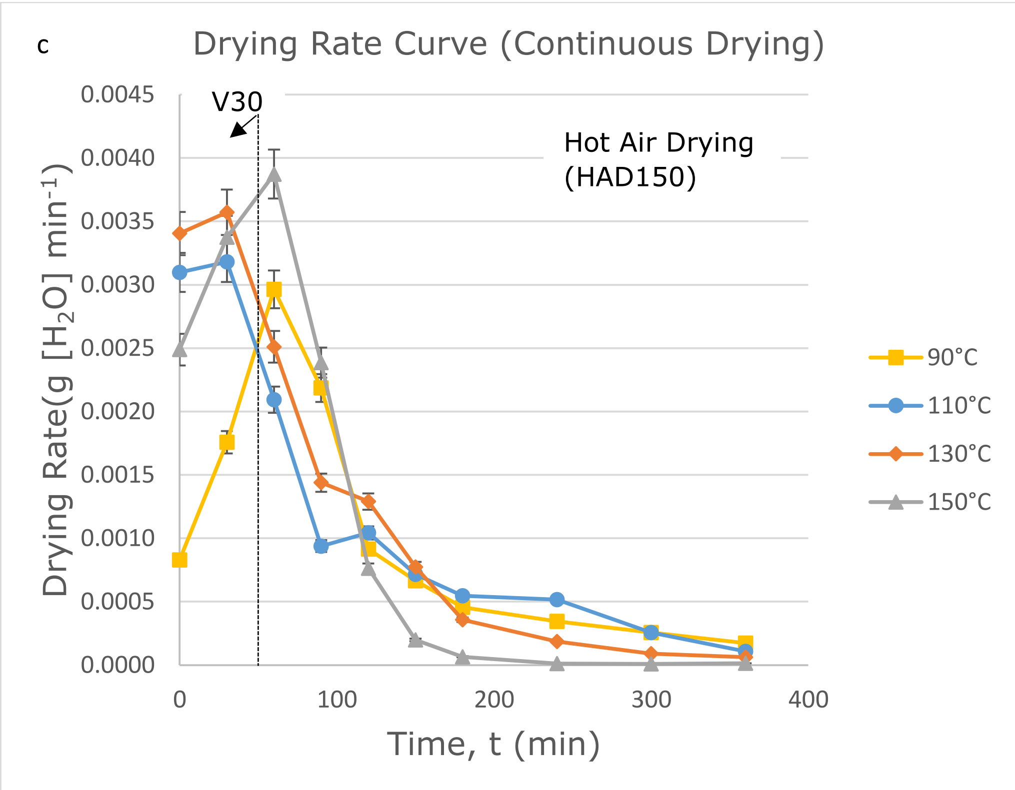


Figure 4.18a, b: Drying rate curve for two-stage vacuum convective drying (VCD) with vacuum pre-drying (30 min) at hot air drying temperature of 150˚C



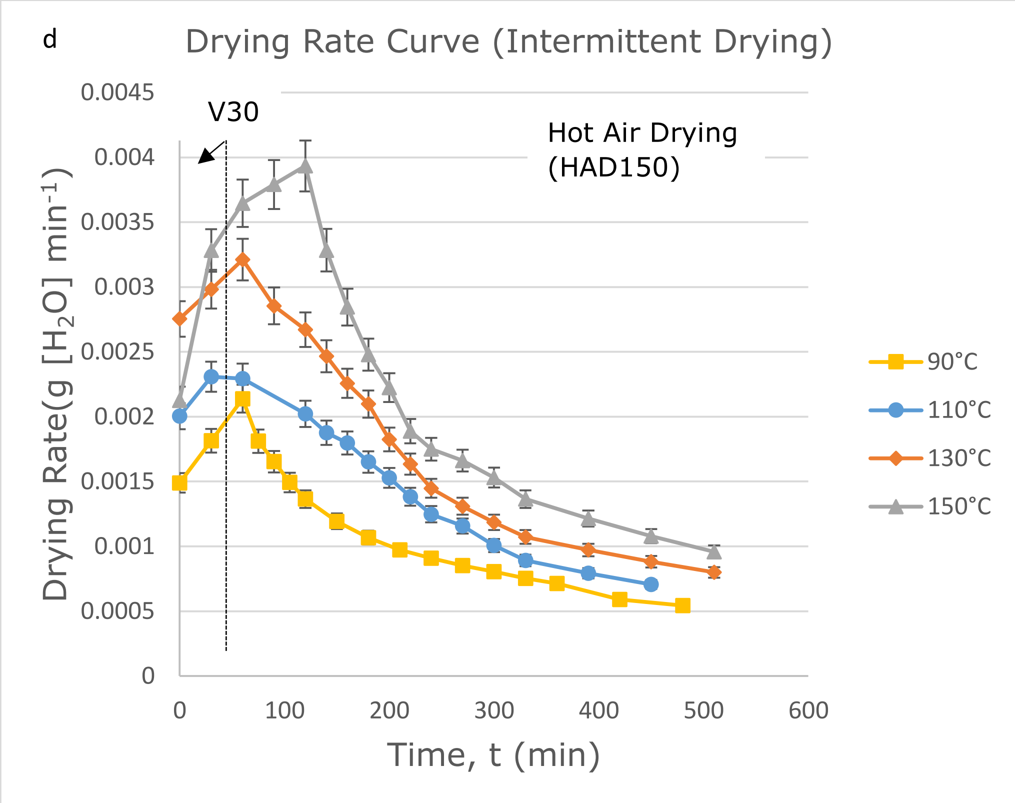


Figure 4.18c, d: Drying rate curve for two-stage vacuum convective drying (VCD) with vacuum pre-drying (30 min) at various hot air drying temperature

Figure 4.18a, b, c, d shows the drying rate curve for rubber with various diameter and various hot air drying temperatures subjected to two-stage vacuum convective drying (VCD) method. There was an increasing rate period for all two-stage VCD, regardless of its sample temperature or sample diameter. The increase in drying rate was mainly due to the higher heat and mass transfer rate for the high step-up temperature during transition from vacuum to hot air drying.

Figure 4.18a shows that rubber sample with 50mm diameter experienced an increment of 35.6% in drying rate during the transition. The internal moisture required longer time to diffuse through 50mm rubber to the surface and this internal heat and mass transfer rates became higher when exposed to high heat. The slow moisture diffusion in thick layer rubber can be improved by using high temperature drying after vacuum pre-drying to enhance the drying kinetics.

Figure 4.18b shows that vacuum drying gave a high drying rate during the initial transient period. The highest drying rate for 50mm sample was 0.0025 g H_2O min^{-1} while the highest drying rate of thinner samples dropped below 0.0030 g H_2O min^{-1} when its moisture ratio was below 0.1 while. Vacuum drying acts as a driven force for moisture to diffuse to surface while hot air drying maintains the vapour pressure of air and evaporation of moisture from the rubber surface. The drying rate for 50mm sample dropped gradually for moisture ratio value of 0.3 to 0.1 while the drying rate for thinner sample dropped drastically once it reached a moisture ratio of 0.1. The beginning of falling rate period differ for thin and thick samples was due to the inherent cross-sectional diameter. Piddlesden (1937) also stated that the drying time of rubber was proportional to the square of its diameter, especially for rubber with low porosity value.

Likewise, Figure 4.18c, d for continuous VCD and intermittent VCD drying rate curve shows that the rubber samples dried at a higher hot air drying (HAD) temperature tends to have a higher drying rate. At higher heating temperature, the internal moisture required shorter time to diffuse to the surface and this internal heat and mass transfer rates became higher when exposed to high heat. The improvement in drying rate was obvious as the Figure 4.18d curves split into four non-overlapping lines, compared to continuous drying. The high drying rate for the second stage intermittent HAD at 150°C may ensure a higher uniformity in heat distribution within rubber particles. A homogeneous heat distribution would allow all parts of rubber dried at the same pace, without any wet rubber particles trapped in the dried products. This showed that the moisture diffusion of crumb rubber can be improved by using high temperature intermittent drying, for a higher heat and mass transfer rate. Thus, intermittent drying of two-stage vacuum convective drying, with vacuum duration of 30 min and hot air drying temperature of 150°C was chosen for subsequent optimization study.

Table 4.7 and 4.8 shows the drying rates for rubber dried for experiments carried out under various drying conditions, included vacuum drying duration, heating temperature and sample diameter. The drying rate value was expressed as the evaporation rate per unit of time, g H_2O min^{-1} in Table 4.7 and 4.8 below. The highest average drying rate value was obtained from the comparison of average drying rate value for vacuum duration of 120 min (V120), 60 min (V60) and 30 min (V30). Statistical analysis (analysis of variance, ANOVA) showed that the change of heating temperature had statistically significant influence on the drying rates of crumb rubber, $p<0.015$, while vacuum drying duration and sample diameter do not had significant effect to drying rates for both continuous and intermittent drying.

Table 4.7: Average drying rate and the highest drying rate values for two-stage vacuum convective drying of crumb rubber in continuous mode

Continuous Drying					
Hot Air Drying (HAD) Temperature		Average Drying Rate for Varies Vacuum Drying Time (Min)			Highest Average Drying Rate
		V120	V60	V30	(g H_2O min^{-1})
	90°C	0.00069 [b]	0.00082 [b]	0.00056 [b]	0.00082
	110°C	0.00117 [a]	0.00155 [a]	0.00187 [a]	0.00187
	130°C	0.00144 [a]	0.00200 [c]	0.00152 [a]	0.00200
10mm	150°C	0.00134 [a]	0.00154 [a]	0.00329 [c]	0.00329
	90°C	0.00085 [b]	0.00136 [a]	0.00100 [a]	0.00136
	110°C	0.00120 [a]	0.00176 [a]	0.00188 [a]	0.00188
	130°C	0.00122 [a]	0.00157 [a]	0.00201 [a]	0.00201
15mm	150°C	0.00173 [a]	0.00153 [a]	0.00182 [a]	0.00182
	90°C	0.00087 [b]	0.00101 [a]	0.00139 [a]	0.00139
	110°C	0.00134 [a]	0.00129 [a]	0.00187 [a]	0.00187
	130°C	0.00203 [a]	0.00206 [a]	0.00161 [a]	0.00206
20mm	150°C	0.00179 [a]	0.00195 [a]	0.00237 [a]	0.00237
	90°C	0.00076 [a]	0.00065 [a]	0.00064 [b]	0.00076
	110°C	0.00097 [a]	0.00096 [a]	0.00077 [b]	0.00097
	130°C	0.00137 [a]	0.00131 [a]	0.00120 [a]	0.00137
50mm	150°C	0.00132 [a]	0.00137 [a]	0.00153 [a]	0.00153

Note: Means in the same column that do not share subscripts differ at $p < 0.05$ probability level.

Table 4.8: Average drying rate and the highest drying rate values for two-stage vacuum convective drying of crumb rubber in intermittent mode

Intermittent Drying					
Hot Air Drying (HAD) Temperature		Average Drying Rate for Varies Vacuum Drying Time (Min)			Highest Average Drying Rate (g H_2O min^{-1})
		V120	V60	V30	
	90°C	0.00280 [a]	0.00263 [a]	0.00368 [b]	0.00368
	110°C	0.00180 [a]	0.00207 [a]	0.00245 [a]	0.00245
	130°C	0.00288 [a]	0.00234 [a]	0.00423 [b]	0.00423
10mm	150°C	0.00197 [a]	0.00251 [a]	0.00414 [b]	0.00414
	90°C	0.00213 [a]	0.00270 [a]	0.00339 [b]	0.00339
	110°C	0.00196 [a]	0.00201 [a]	0.00191 [a]	0.00201
	130°C	0.00320 [b]	0.00306 [b]	0.00337 [b]	0.00337
15mm	150°C	0.00206 [a]	0.00305 [b]	0.00408 [b]	0.00408
	90°C	0.00244 [a]	0.00253 [a]	0.00413 [b]	0.00413
	110°C	0.00171 [a]	0.00229 [a]	0.00218 [a]	0.00229
	130°C	0.00290 [b]	0.00321 [b]	0.00357 [b]	0.00357
20mm	150°C	0.00261 [a]	0.00297 [b]	0.00380 [b]	0.00380
	90°C	0.00176 [a]	0.00151 [a]	0.00149 [a]	0.00176
	110°C	0.00454 [b]	0.00481 [b]	0.00290 [a]	0.00481
	130°C	0.00371 [b]	0.00436 [b]	0.00321 [b]	0.00436
50mm	150°C	0.00165 [a]	0.00228 [a]	0.00393 [b]	0.00393

Note: Means in the same column that do not share subscripts differ at $p < 0.05$ probability level.

From Table 4.7, the 10mm rubber sample subjected to two-stage vacuum convective drying, V30-HAD150, was dried continuously had a 115% higher drying rate compared to the 50mm sample subjected same parameters, continuous V30-HAD150. The results show that thin samples could dry faster than thick samples. Likewise, from Table 4.8, the 10mm sample dried via V30-HAD150, intermittently had 5.07% higher drying rate compared to the 50mm sample dried via V30-HAD150, intermittently. This phenomenon was due to the shorter cross-sectional dimension for thin samples. Thin samples experienced a higher heat and mass transfer rate compared to thick samples, especially when drying with hot air convective dryer. The heat transfer from the surface to core was faster hence the moisture diffusion increased, lead to higher drying rate.

A comparison down the Table 4.7 and 4.8 on the hot air drying temperature of 150°C (HAD150) and 90°C (HAD90) shows 139% and 163.8% higher drying rate for 50mm samples, dried via V30-HAD150, compared to V30-HAD90, for continuous and intermittent mode of drying. From the drying rate, the critical temperature identified for rubber was 150°C, which the drying of rubber was averagely faster than 130, 110 and 90°C. Similarly, a study on the effect of temperature to tomato slices showed that drying revolving the critical temperature would be able to preserve quality and dry efficiently (Correla, 2015). In addition, the high increment in drying rate showed that hot air drying temperature is one of the key parameters to higher drying rate values, especially during intermittent drying process. A higher heating temperature gave a higher average drying rate. As the water molecules were loosely bound to the rubber, less energy was required to remove than at lower temperature. Thus, the drying rate was higher for high temperature intermittent drying.

A comparison across the Table 4.7 and 4.8 shows that two-stage vacuum convective drying (VCD) with a shorter vacuum pre-drying duration, 30 min (V30) was a preferable choice. Most of the highest average drying rate values were from V30, up to 56.25% and 68.75% of all the experiments conducted in continuous VCD and intermittent VCD. On top of that, all the average drying rate (highest) values of intermittent VCD were higher than the average drying rate (highest) values of continuous VCD. The highest drying rate, 0.00481 g H_2O min^{-1} was found for intermittent V60-HAD110, two-stage VCD with 60 min vacuum drying duration (V60) and hot air drying temperature at 110°C (HAD110). Even though intermittent

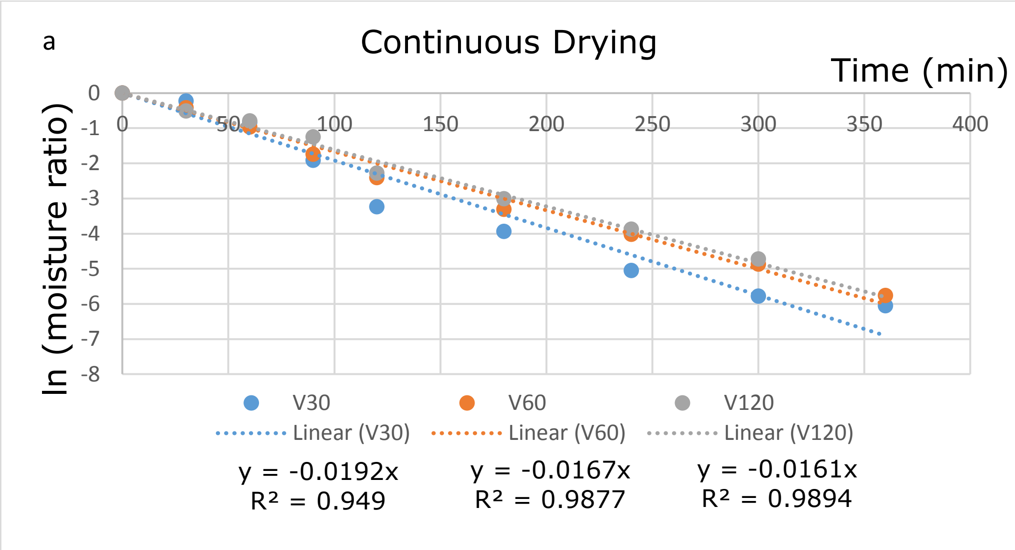
V60-HAD110 might be to dry the sample efficiently, but drying at 110°C took a considerably longer time, which was 60% and 166.7% longer duration than drying at 130°C and 150°C. Thus, the vacuum pre-drying duration should be resorted to 30 min.

4.3.4 Moisture diffusivity

The effective moisture diffusivity was determined by Fick's second law of diffusion, by measuring the slope from a curve plotted from the natural logarithm of moisture ratio. The effective moisture diffusivity values for all samples were calculated from Figure 4.19a, b, c, d, e, f and expressed as m^2s^{-1} in Table 4.9. This behaviour of the moisture diffusivity might be the result of the changing mechanism during drying: in the first stage of vacuum pre-drying, removal of surface moisture might be the main mass transfer mechanism. Then, diffusion of moisture from the core to surface may be predominant as the drying progresses.

The effect of vacuum duration, heating temperature and sample diameter on their respective moisture diffusion were investigated for both continuous and intermittent basis of two-stage vacuum convective drying. The effective moisture diffusivity for various drying conditions was determined by

Fick's second law of diffusion, by measuring the slope from Figure 4.19a, b, c, d, e, f plotted from the natural logarithm of moisture ratio. Their dependence on temperature was estimated by using an Arrhenius-type relationship to develop the Arrhenius equations.



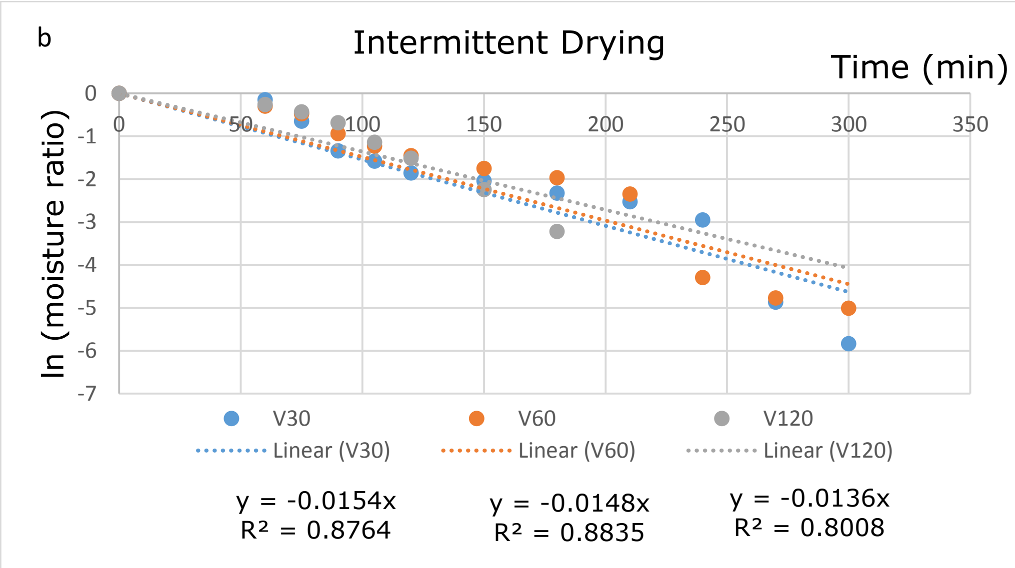
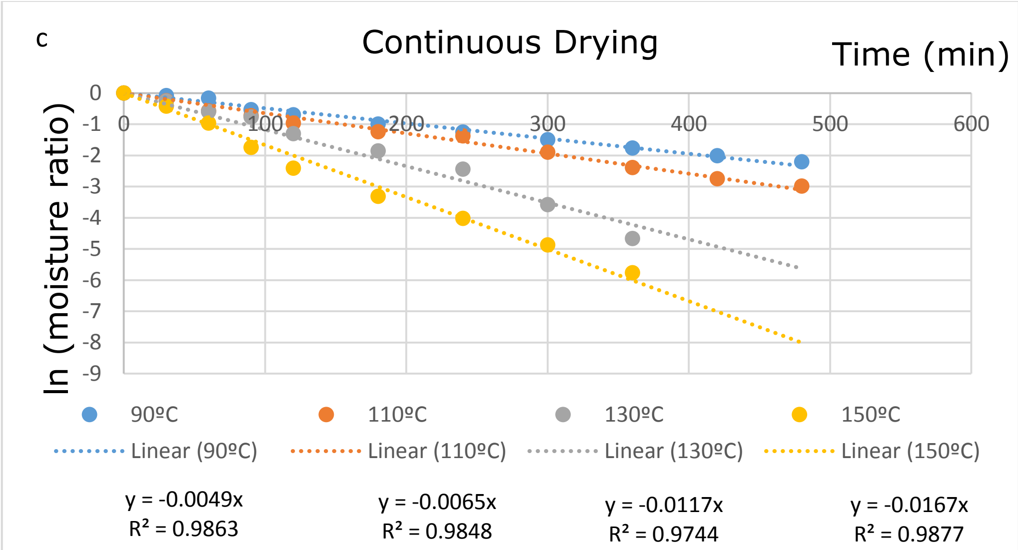


Figure 4.19 a, b: Drying curve of the natural logarithm of moisture ratio versus time for varying vacuum duration of 30 min (V30), 60 min (V60) and 120 min (V120), in continuous and intermittent basis



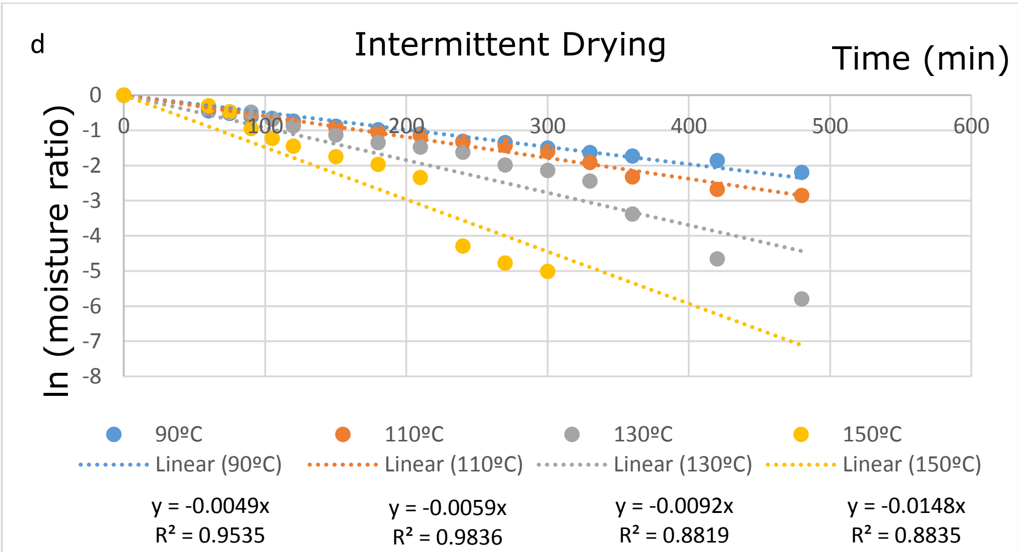


Figure 4.19 c, d: Drying curve of the natural logarithm of moisture ratio versus time for varying drying temperatures (90, 110, 130 and 150 ˚C), in continuous and intermittent basis

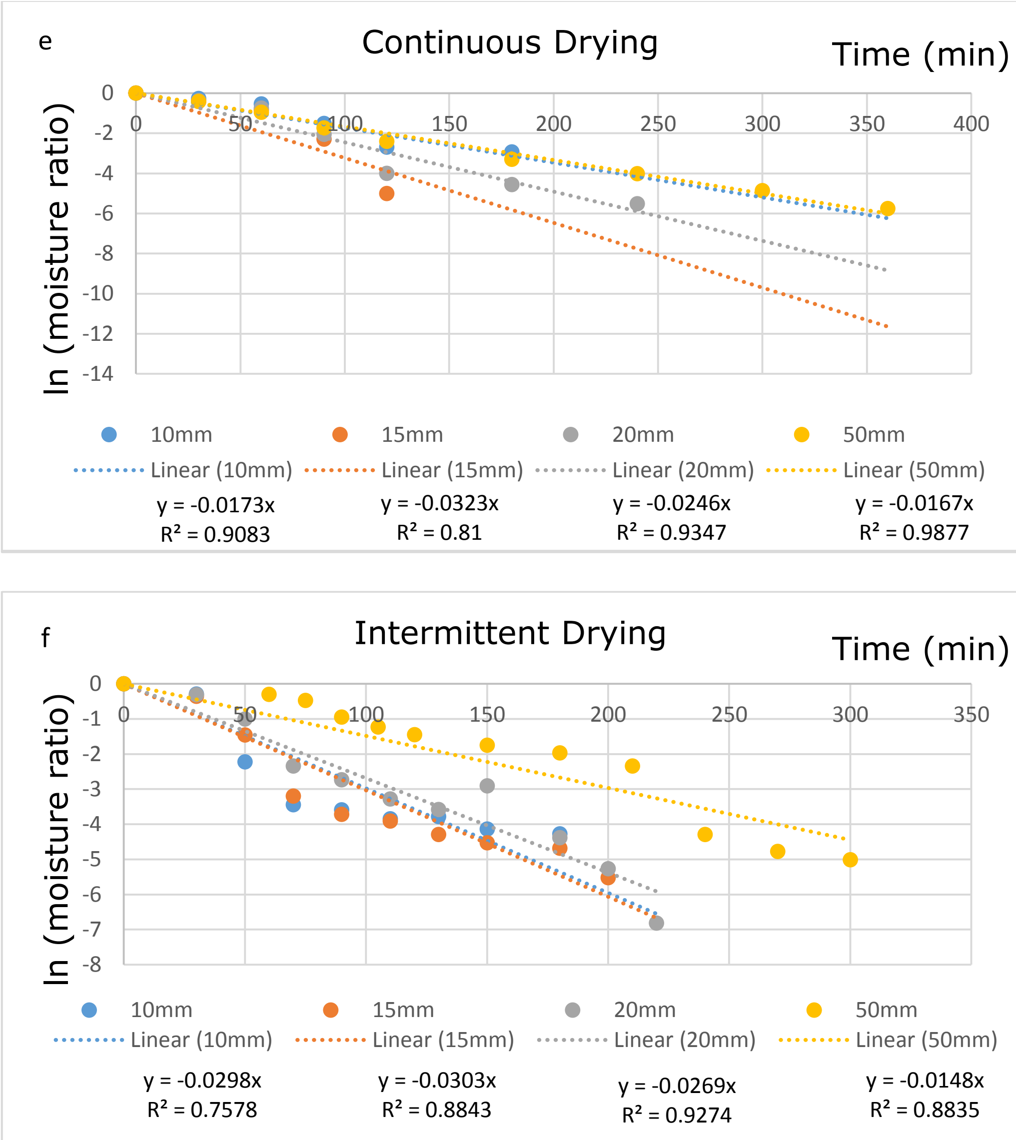


Figure 4.19 e, f: Drying curve of the natural logarithm of moisture ratio versus time for varying sample diameter (10, 15, 20 and 50mm), in continuous and intermittent basis

Table 4.9: Effective moisture diffusivity for two-stage vacuum convective drying (VCD)

Continuous Drying					Intermittent Drying				
Effective Moisture Diffusivity for first drying stage V30, D_{eff} $*10^{-9}$ /m^2 s^{-1}					Effective Moisture Diffusivity for first drying stage V30, D_{eff} $*10^{-9}$ /m^2 s^{-1}				
Temperature/ Diameter	10mm	15mm	20mm	50mm	Temperature/ Diameter	10mm	15mm	20mm	50mm
90°C	0.0352[a]	0.1387[a]	0.2948[a]	1.2378[b]	90°C	0.0691[a]	0.1407[a]	0.2919[a]	1.2378[b]
110°C	0.1046[a]	0.2575[a]	0.5458[a]	1.8979[b]	110°C	0.0716[a]	0.1789[a]	0.2815[a]	1.7588[b]
130°C	0.2420[a]	0.5644[a]	0.8802[a]	3.9609[c]	130°C	0.2163[a]	0.3869[a]	0.9954[a]	3.8109[c]
150°C	0.4555[a]	1.1709[b]	1.1926[b]	6.4089[c]	150°C	0.3100[a]	0.8032[a]	1.4540[b]	4.9184[c]
Effective Moisture Diffusivity for first drying stage V60, D_{eff} $*10^{-9}$ /m^2 s^{-1}					Effective Moisture Diffusivity for first drying stage V60, D_{eff} $*10^{-9}$ /m^2 s^{-1}				
Temperature/ Diameter	10mm	15mm	20mm	50mm	Temperature/ Diameter	10mm	15mm	20mm	50mm
90°C	0.0585[a]	0.1227[a]	0.1982[a]	1.1897[b]	90°C	0.0766[a]	0.1410[a]	0.2043[a]	1.1897[b]
110°C	0.1001[a]	0.1807[a]	0.3450[a]	1.8093[b]	110°C	0.0360[a]	0.1227[a]	0.2739[a]	1.5960[b]
130°C	0.2826[a]	0.4127[a]	0.6305[a]	3.9656[c]	130°C	0.1427[a]	0.3787[a]	0.4503[a]	2.2924[b]
150°C	0.2131[a]	0.5353[a]	1.3205[b]	5.0561[c]	150°C	0.3331[a]	0.6790[a]	1.0539[b]	3.5112[c]
Effective Moisture Diffusivity for first drying stage V120, D_{eff} $*10^{-9}$ /m^2 s^{-1}					Effective Moisture Diffusivity for first drying stage V120, D_{eff} $*10^{-9}$ /m^2 s^{-1}				
Temperature/ Diameter	10mm	15mm	20mm	50mm	Temperature/ Diameter	10mm	15mm	20mm	50mm
90°C	0.0326[a]	0.1002[a]	0.1611[a]	0.8998[a]	90°C	0.0635[a]	0.1129[a]	0.1433[a]	0.8705[a]
110°C	0.0540[a]	0.1793[a]	0.2536[a]	1.6281[b]	110°C	0.0369[a]	0.1083[a]	0.2089[a]	1.3826[b]
130°C	0.1259[a]	0.3355[a]	0.5313[a]	2.7849[b]	130°C	0.0880[a]	0.2512[a]	0.4055[a]	1.6131[b]
150°C	0.1346[a]	0.2777[a]	0.5655[a]	4.4988[c]	150°C	0.2878[a]	1.1568[b]	2.3597[b]	2.3557[b]

Continuous Drying					Intermittent Drying				
Effective Moisture Diffusivity for second drying stage (HAD), D_{eff} *10^{-9} /m^2 s^{-1}					Effective Moisture Diffusivity for second drying stage (HAD), D_{eff} *10^{-9} /m^2 s^{-1}				
Temperature/ Diameter	10mm	15mm	20mm	50mm	Temperature/ Diameter	10mm	15mm	20mm	50mm
90°C	0.324[a]	1.277[b]	2.715[b]	11.399[d]	90°C	0.537[a]	1.094[b]	2.270[b]	9.626[c]
110°C	0.963[a]	2.371[b]	5.026[c]	17.478[d]	110°C	0.557[a]	1.391[b]	2.189[b]	13.678[d]
130°C	2.229[b]	5.198[c]	8.106[c]	36.476[e]	130°C	1.682[b]	3.009[c]	7.741[c]	29.636[e]
150°C	4.195[c]	10.783[d]	10.983[d]	59.020[e]	150°C	2.411[b]	6.246[c]	11.307[d]	38.249[e]
Effective Moisture Diffusivity for second drying stage (HAD), D_{eff} *10^{-9} /m^2 s^{-1}					Effective Moisture Diffusivity for second drying stage (HAD), D_{eff} *10^{-9} /m^2 s^{-1}				
Temperature/ Diameter	10mm	15mm	20mm	50mm	Temperature/ Diameter	10mm	15mm	20mm	50mm
90°C	0.598[a]	1.254[b]	2.026[b]	12.159[d]	90°C	0.669[a]	1.231[b]	1.783[b]	10.385[d]
110°C	1.023[b]	1.847[b]	3.526[c]	18.491[d]	110°C	0.314[a]	1.071[b]	2.391[b]	13.932[d]
130°C	2.888[b]	4.218[c]	6.444[c]	40.529[e]	130°C	1.246[b]	3.306[c]	3.931[c]	20.011[e]
150°C	2.178[b]	5.471[c]	13.496[d]	51.674[e]	150°C	2.908[b]	5.927[c]	9.200[c]	30.650[e]
Effective Moisture Diffusivity for second drying stage (HAD), D_{eff} *10^{-9} /m^2 s^{-1}					Effective Moisture Diffusivity for second drying stage (HAD), D_{eff} *10^{-9} /m^2 s^{-1}				
Temperature/ Diameter	10mm	15mm	20mm	50mm	Temperature/ Diameter	10mm	15mm	20mm	50mm
90°C	0.385[a]	1.185[b]	1.905[b]	10.639[d]	90°C	0.628[a]	1.117[b]	1.418[b]	8.612[c]
110°C	0.638[a]	2.120[b]	2.999[b]	19.251[d]	110°C	0.365[a]	1.071[b]	2.067[b]	13.678[d]
130°C	1.489[b]	3.967[c]	6.282[c]	32.929[e]	130°C	0.871[a]	2.485[b]	4.012[c]	15.958[d]
150°C	1.591[b]	3.283[c]	6.687[c]	53.194[e]	150°C	2.847[b]	11.444[d]	23.344[e]	23.304[e]

Note: Means in the same column that do not share subscripts differ at $p < 0.05$ probability level.

As can be seen in Table 4.9, the effective moisture diffusivity, D_{eff} values for continuous drying were in the range of 0.324 x 10^{-9} to 59.020 x 10^{-9} m^2s^{-1} while the D_{eff} values for intermittent drying were in the range of 0.314 x 10^{-9} to 38.249 x 10^{-9} m^2s^{-1}. Both continuous and intermittent drying results were in accordance with other agricultural and food products, which the typical D_{eff} values is in the range of 10^{-11} to 10^{-9} m^2s^{-1} (Karathanos et al., 1990; Zogzas et al., 1996; Bezerram et al., 2015). The highest moisture diffusivity value found for the two-stage vacuum convective drying (VCD) was 59.020 x 10^{-9} /m^2 s^{-1}, which the sample was subjected to short vacuum pre-drying of 30 min (V30) and hot air drying of 150°C (HAD150) for 50mm samples. The D_{eff} increases with increasing sample diameter.

A comparison of rubber at various diameter (10, 15, 20, 50mm) showed that rubber of smaller size had a lower effective moisture diffusivity than thicker rubber. Likewise, Kabiru et al. (2013) reported that the effective moisture diffusivity coefficients of mango increased with increasing temperature. The results showed that the highest equilibrium moisture diffusivity obtained was 6.99 x 10^{-10} when a 3mm thick mango that were dried at 80°C. Statistical analysis (analysis of variance, ANOVA) with the use of response optimizer tool in Minitab 18 showed that a thicker sample significantly improve the diffusivity values. The evaluated results based on the optimization plot to maximize the effective moisture diffusivity were shows as below:

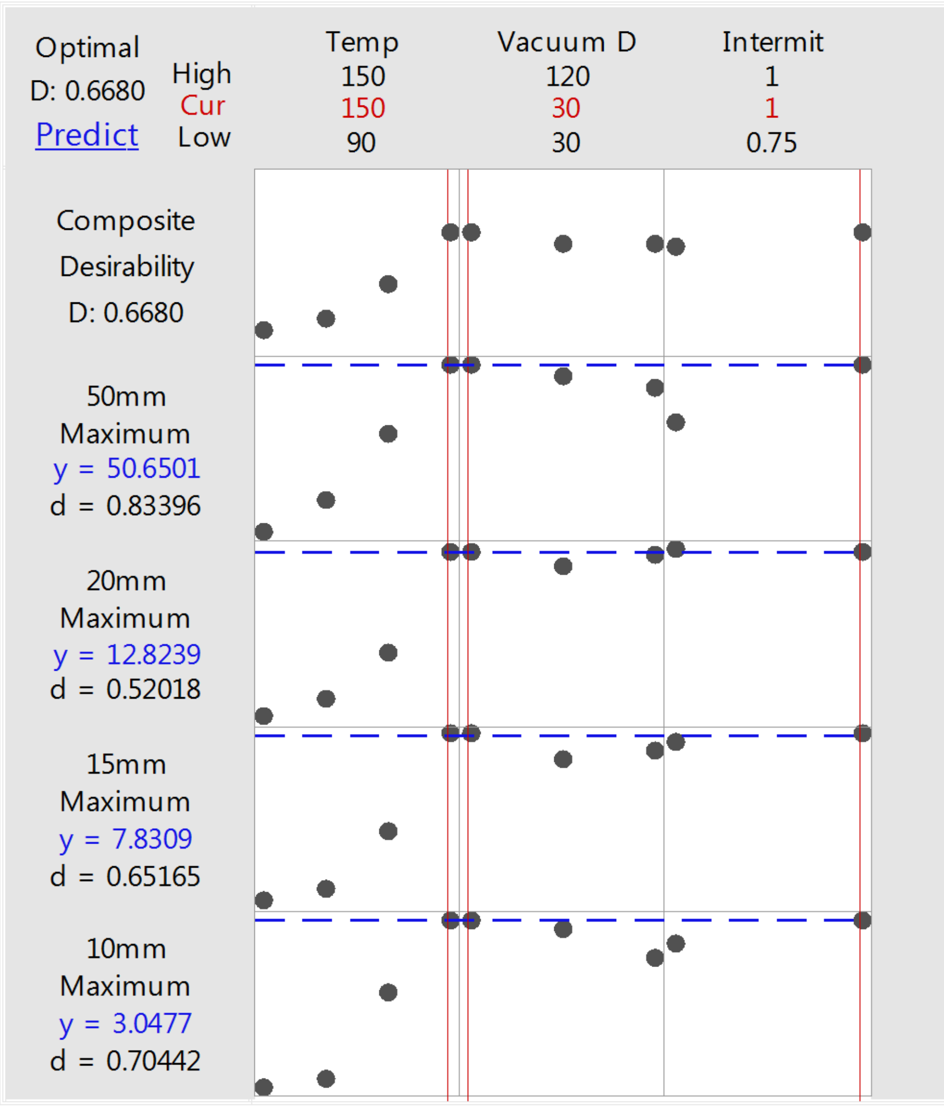


Figure 4.20: The optimization plot for effective diffusivity of various sample sizes

The y values in the optimization plot indicated the yield of the response. Based on the results of 50mm, the optimization plot showed that the vacuum duration of 30 min and heating temperature of 150°C of the two stage vacuum convective drying gave the highest yield among all four sizes, 50.65, which was the most optimum condition in this case.

The dominant controlling mechanism of the drying process can be improved by increasing the drying temperature or reducing the sample diameter, which the improvement can be determined by higher effective moisture diffusivity values. An increase in temperature, a decrease in vacuum duration and a decrease in sample diameter promoted the value of effective moisture diffusivity, D_{eff}. Likewise, vacuum drying study of radish slice showed an increment in effective moisture diffusivity with higher drying temperature and lower sample diameter (Lee, 2009).

This behaviour of the moisture diffusivity resultant from the two-stage vacuum convective drying gave an insight to the fundamental drying characteristic of crumb rubber. The diffusivity value for intermittent drying results was slightly lower than the diffusivity value for continuous drying results, with exception to the two-stage vacuum convective drying with longer vacuum pre-drying duration of 120 min (V120) and high drying temperature at 150°C (HAD150). This might be due to the calculation based on linearized analytical solutions of Fick's diffusion equation, assuming constant diffusivity. Similarly, Cavusoglu (2008) using the same calculation method, but the D_{eff} values obtained for intermittent drying of tomato was higher than continuous. The lower diffusivity found in this study might be due to the short and frequent pulsating of heat, which the drying of samples will be paused for a 5-minute interval for every 15 minutes heating in the hot air convective dryer. This tempering time allows the moisture within crumb rubber to redistribute and diffuse to surface.

However, the transport of moisture might be decreased as there was no heat input for the rubber samples. As the heating resume, the sample temperature increased as the heat was absorbed by the sample's body, which promote the heat and mass transfer. As the duration of sample under constant heat stress was reduced during the tempering period, the D_{eff} values were not as high as continuous drying. In short, a 50mm thick rubber will be used in the stepwise profile study for the second stage hot air drying in two – stage intermittent VCD.

Table 4.10 and 4.11 shows the fitted D_{eff} value and related R^2 value at constant air temperature of 90°C for two-stage VCD. The activation energy was calculated using Eq. 6. The value of activation energy, E_a varies from 29.97 to 54.6 kJ mol^{-1} for continuous drying and 20.07 to 1205.65 kJ mol^{-1} for intermittent drying, as shown in Table 4.10 and 4.11, respectively.

Table 4.10: Fitted equations for D value at constant temperature for varying vacuum duration of 30 min (V30), 60 min (V60) and 120 min (V120) coupled with continuous hot air drying (HAD)

Fitted equations for Deff value at constant air temp, 90 for vacuum pre-drying duration of 30 min

Size	Arrhenius Equation	R^2
10	D_{eff} = $(3.242\text{x}10^{-10})\exp(-54.6/RgT)$	0.9946
15	D_{eff} = $(1.277\text{x}10^{-09})\exp(-45.7/RgT)$	0.9926
20	D_{eff} = $(2.715\text{x}10^{-09})\exp(-30.0/RgT)$	0.9894
50	D_{eff} = $(1.140\text{x}10^{-08})\exp(-36.1/RgT)$	0.9855

Fitted equations for Deff value at constant air temp, 90 for vacuum pre-drying duration of 60 min

Size	Arrhenius Equation	R^2
10	D_{eff} = $(5.978\text{x}10^{-10})\exp(-31.9/RgT)$	0.8081
15	D_{eff} = $(1.254\text{x}10^{-09})\exp(-33.5/RgT)$	0.9641
20	D_{eff} = $(2.026\text{x}10^{-09})\exp(-40.0/RgT)$	0.9875
50	D_{eff} = $(1.216\text{x}10^{-08})\exp(-32.8/RgT)$	0.9683

Fitted equations for Deff value at constant air temp, 90 for vacuum pre-drying duration of 120 min

Size	Arrhenius Equation	R^2
10	D_{eff} = $(3.850\text{x}10^{-10})\exp(-32.8/RgT)$	0.9348
15	D_{eff} = $(1.185\text{x}10^{-09})\exp(-24.0/RgT)$	0.8167
20	D_{eff} = $(1.905\text{x}10^{-09})\exp(-29.0/RgT)$	0.9369
50	D_{eff} = $(1.064\text{x}10^{-08})\exp(-34.3/RgT)$	1

Table 4.11: Fitted equations for D value at constant temperature for varying vacuum duration of 30 min (V30), 60 min (V60) and 120 min (V120) coupled with continuous hot air drying (HAD)

Fitted equations for Deff value at constant air temp, 90 for vacuum pre-drying duration of 30 min

Size	Arrhenius Equation	R^2
10	D_{eff} = $(5.370x10^{-10})exp(-35.5/RgT)$	0.879
15	D_{eff} = $(1.094x10^{-09})exp(-37.9/RgT)$	0.941
20	D_{eff} = $(2.270x10^{-09})exp(-38.5/RgT)$	0.853
50	D_{eff} = $(9.626x10^{-09})exp(-31.4/RgT)$	0.964

Fitted equations for Deff value at constant air temp, 90 for vacuum pre-drying duration of 60 min

Size	Arrhenius Equation	R^2
10	D_{eff} = $(6.687x10^{-10})exp(-1164.9/RgT)$	0.8917
15	D_{eff} = $(1.231x10^{-09})exp(-598.8/RgT)$	0.929
20	D_{eff} = $(1.783x10^{-09})exp(-449.5/RgT)$	0.9981
50	D_{eff} = $(1.039x10^{-08})exp(-23.1/RgT)$	0.9838

Fitted equations for Deff value at constant air temp, 90 for vacuum pre-drying duration of 120 min

Size	Arrhenius Equation	R^2
10	D_{eff} = $(6.282x10^{-10})exp(-1205.6/RgT)$	0.9849
15	D_{eff} = $(1.117x10^{-09})exp(-1135.4/RgT)$	1
20	D_{eff} = $(1.419x10^{-09})exp(-1024.6/RgT)$	0.9884
50	D_{eff} = $(8.612x10^{-09})exp(-20.1/RgT)$	0.9724

In the present study, the value of E_a varied from 20.07 to 1205.65 kJ mol^{-1} at different sample diameter of rubber, as shown in Table 4.10 and 4.11. For continuous drying process, the activation energy for thinner samples (10, 15, 20 mm) were significantly different from thicker samples (50mm). As the moisture removal from rubber was governed by diffusion phenomenon, it was deduced that at a lower sample diameter (10mm), the rubber surface had a better contact with heated air, which results in a greater absorption of heat. Thus, the moisture gradient of sample increased, with an increase in the moisture diffusivity. Similarly, a study on the effect of air velocity to apricots showed that the diffusivity was reduced at lower moisture gradient.

The second-degree polynomial equation was fitted on the data to determine the prediction of activation energy based on the sample diameter. Based on the curve fitting, a basic prediction on the activation energy of continuous drying V30 and intermittent drying V30 can be done for two-stage vacuum convective drying with vacuum pre-drying of 30 and 120 min, respectively. The equation for 30 min was $y = 0.0638x^2 - 4.328x + 92.959$ while the equation for 120 min was $y = -0.3905x^2 - 6.29x + 1310.6$, with of $R^2 = 0.9515$ and 0.999, respectively.

4.3.5 Rubber quality of two-stage Vacuum Convective Drying (VCD)

Both visual attributes and textural attributes were tested to study the effect of vacuum - convective drying (VCD). The visual attributes were measured using colour measurement system (Hunter Lab, USA) while the textural attributes were measured by a TA-XT. Plus Texture Analyser (Stable Micro Systems, Co., UK). Significant alterations in the colour quality and texture of rubber samples were observed for every single experiment run, including both continuous drying and intermittent drying.

4.3.5.1 Colour analysis

Colour characteristics of crumb rubber were studied by evaluation of total colour change via measuring lightness (L*), redness (a*) and yellowness (b*) using a Hunter Lab calorimeter for both continuous drying and intermittent two-stage vacuum convective drying (VCD). Table 4.12 shows the colour parameters of standard rubber colour. The effect of vacuum duration and heating temperature to the total colour change (ΔE) and colour parameters (L*, a*, b*) were as shown in Table 4.13 and 4.14 for crumb rubber samples that undergo two-stage vacuum-convective drying, for continuous and intermittent basis, respectively.

Table 4.12 Colour parameters of fresh crumb rubber and standard rubber colour

	L*	a*	b*
Fresh Rubber Colour	50.53 ± 6.513 [a]	1.65 ± 2.55 [e]	13.35 ± 7.82 [g]
Standard Rubber Colour	18.9 ± 3.96 [b]	7.10 ± 1.70 [f]	4.00 ± 0.35 [g]

Table 4.13 Colour parameters of crumb rubber for two-stage continuous vacuum convective drying

		Continuous drying			
Vacuum Duration	Heating Temperature	L*	a*	b*	ΔE
HAD	90 (°C)	27.07 ± 0.60 [b]	6.33 ± 1.46* [f]	13.67 ± 1.76 [g]	12.75 [h]
VD	90 (°C)	25.50 ± 2.95 [b]	3.07 ± 1.53* [e]	9.53 ± 2.67 [g]	9.93 [h]
V120*	90 (°C)	11.00 ± 3.59 [c]	3.70 ± 1.99* [e]	6.70 ± 5.42 [g]	9.01 [h]
	110 (°C)	11.80 ± 0.78 [c]	3.00 ± 0.68* [e]	5.00 ± 1.04 [g]	8.26 [h]
	130 (°C)	11.30 ± 7.76 [c]	4.60 ± 1.18* [e]	6.80 ± 5.37 [g]	8.48 [h]
	150 (°C)	15.40 ± 2.82 [c]	4.30 ± 1.07* [e]	11.20 ± 2.30 [g]	8.48 [h]
V60*	90 (°C)	8.40 ± 2.33 [d]	5.30 ± 4.10* [f]	8.60 ± 2.19 [g]	11.60 [h]
	110 (°C)	8.40 ± 1.53 [d]	2.80 ± 1.94* [e]	1.10 ± 0.31 [g]	11.71 [h]
	130 (°C)	9.90 ± 3.22 [d]	3.30 ± 1.41* [e]	7.60 ± 4.59 [g]	10.41 [h]
	150 (°C)	10.10 ± 2.88 [d]	3.70 ± 2.46* [e]	6.40 ± 2.64 [g]	9.73 [h]
V30*	90 (°C)	5.80 ± 3.61 [d]	7.40 ± 1.37* [f]	6.60 ± 0.95 [g]	13.36 [h]
	110 (°C)	6.90 ± 2.40 [d]	3.00 ± 1.37* [e]	7.50 ± 4.56 [g]	13.16 [h]
	130 (°C)	8.00 ± 5.74 [d]	3.80 ± 1.80* [e]	6.40 ± 4.84 [g]	11.64 [h]
	150 (°C)	9.20 ± 4.07 [d]	4.80 ± 1.24* [e]	2.50 ± 3.33 [g]	10.08 [h]

Anova analysis shows there is statistical significant influence of vacuum duration and heating temperature to L, a* and b* values, $p<0.05$.

Based on Table 4.12, the fresh rubber had high lightness (L*) value, redness (+a*) and yellowness (+b*) while the standard rubber colour had a 62.6% lower L* value, 330% higher a* value and 70% lower b* value, compared to the fresh rubber. A lower total colour change value indicated less variation with the standard rubber colour. The existing drying process changed all three colour parameters (L*, a*, b*), causing a colour shift towards the darker region, which the L* decreased, a* increased and b* decreased due to the browning effect. Gupta et al. (2013) studied the effect of sample size, heating temperature and air flow velocities to the drying time of cauliflower, which the colour change of the dried cauliflower was affected by the heating temperature and air flow velocities, but independent of the sample size. Similarly, Omolola (2015) reported that drying time affected both the colour change of dried banana. Thus, the overall colour change result for rubber drying at various heating temperatures and sizes was in accordance to the literature.

A comparison across the same heating temperature at 90°C in Table 4.13 showed that 120 min vacuum duration (V120) gave a 22.33% and 32.56% lower colour change value compared to 60 min (V60) and 30 min (V30) vacuum duration. Statistical analysis using Anova showed that a change in vacuum duration had significant effect to the colour change, $p<0.01$. The colour change of rubber was dependent to oxygen exposure, heat exposure and drying time. Oxidation can be retarded by drying the object in an oxygen-free environment (Shashoua, 1993). A longer vacuum drying process helps to reduce oxygen exposure, but there was still Maillard reaction in crumb rubber, which caused visible browning during vacuum drying. Nevertheless, the two-stage continuous VCD with 60 and 120 min vacuum pre-drying duration had a lower colour change, ΔE compared to conventional drying method. When the rubber sample was dried via long vacuum duration (V120), coupled with hot air drying temperature of 110°C (HAD110), the final colour became closer to standard rubber colour, with the total colour change, ΔE as 8.26. The L*, a* and b* was 56.4%, 52.60% and 36.58%, lower than the colour parameters of hot air dried rubber, respectively, with a shorter drying time of 9.09%. Heating temperature also have a significant effect to rubber drying process, $p<0.002$. There was a significant colour improvement with the use of two-stage vacuum convective drying (V120-HAD110) subjected to 120 min vacuum pre-drying (V120) and heating temperature at 110°C (HAD110), in continuous basis. In short, the total colour change, ΔE for two-stage continuous VCD of rubber can be lower with increasing heating temperature (from 90°C to 150°C) and increasing vacuum duration (from 30 min to 120 min).

Table 4.14 Colour parameters of crumb rubber for two-stage intermittent vacuum convective drying

Intermittent drying					
Vacuum Duration	Heating Temperature	L*	a*	b*	ΔE
HAD	90 (°C)	42.67 ± 2.56 [a]	3.83 ± 1.40* [e]	18.77 ± 3.57 [g]	28.24 [i]
VD	90 (°C)	22.35 ± 8.41 [b]	5.80 ± 0.57* [f]	14.20 ± 2.40 [g]	12.08 [j]
V120*	90 (°C)	5.50 ± 1.72 [d]	3.10 ± 1.47* [e]	4.10 ± 1.98 [h]	13.98 [j]
	110 (°C)	9.10 ± 3.27 [d]	1.80 ± 0.50* [e]	9.10 ± 4.21 [h]	12.25 [i]
	130 (°C)	15.00 ± 1.63 [c]	1.10 ± 2.25* [e]	12.40 ± 4.36 [g]	11.03 [j]
	150 (°C)	29.90 ± 4.31 [b]	8.30 ± 1.35* [f]	17.70 ± 3.25 [g]	17.61 [j]
V60*	90 (°C)	5.10 ± 2.90 [d]	3.00 ± 1.12* [e]	5.00 ± 2.48 [h]	14.43 [j]
	110 (°C)	7.30 ± 0.93 [d]	4.80 ± 3.06* [e]	6.60 ± 4.71 [h]	12.11 [i]
	130 (°C)	10.50 ± 1.59 [c]	7.70 ± 2.63* [f]	11.20 ± 3.12 [g]	11.08 [j]
	150 (°C)	21.20 ± 3.92 [b]	7.40 ± 1.77* [f]	14.80 ± 5.37 [g]	11.05 [j]
V30*	90 (°C)	3.90 ± 0.73 [d]	1.70 ± 0.65* [e]	2.20 ± 0.98 [h]	16.04 [i]
	110 (°C)	5.70 ± 2.95 [d]	1.80 ± 1.76* [e]	4.00 ± 2.26 [h]	14.22 [j]
	130 (°C)	7.00 ± 0.91 [d]	2.00 ± 1.54* [e]	5.00 ± 1.01 [h]	12.99 [j]
	150 (°C)	15.60 ± 0.25 [c]	5.70 ± 0.55* [f]	13.00 ± 0.61 [g]	9.69 [i]

Anova analysis shows there is statistical significant influence of vacuum duration and heating temperature to L, a* and b* values, $p<0.05$.

Based on Table 4.14, the total colour change, ΔE of rubber subjected to two-stage intermittent VCD decreased with increasing vacuum duration (from 30 min to 120 min). The decreasing trend was even more obvious at higher drying temperature, 150°C (HAD150). The colour change for rubber that was 30 min vacuum pre-dried in the two-stage intermittent VCD with HAD150 for was 44.97% and 12.31% lower than vacuum pre-drying for 120 and 60 min, respectively. This lower colour change was because of the overall short drying time when the rubber samples were dried quickly with high intermittent temperature during the second stage VCD. The colour change for samples dried with short drying time was normally low (Zhu, 2009). In addition, the heat was stopped for 5 min after every 15 min heating during the intermittent drying. The surface temperature of samples during this tempering period were in equilibrium with the surrounding temperature, as there was residual heat inside the heating chamber. When the temperature gradient was reduced, the moisture diffusivity increased and hence, reduced the total colour change.

Based on Table 4.14, the total colour change, ΔE of rubber subjected to two-stage intermittent VCD also decreased with increasing heating temperature (from 90°C to 150°C). The drying at 150°C had the highest total colour change value for vacuum pre-drying of 120 min during the two-stage intermittent VCD. The total colour change at drying temperature of 150°C was 59.66%, 43.79% and 25.97% higher than the total colour change of crumb rubber samples at drying temperature of 130, 110 and 90°C during intermittent VCD drying, respectively. The main reason of this adverse colour change was due to the prolonged drying time. Formation of rubber skin at the early drying stage reduce the moisture diffusivity from the core to surface (Rahman, 1985). Hence, the prolonged drying period lead to a high total colour change.

Rubber with high heat exposure for prolonged period tends to have a higher colour change value. High temperature drying does not necessarily lead to high total colour change, but longer drying duration will change the rubber colour severely. Intermittent drying at high temperature was one of the way to prevent severe browning while obtain the high drying efficiency of products. The total colour change and the drying kinetics for rubber dried under intermittent drying was much higher than continuous two-stage vacuum convective drying, with a short effective drying time. Nadian (2015) reported on the controlling of drying process with cost-effective colour measuring system to improve apple slices colour change. Likewise, a study on edible bird's nest showed that the intermittent drying of heat pump was able to minimize colour change (Gan, 2017). In this study, the time taken for the completion of rubber drying through intermittent process reduced by 16.67%, 0%, 25% and 13.33% for the second stage hot air drying temperature at 150, 130, 110 and 90°C, respectively. It was an improvement on the corresponding continuous drying runs. This showed that intermittent high temperature drying could be a good alternative to continuous drying of crumb rubber.

On top of that, the rubber subjected to two-stage vacuum convective drying (VCD) with 30 min vacuum pre-drying duration and hot air drying temperature of 150°C (V30-HAD150) had a golden yellow or light brown colour. The L*, a* and b* values for V30-HAD150 were high, amounting to 15.60 ± 0.25, 5.70 ± 0.55 and 13.00 ± 0.61, respectively. Based on the comparison of Table 4.13 and 4.14, the ΔE of intermittent V30-HAD150 was 3.87% lower than the continuous counterpart and 17.31% higher than continuous VCD, V120-HAD110. However, the drying of 50mm rubber via continuous two-stage V120-110 took 300 min compared to intermittent two-stage V30-HAD150 that only required 90 min. The V30-HAD150 only required 30% of the total drying time used by V120-110 to achieve similar colour value. Hence, V30-HAD150 was a preferable option for quick surface moisture removal and drying rate improvement in rubber drying process. It was observed that by using an appropriate heating temperature and cycle time, it was possible to reduce significantly the drying time to reach the desired moisture content with improved product colour. Below image shows the colour change of rubber for various drying temperature.

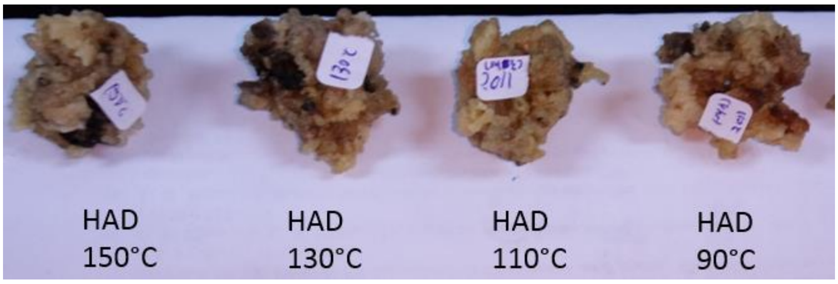


Figure 4.21 Colour change for rubber dried at various heating temperature

The continuous drying process changed all three colour parameters (L*, a*, b*), causing a colour shift towards the darker region, which the L* decreased, a* increased and b* decreased due to the browning effect. By using intermittent drying to replace continuous drying of crumb rubber, acceptable colour quality of the sample at the required final moisture content was not yet achieved. Thus, the experiments were continued by employing a step change in drying air temperature with time-varying strategies.

4.3.5.2 Texture analysis

Textural properties, mainly hardness (H) and stickiness (S) was evaluated with a TA-XTPlus Texture Analyser (Stable Micro Systems, Co., UK) through compression test. The texture characteristic measured represents the physical changes of crumb rubber during the drying process. These textural changes were determined by measuring the toughness and adhesiveness between interfaces, i.e. the surface between dried rubber and probe. Many studies had been focusing on the effect of drying techniques to the dried rubber properties, but more substantial knowledge was required to understand the influence of drying process on the structure property relationships and drying properties of rubber. The statistical analysis (Anova) results revealed a non-significant trend for the texture properties of dried crumb rubber samples subjected to various drying conditions, including two-stage vacuum convective drying (VCD) with various vacuum duration (30, 60 and 90 min), followed by hot air drying (HAD) at various heating temperature (90, 110, 130 and 150°C). The effect of drying conditions, including vacuum drying duration and heating temperature on texture properties of crumb rubber were examined for both continuous and intermittent drying and shown in Figure 4.22a, b, c, d.

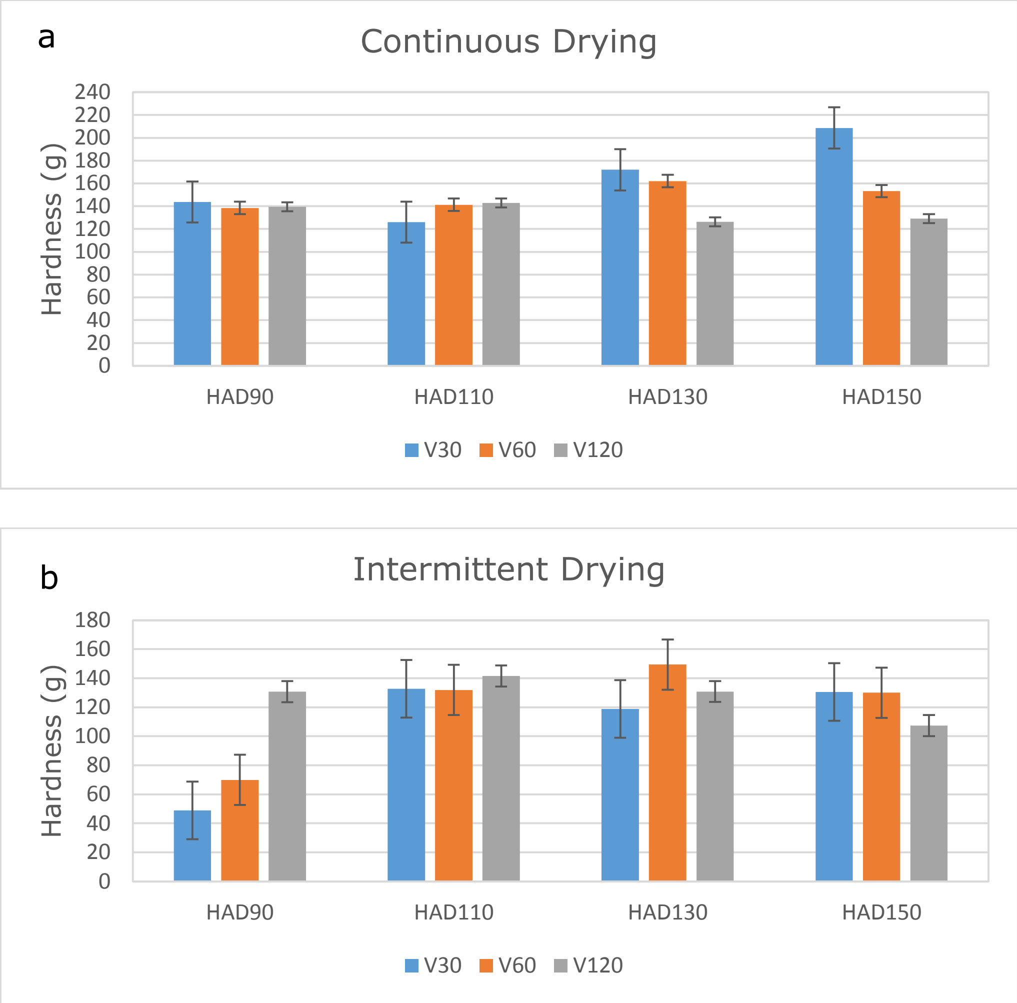


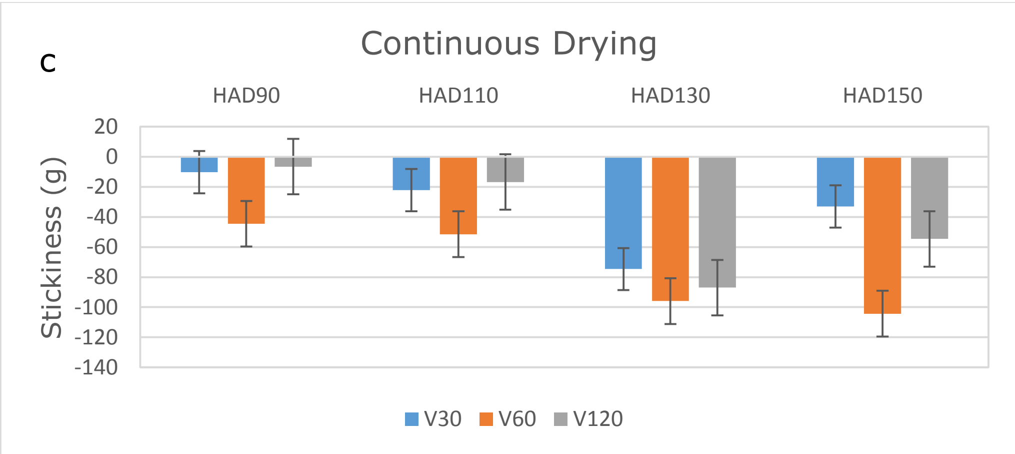
Figure 4.22a, b: Texture properties (hardness) of dried crumb rubber samples subjected to two-stage vacuum convective drying (VCD) with various vacuum duration (30, 60 and 90 min), followed by hot air drying (HAD) at various heating temperature (90, 110, 130 and 150°C)

As shown in Figure 4.22a, the effect of vacuum drying to the hardness of rubber was more significant at high hot air drying temperature. At higher hot air drying temperature, 150°C (HAD150) with a lower vacuum pre-drying period, 30 min (V30) would lead to a harder rubber material. The hardness of rubber subjected to two-stage drying of V30-HAD150 was 45.18% higher than rubber dried at lower temperature, 90°C

(HAD90) while the longer vacuum pre-drying duration, 120 min (V120) coupled with hot air drying temperature, 150°C (HAD150) was 38.1% softer than V30-HAD150, for continuous drying.

For the intermittent counterpart in Figure 4.22b, the lowest hardness was found on rubber subjected to V30 and HAD90. The intermittent V30-HAD90 was only 48.84 g, which was 76.6% softer than the continuous V30-HAD150. This was due to the low heating temperature, which prevented the formation of rubber skin. The hardness value ranged from 48.84 g to 208.73 g for both continuous and intermittent VCD, with the critical hardness values fall on 183.45g. Any samples with hardness higher than this critical hardness values would have experience structure changes. There was a total of 87.5% experiments carried out during intermittent drying was softer than continuous drying. This was a favourable phenomenon as the rubber processor prefer rubber with their natural tackiness and softness. Thus, intermittent drying should be used for the two-stage drying process.

The average hardness values for continuous VCD drying and intermittent VCD drying were 148.64g and 118.54g, respectively while the average stickiness values were -50.06 g and -35.24 g, respectively. The rubber dried with two-stage VCD had a softer texture and higher stickiness compared to standard rubber. The continuous V30-HAD150 with highest hardness value of 208.73 g also had a low stickiness value of -32.94 g, which was 3.09 times higher than the standard rubber stickiness. Similarly, the hardness and stickiness value of V30-HAD150 intermittent counterpart was 28.9% and 173.3% higher than standard rubber hardness.



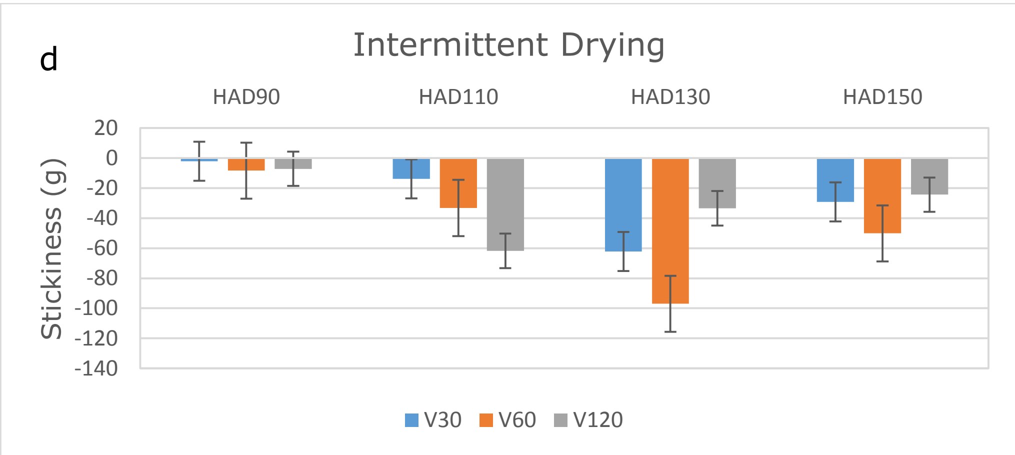


Figure 4.22c, d: Texture properties (stickiness) of dried crumb rubber samples subjected to two-stage vacuum convective drying (VCD) with various vacuum duration (30, 60 and 90 min), followed by hot air drying (HAD) at various heating temperature (90, 110, 130 and 150˚C)

Prior drying, crumb rubber is a wet, discrete solid. However, the state of material changed when it is exposed to high heat. Typically, prolonged drying of rubber at low drying temperature gave a less sticky texture and a harder rubber due to oxidative hardening effect when the new crosslinks were formed. Rubber has a type of diene elastomers that have electron-donating groups attached to the diene, so it was usually unstable to heat and have poorer heat resistance (Rahman, 1985).

From Figure 4.22c, d, the rubber achieved maximum stickiness for vacuum pre-drying of 60 min (V60) coupled with hot air drying temperature of 150°C (HAD150), for continuous drying while the minimum stickiness was found in intermittent V30-HAD90, with a value of -2.2 g. The stickiness level of V60-HAD150 and V30-HAD90 were 9.79 times higher than and 4.84 times lower than standard rubber. The main reason for the high stickiness could be due to prolonged drying at high heating temperature, inversely, the low stickiness was due to prolonged drying at low hot air drying temperature. All stickiness value for both continuous and intermittent drying of two-stage VCD with second stage hot air temperature at 90°C (HAD90) ranged from -2.2 to -44.51. The low stickiness value was accompanied by average hardness value of 111.89 g. The stickiness value was not well correlated to the rubber hardness, as there was no clear trend in both stickiness and hardness values. The lack of precision in determination of the effect of varying operation parameters to both hardness and stickiness could be due to the inconsistent change of rubber texture when exposed to heat. Generally, low temperature drying of rubber will have low stickiness value while high temperature drying of rubber will have high stickiness value.

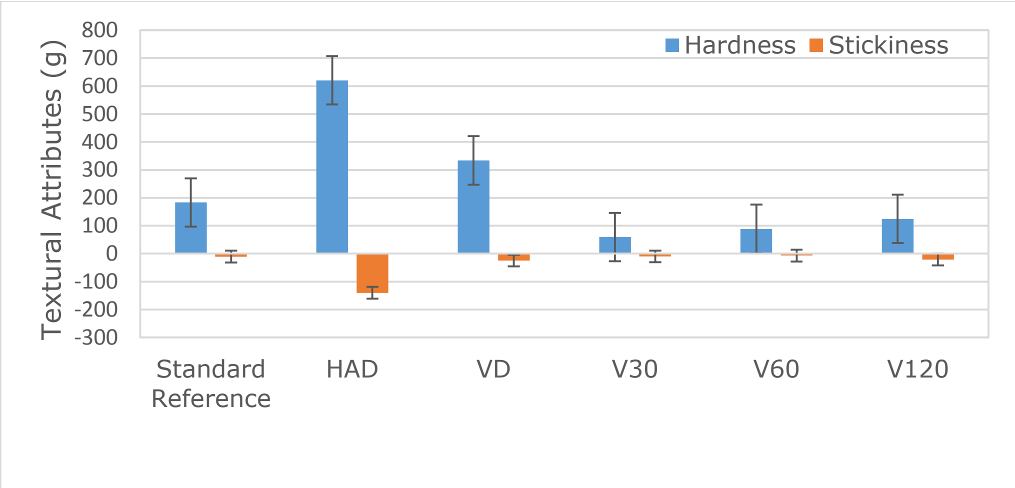


Figure 4.22e: Texture properties (hardness and stickiness) of dried crumb rubber samples subjected to various drying methods

Drying of crumb rubber tends to lead to a deformation of rubber structure, which reduce the porosity of rubber. At high heating temperature, the high heat stress lead to the collapse of rubber structure. The shrinkage due to rupture of rubber structure lead to a more compact and harder rubber material. In addition, when the moisture was removed rapidly from the materials, the rubber shrinks and turned sticky. Thus, the final dried rubber was commonly harder and stickier compared to the raw rubber.

From Figure 4.22e, the hot air dried rubber had a hardness level of 620.69 g, which was almost 85.91% higher than vacuum dried rubber with a hardness level of 333.85 g. A comparison of the lab dried rubber with factory products, the standard rubber, showed that hot air dried rubber was 70.44 % harder. Similarly, vacuum dried rubber also showed a higher hardness value compared to standard rubber, 45.05% harder. The longer the vacuum pre-drying duration, the higher the hardness and stickiness level of rubber. Thus, this showed that two-stage vacuum convective drying would be able to produce rubber with higher softness.

In present study, three different vacuum durations for first stage vacuum drying, four different heating temperatures for second stage convective drying and four sample diameter were investigated simultaneously. Statistical analysis (analysis of variance, ANOVA) with the use of response optimizer showed that the intermittency strategies and hot air drying temperature had statistically significant influence on only stickiness values. The regression equation 3.4 and 3.5 found for hardness and stickiness in relation to the intermittent ratio (IR) and heating temperature (T) is as below:

Hardness = 135.2 – 27.5 (IR0.75) + 27.5 (IR1) – 38.9 (T90) – 5.8 (T110) + 10.2 (T130) + 34.4 (T150) (3.4)

Stickiness = 30.91 + 4.055 (IR0.75) – 4.055 (IR1) + 24.74 (T90) + 12.88 (T110) - 37.5 (T130) – 0.12 (T150) (3.5)

It was observed that by using a good combination of vacuum pre-drying duration, hot air heating temperature and intermittent drying strategy, it was possible to reduce significantly the drying time to reach the desired moisture content with improved product quality. From the results, vacuum pre-drying for 30 min (V30) and hot air drying at high temperature at 150°C (HAD150) showed a desirable drying characteristic, with high drying rate for 50mm, and acceptable colour and textural changes. The V30-HAD150 was selected based on drying rate as the quality was within acceptable range.

Figure 4.23 shows the optimization plot for the textural change due to intermittent ratio and heating temperature. The optimization plot indicates that intermittency strategies did improve the textural properties. However, when the heating temperature was increased, the desirability reduced. Nevertheless, stepwise drying was able to solve the undesirable effects as the drying temperature was reduced prior to the occurrence of any quality changes.

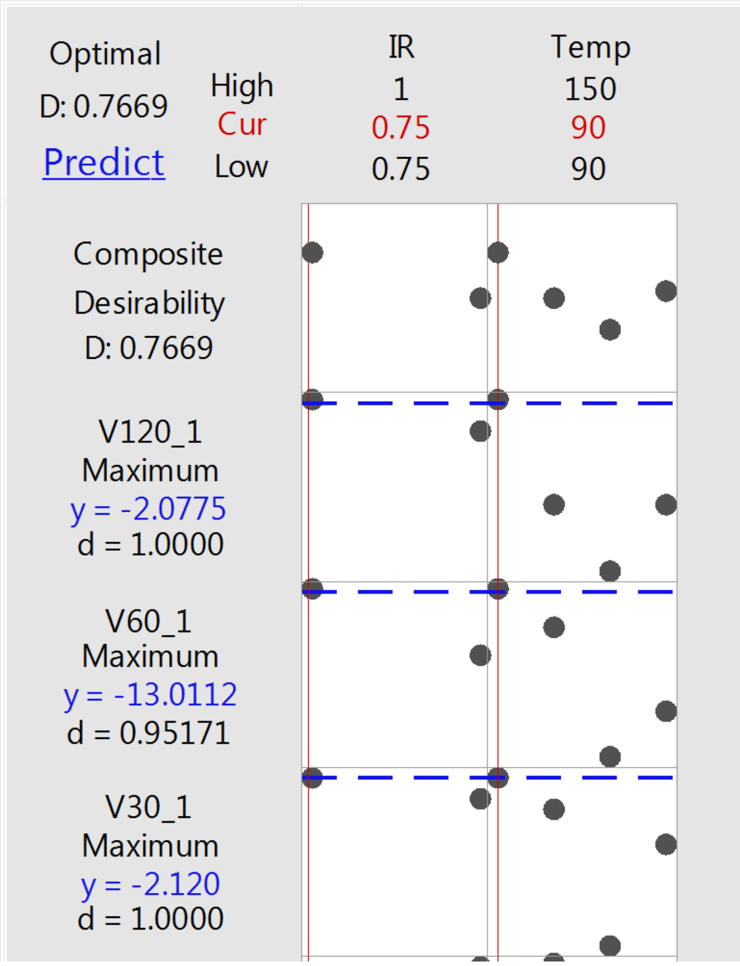


Figure 4.23: The optimization plot for textural changes

To conclude the findings, a rubber dried with two-stage VCD, with a shorter vacuum duration, a higher heating temperature and a smaller size rubber tends to gain higher hardness and stickiness. Hence, it was worth further investigate the stepwise profile of vacuum convective drying (VCD), with HAD150 as the starting temperature to further improve the colour and texture attributes. The stepwise changes in drying conditions were able to minimize the total energy requirement.

4.4 Stepwise profile of two-stage high temperature intermittent Vacuum Convective Drying (VCD)

In this study, multiple responses, such as low final moisture content, high drying rate, low total colour change (ΔE), low hardness and high stickiness were desirable. Two parameters (vacuum pre-drying duration and sample diameter) tested in earlier experiments were fixed during the subsequent study of the two-stage vacuum convective drying (VCD) while hot air drying (HAD) temperature was adaptable to the stepwise drying process.

In this study, the effect of stepwise change (step-up and step-down) of heating temperature during two – stage intermittent vacuum convective drying (VCD) was determined. By using time-varying stepwise temperature profile coupled with the intermittent drying process, acceptable colour quality and texture properties of the sample at the required final moisture content was possible. The stepwise profile based on the right changing point could preserve the colour and texture quality of products (Pei, 2014). Changing point for the temperature is one of the important aspects that prevent product quality deterioration and prolonged drying time, which normally determined by the moisture content of sample. By selecting the optimum intermediate moisture content as critical changing point, drying time can be reduced (Jomlapelatikul, 2016). Every point of change aimed to maintain the high drying rate and reduce the quality degradation of samples. Thus, the critical moisture content was determined by recording the onset of browning time during intermittent drying.

4.4.1 Onset browning time

Table 4.15 shows the onset of the browning time results for intermittent drying. In most applications, moisture content is expressed on a wet basis, which indicated the water mass in grams per 100 grams of the total mass. The %wet basis (w.b.) moisture content of rubber was calculated from the rubber weight measured for stepwise profile during two-stage intermittent VCD drying. For simplification and the comparison purpose, both wet basis and dry basis were used for calculations and comparison to identify the browning time.

The onset of browning time results was evaluated to determine the point of change based on the right combination of vacuum pre-drying duration and hot air drying (HAD) temperature. The point where it began to turn into an unacceptable browning or blackening via L*a*b* colour measurement was recorded as onset of browning time.

Table 4.15: Onset of browning during two-stage intermittent vacuum convective drying

Rubber Samples	Drying Temperature / ºC	Moisture Content /% w.b.	Moisture Content /% d.b.	Browning Time / min
V120	HAD90	7.58 [a]	11.22 [b]	360 [c]
	HAD110	7.68 [a]	11.36 [b]	300 [c]
	HAD130	7.64 [a]	11.30 [b]	240 [c]
	HAD150	7.71 [a]	11.41 [b]	180 [d]
V60	HAD90	7.24 [a]	10.71 [b]	330 [c]
	HAD110	6.46 [a]	9.56 [b]	270 [c]
	HAD130	6.84 [a]	10.12 [b]	180 [d]
	HAD150	7.85 [a]	11.61 [b]	140 [d]
V30	HAD90	8.06 [a]	11.93 [b]	330 [c]
	HAD110	8.30 [a]	12.28 [b]	240 [c]
	HAD130	7.47 [a]	11.05 [b]	130 [d]
	HAD150	6.84 [a]	10.12 [b]	90 [d]

Note: Means in the same column that do not share subscripts differ at $p < 0.05$ probability level.

From Table 4.15, the onset browning time for vacuum pre-drying duration of 120 min (V120) was slower than shorter vacuum pre-drying duration, 30 min (V30) and 60 min (V60). During the vacuum pre-drying, rubber samples were not exposed to oxygen. Hence, a longer vacuum pre-drying will lead to a delay in colour degradation. For example, the onset of browning time and moisture content % (w.b.) for V120-HAD90 was 9% and 5.96% higher than V30-HAD90 while V30-HAD150 was 72.72% and 15.13% lower than V30-HAD90. This showed that the effect of drying temperature was more significant compared to the effect of vacuum duration. When the heating temperature increased, the total drying time to reach the final moisture content was shorter, which the reduced heat exposure duration was able to improve rubber quality. By applying high temperature drying at reasonable time frame, a low total colour change (ΔE) values to dried rubber samples was possible.

Several researchers have reported the advantage of using this technique to yield better colour quality (Fakuiotat et al., 2008; Cuervoa et al., 2008; Chua et al., 2001). Fakuiotat et al. (2008) stated step-wise changes from high heating temperature to lower drying temperature, especially at the final stage would be able to avoid severe browning of tomato samples. Similarly, Chua et al. (2001) had reported a better colour quality for the banana samples through step-down temperature profile, even though both step-up and step-down of the temperature profile can reduce colour degradation. A study comparing the drying of carrot via step-up temperature 60°C/ 70°C and step-down temperature 70°C/ 60°C showed that step-up mode gave the shortest drying time (Jomlapelatikul et al., 2016). Whereas, a stepwise profile of lemon balm with step-up temperature 40°C/ 50°C degraded the herb quality (Cuervoa et al., 2016; Cuervoa et al., 2008).

However, the stepwise drying process was limited to 50mm thick rubber sample, with first stage vacuum drying of 30 min (V30) and second stage convective drying of 150°C (HAD150), 130°C (HAD130) and 110°C (HAD110) as the main drying temperature. Drying kinetics of crumb rubber were not required to predict the drying behaviour, for stepwise profile of drying parameters, as each individual parameter was evaluated earlier. The main reason for current selection was the high effective moisture diffusivity compared to other drying conditions and sample diameter. The large surface area per unit volume enable the heat transfer to the rubber samples, which further promote the internal moisture diffusion. Even though a longer vacuum pre-drying duration, 120 min (V120) gave a better colour quality compared to vacuum pre-drying of 30 min (V30), but V120 prolonged the total drying time. Thus, the point of change was determined based on the critical moisture content and onset of browning time recorded for V30.

Referring to Table 4.15, the rubber turned brown 72.7% faster when vacuum pre-drying of 30 min (V30) was coupled with hot air drying temperature at 150°C (HAD150), compared to 90°C (HAD90). This result was as expected as a higher heating temperature will lead to higher colour change and shorter browning time. When the heating temperature undergoes stepwise profile before the onset browning time, the colour quality of crumb rubber can be preserved. At the same time, the total drying time can be shortened. Thus, the heating temperature for second stage vacuum convective drying during the stepwise profile was preferable to be 150°C for a duration of 90 min or less, to minimize the total drying time, maintain the high drying rate without any degradation to the rubber properties. The analysis of variance for onset browning time shows that the p-value is < 0.004, which the differences of the browning time in relation to drying temperature is significant.

Figure 4.24 below shows the three change points for the time-varying stepwise drying profile. The stepwise profile was designed to reduce the colour quality degradation, when high temperature was used for rubber drying. The initial moisture content of crumb rubber for this study ranged between 33.15 - 44.47% (d.b.). The two-stage vacuum – convective drying began with 30 min vacuum drying (V30) at 90°C, then step-up to 150°C via intermittent hot air drying (HAD150).

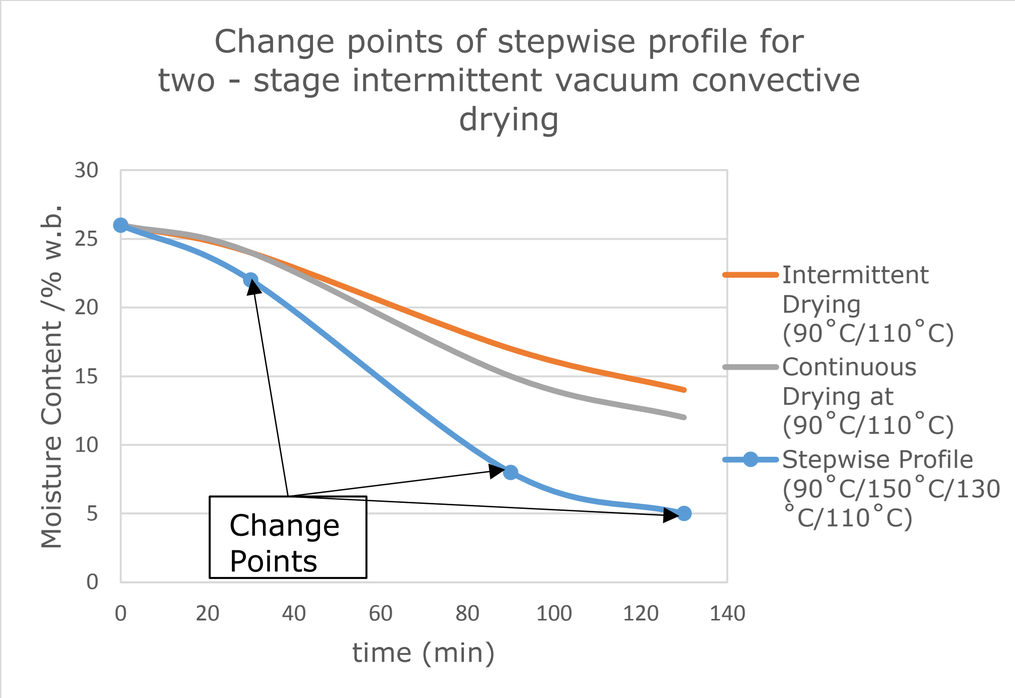


Figure 4.24: Change points for stepwise temperature profile of two stage intermittent vacuum convective drying (VCD)

The HAD150 was stopped after 60 min, before the moisture content decreased lower than 8% (w.b.). As shown in Table 4.15, browning began at the moisture content below 6.84% (w.b.) when the rubber was dried with intermittent VCD at 150˚C. Therefore, the reduction of heating temperature (step down profile) to 130˚C could prevent browning and minimize the total drying time. The HAD130 was continued for 40 min until the moisture content reached 5% (w.b.). The rest of the drying was performed by further step down the intermittent drying temperature to 110 ºC for complete dryness of crumb rubber. Table 4.16 below is the drying parameters at different stage of the optimized process.

Table 4.16: Drying parameters at different stage of the optimized process

Drying temperature / °C	Effective drying time / min	Overall drying time / min	Moisture reduction to / %w.b.
Vacuum pre-drying at 90°C (V90)	30	30	30
HAD150	45	60	8
HAD130	30	40	5
HAD110	75	25	1

4.4.2 Application of stepwise profile to two-stage intermittent Vacuum Convective Drying (VCD)

This work indicates that stepwise profile of VCD, especially controlling of the heating air temperature can improve the rubber drying rate, in term of reducing the total drying time. In comparison with results obtained under traditional operation conduct with inlet air in steady-state condition or intermittent drying, this new method of drying able to dry rubber up to the manufacturing standard, referring to the standard rubber quality and industrial specification. The moisture evaporated during each stage of stepwise drying process, including standard deviation, is shown in Figure 4.25.

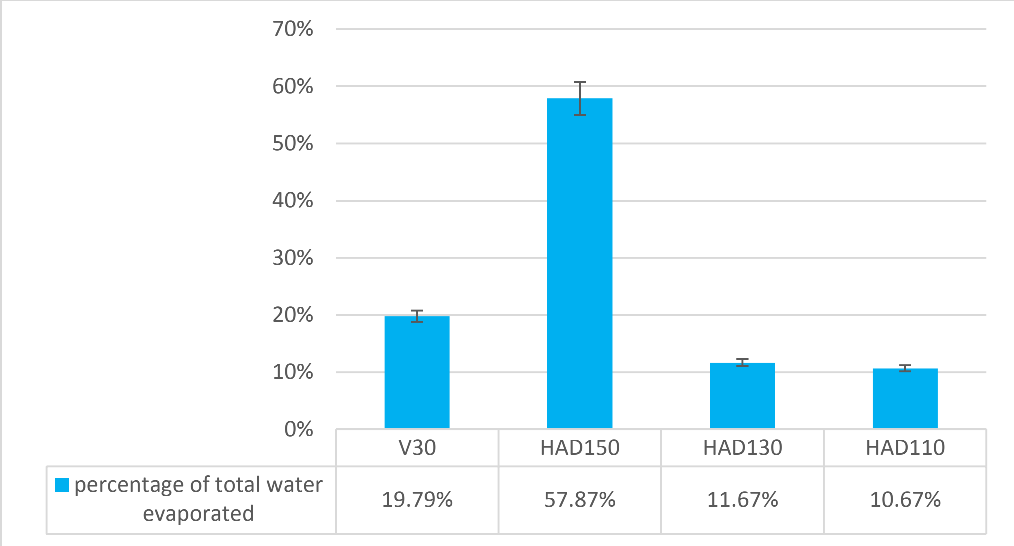


Figure 4.25: Moisture evaporation for stepwise drying

Based on Figure 4.25, the initial 30 min of vacuum drying removed the 19.79% of moisture, while majority of the moisture, 57.87% was removed during the intermittent drying of hot air drying (HAD150). The high heating temperature provided sufficient latent heat required for evaporation, hence, the drying rate and effective moisture diffusivity increased during this stage. The drying rate was maintained during the tempering period as the rubber material was sufficiently hot to supply the latent heat required for the moisture evaporation. Thus, the drying was continuous even at the absence of external heat source. The subsequent step-down drying at 130ºC (HAD130) removed the remaining 11.67% of moisture to reach 5% (d.b.), while the final drying stage for HAD110 took additional 80 min to remove the remaining 10.67% of moisture.

Aside from moisture changes, monitoring of the temperature distribution of time varying stepwise temperature profile allowed a better understanding of quality changes that occurred during the optimization studies. The temperature at two locations in the rubber samples during drying was recorded with infrared thermometer, by measuring the surface temperature and internal rubber temperature, as shown in Figure 4.26.

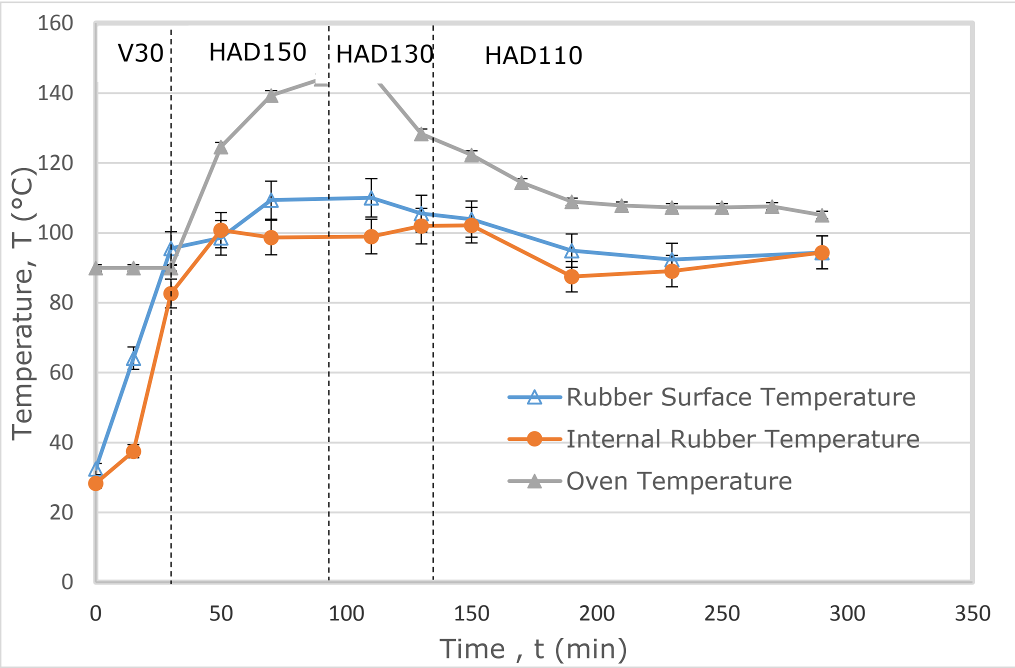


Figure 4.26: Temperature development of 50mm thick rubber during stepwise drying

Figure 4.26 shows a quick increment of rubber temperature, for both surface and internal temperature reading for the initial 30 min of vacuum pre-drying period. The highest temperature differences for the surface and internal rubber recorded was 26.6°C at the initial transient period. The temperature gradient within rubber became lesser after 50 min of drying time. As the oven temperature increased to 145.6°C, the maximum temperature recorded for the surface and internal rubber was 110 and 101.93°C, after 110 min of drying time. At this point, the step-down of temperature from 150 to 130°C began and the rubber temperature was further reduced the end of drying process. By understanding the relationship of moisture change with the temperature development in the rubber core, the fundamental drying mechanism of rubber could be understood better. As shown in Figure 4.26, there was a drastic moisture changes with increasing rubber temperature. As the internal rubber temperature increased for 72.43°C, there was 18.89% of moisture left in the rubber samples. A majority of the moisture were eliminated during the increasing temperature period. This shows that most of the moisture was not bound to the rubber while the remaining bound moisture took considerable longer time to dry. The remaining moisture was dried within 100 min as the rubber temperature was maintained within 98.67 to 102.20°C. As the high temperature intermittent drying period began, the temperature increment became slower. The slower increment in temperature also ensure that the rubber would not undergone extreme heat stress while able to enjoy the high drying rate. To further understand the moisture ratio and drying rate changes during the stepwise drying process, moisture ratio over drying time chart and drying rate over drying time chart was shown in Figure 4.27, included standard-deviation bars. Both figures are a variation of moisture ratio and drying rate of 50mm rubber with respect to drying time for stepwise profile, continuous drying and intermittent drying; x-axis includes the tempering periods.

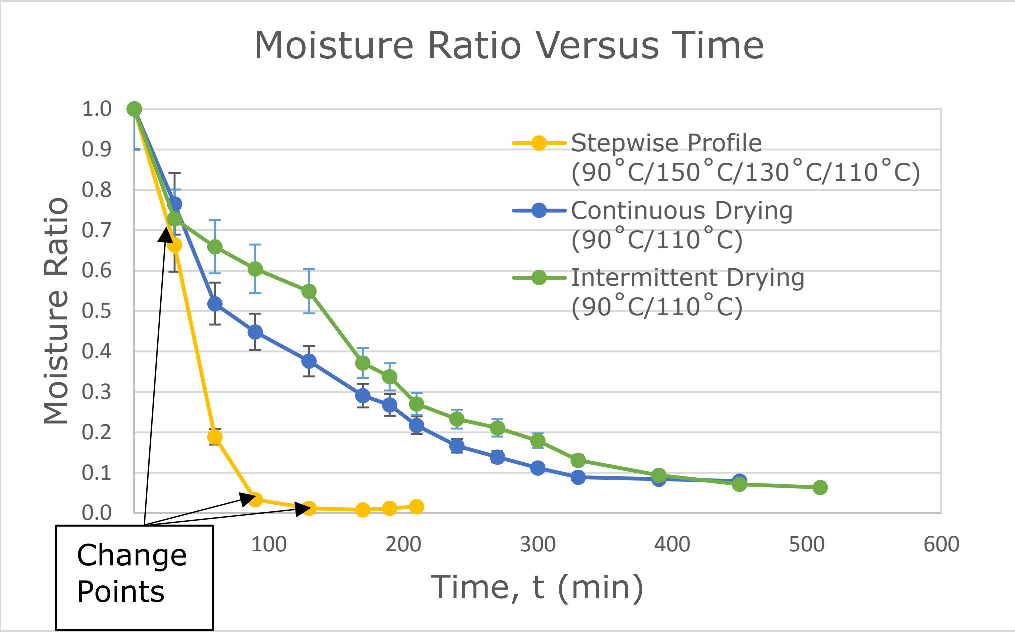


Figure 4.27: Moisture ratio versus time for stepwise profile and constant profile of continuous and intermittent drying

Figure 4.27 shows that all the drying curves for stepwise drying, continuous drying and intermittent drying are exponential. The moisture content of rubber decreased with time. For the two-stage vacuum convective drying (VCD), the moisture content of the samples could be successfully reduced from initially 33.15 - 44.47% (d.b.) to lower than 0.2% (db.) within 210 min. Compared to the continuous drying and intermittent drying of rubber samples via two-stage vacuum convective drying (VCD), 53.3% and 58.8% of the total drying time were saved. This showed that the stepwise drying was a possible solution for improving the rubber drying process. The gradient of curve in Figure 4.27 indicates the drying rate. The drying rate curves for stepwise drying and two-stage continuous VCD shows three drying periods, which are initial transient period, increasing rate period and falling rate period. Whereas, there is only falling rate period for the two-stage intermittent VCD. The step-up temperature from vacuum drying at 90°C to 150°C increased the drying rate to a maximum of 0.00308 g H_2O min^{-1}, which the value was higher than two-stage continuous VCD and intermittent VCD. For better understanding of rubber drying characteristics during stepwise drying, drying rate plotted versus moisture ratio in Figure 4.28.

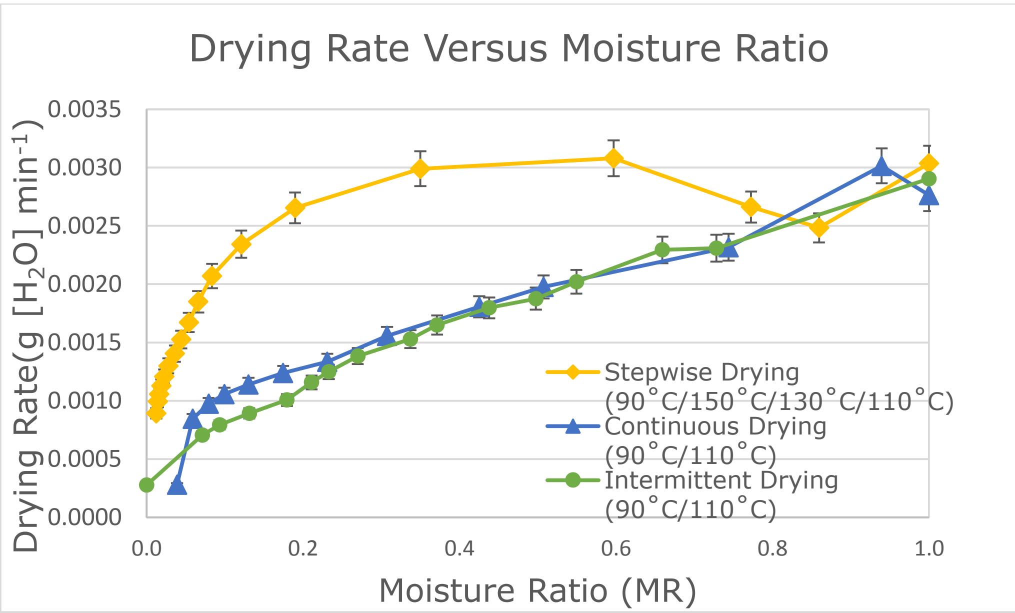


Figure 4.28: Drying rate versus moisture ratio for stepwise drying, continuous drying and intermittent drying

Based on Figure 4.28, there was a deflection point for stepwise drying rate curve, which clearly representing two drying stages. There was a great increment of drying rate until the moisture ratio reach 0.35. The higher drying rates during the time-varying step-down drying of the second stage of vacuum convective drying showed that the moisture was rapidly removed from the rubber. In contrast, the continuous drying and intermittent drying undergone falling rate period. The drying rate was decreased with decreasing of moisture content, especially when the moisture of the rubber was less than 10%. It was found that the final moisture content of samples dried by stepwise drying, continuous drying and intermittent drying were 0.0009, 0.0003 and 0.0003 $gH_2O\ gDM^{-1}$, respectively. The effective moisture diffusivity for stepwise was determined by Fick's second law of diffusion, by measuring the slope from Figure 4.29. From the calculation, the total effective moisture diffusivity, D_{eff} values for continuous drying were 3.2372 x 10^{-9}, with D_{eff} values of 1.1399 x 10^{-9} for vacuum pre-drying and D_{eff} values of 2.0973 x 10^{-9} for hot air drying.

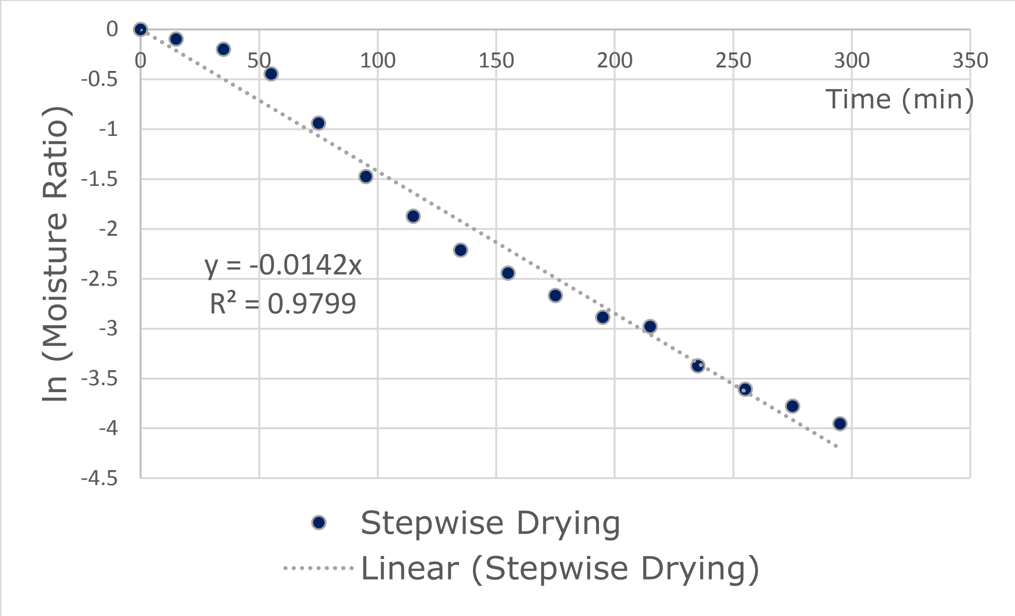


Figure 4.29: Drying curve of the natural logarithm of moisture ratio versus time for stepwise drying of crumb rubber

This stepwise temperature profile leads to high effective moisture diffusivity and high drying rate. As drying progressed, there was increasingly more air in the space previously occupied by water, which improve the moisture diffusion to vapour diffusion. However, the rubber skin formation inhibit moisture diffusion to the surface and lead to a lower drying rate. The drying was limited by the moisture diffusion within the rubber sample during the falling rate period.

4.4.3 Rubber quality of stepwise profile for two-stage intermittent Vacuum Convective Drying (VCD)

The final product quality, including total colour change (ΔE) values and textural attributes for the stepwise drying was determined. The data pertaining to the final product quality dried by stepwise two-stage intermittent vacuum convective drying (VCD) is presented in Figure 4.30, 4.31, and 4.32.

4.4.3.1 Colour analysis

The colour change (ΔE) values of the rubber samples through optimization stepwise drying process was plotted in Figure 4.30 below:

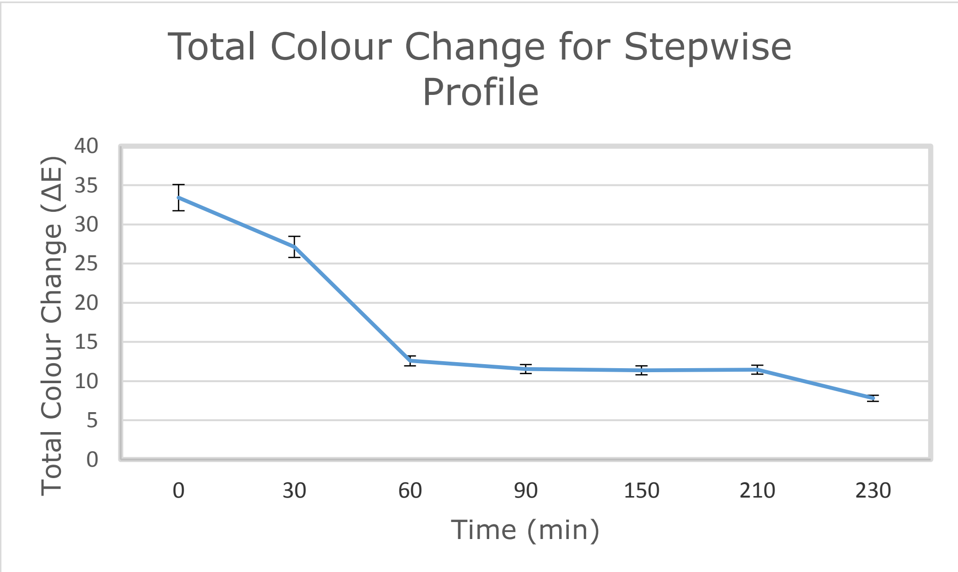


Figure 4.30: Total colour change for rubber dried with stepwise drying

Figure 4.30 shows the colour change (ΔE) values of crumb rubber throughout the stepwise drying process. The stepwise drying gave better colour quality than other drying condition as it had a higher lightness (L*) value of 16.3 ± 3.89, lower a* value of 5.1 ± 0.64 and lower b* value of 11.1 ± 0.35. By understanding the moisture content – browning profiles for crumb rubber, the browning of crumb rubber when drying at different drying temperature could be controlled. As the preference of the rubber colour is golden yellow or golden brown, the occurrence of browning need to be optimize via the stepwise drying process. The introduction of intermittent during high temperature drying and gradually decrease the drying temperature also prevent overheating of the rubber samples without prolonging the drying time. The final colour change (ΔE) value, 7.82 for stepwise drying had the closest value to the standard rubber colour compared to other drying conditions. Results obtained in this study suggested that stepwise drying coupled with intermittent drying via two-stage vacuum convective drying technique were the best drying condition for optimal browning of crumb rubber. Figure 4.31 shows the rubber had a uniform brown colour distribution on the surface and there was no white spots or blackening of the sample.

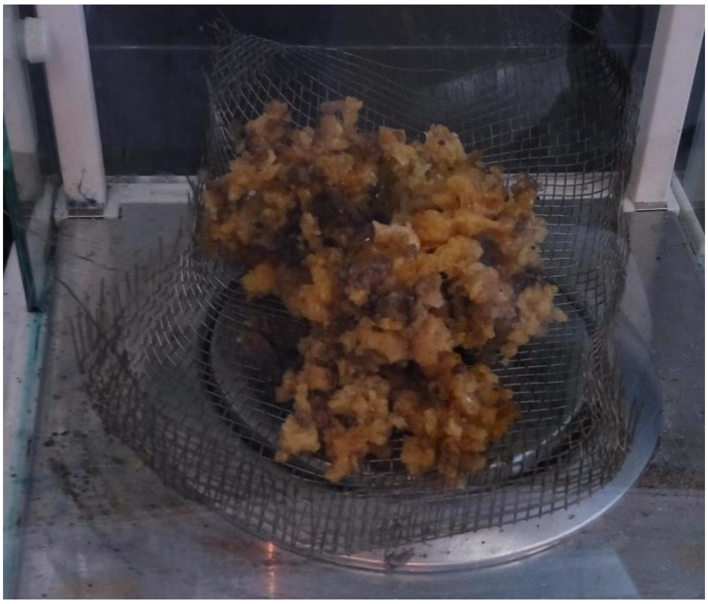


Figure 4.31: Final colour for dried rubber

4.4.3.2 Texture analysis

The stepwise profile for the second stage of hot air drying lead to a high hardness and stickiness rubber material. As the rubber was dried intermittently, the heat stress of rubber reduced. At this state, the crystalline order of the rubber turned into amorphous state. At amorphous state, rubber returned to their tangled and disordered state, which lead to a decrease of hardness value and stickiness value to 51.16% and 35.54% lower. It was seen here that the hardness changed as rubber samples decreased with moisture content. Subsequently, the rubber experienced progressive hardness, unlike the initial state where there was a 54.18% of hardness increment for the first 60 min drying. The textural change of the rubber samples through optimization stepwise drying process was plotted in Figure 4.32 below:

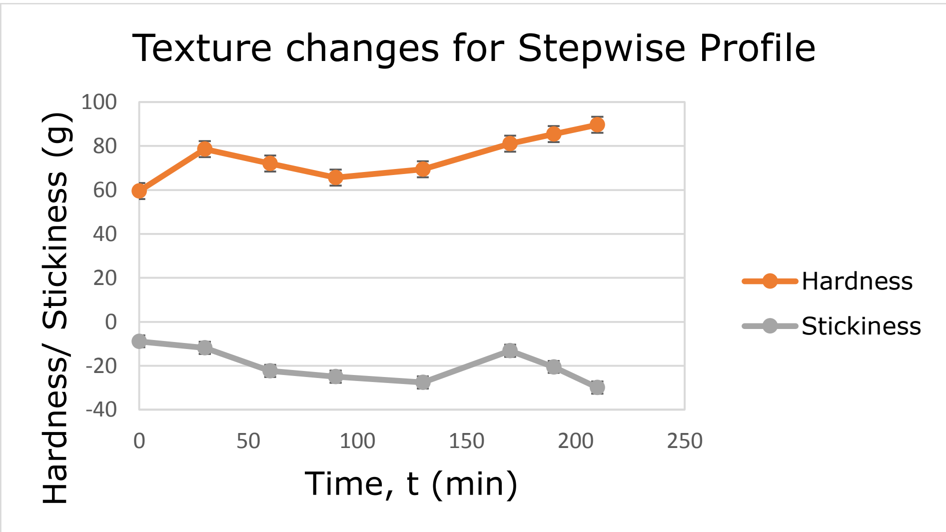


Figure 4.32: Textural change for rubber dried with stepwise drying

Based on Figure 4.32, the maximum hardness (H), 89.66g and maximum stickiness (S), -29.87g were recorded when the rubber was fully dried. The rubber hardness and stickiness increased to the max after vacuum pre-drying and hot air drying at HAD150. Hardness increased with increase in heating temperature due to the crystallisation of crumb rubber (Whitby, 1937; Pickles, 1924) and localised variations in the moisture content (Omolola et at., 2015). In addition, the high internal stresses caused collapse of rubber structure and became the material became compacts.

Likewise, the rubber stickiness increased from -8.88 to -29.87 throughout the drying process. The increment of rubber stickiness showed that the oxidation process or depolymerization had occurred. The stickiness or tackiness or rubber was a desired phenomenon, which improved the processability. With this stepwise drying method, the final rubber products were moderately hard with higher sticky compared to the standard rubber properties when the drying process were completed within pre-determined time frame. This suggests that the two-stage intermittent vacuum convective drying with a time-varying stepwise temperature profile could offers an alternative to the single stage vacuum drying or hot air drying for improving the rubber textural attributes.

CHAPTER 5

Conclusions

5.1 Research summary

One of the major challenge for the rubber processors during the rubber drying was to reduce the drying time and moisture content, without affecting the quality, particularly on the incorporation of wet particles in dried rubber, colour changes, hardness and stickiness. Existing practise of drying crumb rubber at high heating temperature via hot air convective dryer for a prolonged period causes poor product quality and low drying efficiency. By using two-stage vacuum convective drying process, with a low pressure drying at the initial transient period and a stepwise temperature profile at the falling rate period, the heat and mass transfer rate increased, thus, lowers the total drying time required. The shorter drying time was ensured by the two-stage VCD with a shorter vacuum pre-drying duration. However, the two-stage VCD method experienced uneven surface browning of rubber and skin formation on the samples. Thus, a subsequent study on intermittent drying was carried out.

5.2 Two-stage intermittent Vacuum Convective Drying (VCD)

The two-stage vacuum convective drying consisted of two stages. The first stage was the vacuum pre-drying (V), while the second stage was hot air drying (HAD) to finish the drying process. The experiments were conducted at three operating parameters, including vacuum pre-drying duration, hot air drying temperature and sample diameter to evaluate the drying curves and the quality change. The initial transient period during the first stage was connected to the increasing rate period and reached the critical moisture content before the falling rate period began. Two-stage vacuum convective drying (VCD) at a lower vacuum pre-drying duration (30 min) – V30, a higher hot air drying temperature (150°C) - HAD150 and a lower sample diameter (10mm) had the highest drying rate, 0.0033 g H2O min^{-1}, among all test parameters for continuous drying. It was observed that the heating temperature affected the drying rate and total drying time. To sum up, it can be affirmed that combining low pressure at first stage, high temperature and intermittent drying at second stage were useful in hindering quality degradation of rubber.

A series of investigation, involves a comparative study of intermittent ratio and intermittent duration was conducted for vacuum - convective drying. The intermittent ratio of 0.75 and intermittent duration of 15 min was decided based on the comparison of drying time and drying kinetic. Then, another comparison of engineering properties (drying rate and effective moisture diffusivity) and quality parameters (colour, hardness and stickiness) were tested to determine the suitable process conditions (continuous / intermittent) for rubber drying. The results showed that rubber samples experience a higher moisture loss with higher drying rate during the equipment change from vacuum dryer to hot air convective dryer.

5.3 Stepwise profile of two-stage intermittent Vacuum Convective Drying (VCD)

The improved drying system provides incomparable benefits in terms of short drying time, premium quality products and flexible drying capacities. The improvement of technology is attractive from an economic point of view as there is a higher demand for crumb rubber with consistent quality. When the rubber is not dried properly, costs rise for all subsequent processes due to adverse quality changes. Hence, the choice between drying technologies is typically influenced by the impact of quality changes on revenue. While the proposed two-stage vacuum convective drying technology is unlikely to show large cost advantages over other technologies, it may be a more profitable technology if revenue increases with the improved rubber quality. The impact on revenue and profit depends not only on the change in quality but also on the extent of the premium paid for higher quality rubber. Thus, this stepwise profile should be considered to apply in rubber processing plants.

5.4 Future Studies

The present work covers only experimental-based investigation of the quality problem in rubber drying. A model-based investigation or simulation could be considered to identify alternative rubber drying technology.

Optimization in drying time also mean that less energy is also required for the mastication process. However, the analysis of specific energy consumption is not covered in this study. The investigation on both the energy consumption and carbon footprint would be an advantage for the proposed optimized drying test or any other more efficient rubber drying method.

The technology would also be more attractive from an economic point of view if the drying cost is lowered. However, the cost analysis is not within current scope of research. A thorough comparison of the revenue and costs associated with alternative rubber drying technologies could be considered as subsequent research topic.

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