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# Effects of latex type and processed-mica waste loading on the structural and thermo-physical properties of natural rubber latex foam composites



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## ABSTRACT

Mica waste, a type of commercial waste produced in bulk quantities by the mica mining industry, was processed and added to Natural Rubber Latex Foams (NRLF) made of centrifuged and creamed latex. Following the Dunlop method, NRLF composites with various Processed Mica Waste (PMW) loadings (0, 2, 4, 6, 8, and 10 Phr) were prepared, and the thermo-physical characteristics were compared. Thermal conductivity, electrical resistivity of NRLF against the latex type and mica loading were compared for the first time. NRLFs prepared using creamed latex exhibit 33 and 50  $\mu\text{m}$  (width and height) cell diameter, 3 ibf hardness, 281 % swelling index improvements and  $0.05 \text{ Wm}^{-1}\text{K}^{-1}$  thermal conductivity, and 1 °C glass transition temperature ( $T_g$ ) reductions than centrifuged NRLF and indistinguishable electrical resistivity. With the addition of mica (0–10 Phr), both NRLF types showed a similar ascending trend in hardness (42 ibf), water absorption (16 %),  $T_g$  (7 °C), thermal conductivity ( $0.54 \text{ Wm}^{-1}\text{K}^{-1}$ ), electrical resistivity ( $69 \times 10^3 \Omega \text{ m}$ ) with decreasing gel time (3 min) and swelling index (550 %). The key objective of this research was to prepare PMW-filled NRLF and compare structural, electrical and thermo-physical properties, for the first time, against mica content and latex type.

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## 1. Introduction

Natural rubber foams which are composed of cellular structures cover a special class of products and are widely used in applications where properties such as buoyance [1], light weight [2], energy absorption [3] and thermal insulation ability [3–5] are important. They have a high demand in a wide spectrum of sectors such as transportation [6,7], furniture [8], construction [9], and packaging

industry [10]. There are two main routes of manufacturing foam rubber known as dry rubber route [11] and latex route [12]. In dry rubber route, blowing agents which could be thermally decomposed to generate gases are compounded with dry rubber and subsequently vulcanized under heat and pressure [13–15]. In the latex route, air-incorporated latex through vigorous mechanical agitation is gelled using a suitable gelling agent [16] and expanded to form cellular structures. The cellular structures are then

**Abbreviations:** NRLF, Natural Rubber Latex Foams; NR, Natural Rubber; NRL, Natural Rubber Latex; NRS, Non-Rubber Substances; PMW, Processed Mica Waste; SA, Sodium alginate; SLS, Sodium lauryl sulfate; ZDEC, Zinc diethyldithiocarbamate; ZMBT, Zinc 2-mercaptobenzthiolate; DAHP, Diammonium hydrogen phosphate; DPG, Diphenylguanidine; SSF, Sodium silico fluoride; DRC, Dry Rubber Content; TSC, Total Solid Content; VFA, Volatile Fatty Acid number; MST, Mechanical Stability Time; SEM, Scanning Electron Microscope; DSC, Differential Scanning Colorimetry; PVC, Polyvinyl chloride.

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vulcanized to obtain latex foam rubber [17]. The latex route has many advantages such as low energy consumption [17] and operational easiness [18] over the dry rubber route.

Among the latex foam rubber, natural rubber latex has a high demand due to its inherent properties in dry and latex rubber such as tensile strength (500–3500 psi) [19], green nature [16,19], and impact absorbency [20,21]. The performance of foam rubber depends on the cell microstructure (shape, size, wall thickness and type), density of continuous phase, filler characteristics, etc [2,12]. Therefore, characteristics of latex compound used for preparation of foam rubber play a major role in controlling foam performance. Ability to generate stable air bubbles with high surface tension and control of shrinkage are among vital factors to be considered in the process of production of foam rubber. Therefore, centrifuged latex, the extensively used form of concentrated latex used in manufacture of foam rubber has high total solid content that promotes the stability of the air bubbles and low shrinkage in the foam [17,22].

Creamed natural rubber latex which could be considered a greener material is another form of concentrated natural rubber latex produced through a phase separation process assisted by chemical substances known as creaming agents. Creamed latex also has comparable properties to centrifuged latex [23]. Creamed latex could be manufactured to have a higher TSC level than centrifuged latex which allows them to be used in manufacturing of foam rubber. In addition, simplicity of machinery, user friendly operational practices, low energy consumption in manufacture and associated low production cost, possibility of manufacture in small volumes promoting small scale foam rubber industries are some of the non-technical factors which justify the investigations on the potential of creamed latex for manufacture of foam rubber despite the extended time taken for creaming process of latex [24,25]. Minor quality variations such as relatively high non-rubber material content present in latex and availability of creaming agent residues in creamed latex may perhaps influence the foam properties. However, the comparative studies on the properties of foam rubber prepared using centrifuged latex and cream latex could not be readily found in the literature. Suk sup et al. [23,26] has reported the use of creamed latex in foam rubber manufacture perhaps for the first time. This study has shown the potential of using creamed natural rubber latex to manufacture foam rubber with lower porosity compared to its centrifuged latex counterpart. However, this study has focused only on the strength related properties and density of latex foam.

Surprisingly, a majority of works in this field is focused only on the natural rubber latex concentrated through centrifugation i.e. Centrifuged latex. Furthermore, it could be seen that most of the studies have focused on limited common foam properties such as tensile properties, hardness and density. In addition to these properties, some other properties such as electrical conductivity, thermal conductivity, thermal decomposition etc. are also important in the selection of foam rubber for their applications. However, these properties have not been widely reported in the literature for natural rubber latex-based foam rubber, either manufactured from centrifuged latex or creamed latex as well as from filled latex compounds.

In order to increase the value of the product offering advanced/improved properties or to meet the competitiveness in the market, natural rubber latex incorporated with different fillers has been investigated to manufacture foam rubber products. Table 1 illustrates the detailed information of previous studies on NRLF with various filler types.

Use of natural rubber latex incorporated with Micro and nano fillers derived from by-products or waste from other industries in manufacture of NRLF have drawn the interest in the recent past due

to competitive market scenarios and growing environmental concerns together with promotion of green products. In line with these requirements, various fillers derived from industrial and agricultural waste such as rice husk, kenaf fiber, eggshell powder have been studied in preparation of NRLF composites [27,36,47].

Similar to various waste materials used in preparation of latex composites, mica waste which is remained in the mica mining industry has a potential to incorporate into latex to manufacture latex foam composites. Utilization of mica as a resource material has already been studied for various applications in different industries including polymer industry [48–51]. Table 2 presents several studies of mica as a filler in polymers, polymer blends and composites.

Dananjaya et al. [70] has reported the possibility of incorporation of processed waste mica into NRLF to improve several physio-mechanical properties [35]. However, to the best of the authors' knowledge, conductive properties and several important comparisons of physical properties for industrial usage (gelling time, hardness, water absorption and swelling) have not yet been studied using waste mica as a filler for preparation of NRLF. Considering this fact along with the limited amount of works carried out on creamed rubber latex-based foam composites and the associated economic, environmental and technical advantages, a series of work on the waste mica filled NRLF composites made out of creamed latex was conducted by the authors.

The work presented in this study focuses the selected properties including some rarely or so far not reported properties in NRLF composites such as electrical and thermal conductivity of mica filled foam rubber manufactured using natural rubber creamed latex in comparison to the centrifuged latex-based counterparts. The effect of mica loading on the selected properties is also examined. Industrial waste as a raw material to a separate industry is a better solution for the burning problem of environmental pollution and waste management. The potential of the usage of a dust waste to increase properties of natural rubber latex foams is presented here. Many foam industries tackle with the heat generation on the latex foams due to their reduced thermal conductivity. Therefore, the study on thermal conductivity adding thermally conductive mica into NRLF composites provides a greater intention on real world concerns.

## 2. Experimental

### 2.1. Materials

Dartonfield rubber processing factory, Sri Lanka provided Natural rubber field latex (30 % (w/w) DRC). Mica powder waste (7 % from the total production) generated from mica mining and processing industry was collected. Industrial grade sodium alginate (SA), sodium lauryl sulfate (SLS), (poly(dicyclopentadiene-co-P-cresol)), sulfur, zinc oxide (ZnO), zinc diethyldithiocarbamate (ZDEC) and zinc 2-mercaptobenzhiolate (ZMBT), diammonium hydrogen phosphate (DAHP), ammonia, potassium oleate soap, diphenylguanidine (DPG), sodium silico fluoride (SSF) were kindly provided by Richard Pieris Natural Foams Ltd., Biyagama, Sri Lanka. Lak latex processing company, Agalawatta, Sri Lanka supplied centrifuged natural rubber latex.

### 2.2. Preparation of creamed latex

Field natural rubber latex (NRL) was preserved with addition of 10 % ammonia at 0.7 % (w/w). Subsequently, preserved NRL was converted to creamed latex following the recommended cream rubber manufacturing procedure reported in the literature through

**Table 1**

The list of previous studies on NRLF filled with different fillers and their properties.

Author	Ref.	Study	Objectives	Research findings
Kudori et al.	[27]	The effects of filler contents and particle sizes on properties of green kenaf-filled natural rubber latex foam.	To assess the effect of kenaf content and size on the mechanical properties of NRLF	Kenaf fiber addition from 0 to 7 Phr increased modulus at 100 % (M100) from 0.2 MPa to 0.28 MPa, 40 % of compression strength, 36 % compression set, and foam density by $76 \text{ kgm}^{-3}$ . 0.12 MPa reduction of tensile strength reduction and 0.1 MPa development of tensile modulus (100 %) from control to 7 Phr kenaf loading in NRLF
	[28]	Kenaf core and bast loading vs. properties of natural rubber latex foam (NRLF)	To compare density, tensile properties, swelling percentage, compression, microstructural character and accelerated aging of kenaf core and bast filled NRLF	
Surya et al.	[12]	Effect of partial replacement of kenaf by empty fruit bunch (EFB) on the properties of natural rubber latex foam (NRLF)	To study the properties of NRLF by changing the ratio of kenaf and fruit bunch lading	Compression set, compressive strength, density was improved 60 %, 0.04 MPa, $0.8 \text{ kgm}^{-3}$ and swelling, tensile strength was reduced by 240 %, 0.8 MPa respectively from 0 to 7 Phr fruit bunch loading in NRLF
Bashir et al.	[29]	Mechanical, thermal, and morphological properties of (eggshell powder)-filled natural rubber latex foam.	To study the potential reinforcing effect of egg shell powder on NRLF properties	Egg shell powder filling from 0 to 10 Phr doubled M100, 0.25 MPa compression stress, 23 % compression set, and 0.05 MPa tensile strength of natural rubber latex foam composites
Rathnayake et al.	[30]	Imparting antimicrobial properties to natural rubber latex foam via green synthesized silver nanoparticles.	To synthesis of silver nano particles inside the liquid dispersion centrifuged NRL and its routine for making antimicrobial NRLF material more suitable and practical method to make silver nano-particles incorporated natural rubber latex foam not only in lab-scale synthesis but also in large-scale production	Silver nano particles made a toxic environment for bacteria
	[31]	Novel Method of Incorporating Silver Nanoparticles into Natural Rubber Latex Foam	To present a more suitable method to prepare silver nano particles filled NRLF for both lab scale and industrial scale production	Larger diameter of bacterial inhibition zone could be observed in silver nano particles filled NRLF than controlled samples
	[32]	Enhancement of the antibacterial activity of natural rubber latex foam by the incorporation of zinc oxide nanoparticles.	To synthesize and characterize zinc oxide nanoparticles filled NRLF	10 times antibacterial activity of 4 Phr nano zinc oxide filled NRLF with compared to micro-zinc oxide filled NRLF after 6 h
	[33]	Synthesis and characterization of nano silver based natural rubber latex foam for imparting antibacterial and anti-fungal properties.	To produce a novel NRLF that has antimicrobial properties against two major groups of bacteria and fungi	Silver nano particles prepared from chemical reduction method of silver nitrate by tri-sodium citrate enhanced anti-bacterial and anti-fungal properties of NRLF
Phomark et al.	[34]	Antibacterial effect of Ag-doped $\text{TiO}_2$ nanoparticles incorporated natural rubber latex foam under visible light conditions.	To synthesize antimicrobial NRLF with the incorporation of Ag-doped $\text{TiO}_2$ nanoparticles	Incorporation of silver, zinc, Ag-doped $\text{TiO}_2$ nanoparticles increased the antimicrobial properties of the NRLF
	[1]	Natural Rubber Latex Foam Reinforced with Micro- and Nanofibrillated Cellulose via Dunlop Method.	To develop NRLF composites with improved absorption capacity and mechanical properties	$0.125 \text{ kgdm}^{-3}$ of density, $10 \text{ moldm}^{-3}$ of crosslink density, 0.45 MPa of tensile modulus enhancement at 20 Phr nano cellulose filled NRLF than 0 Phr composites
Datta et al.	[20]	Preparation, morphology and properties of natural rubber composites filled with untreated short jute fibers.	To obtain the NRLF filled with short jute fibers and examine the effect of applied fibrous filler content on the tear strength, tensile properties, behavior under cyclic compression, rebound resilience, hardness, abrasion resistance, density and swelling parameters of NRLF	Three times increase of 100 % tensile modulus, a reduction of swelling ratio could be experienced with adding short jute fibers up to 10 Phr into NRLF.
Dananjaya et al.	[35]	Waste mica as filler for natural rubber latex foam composites	To process waste mica dust into industrially consumable form of filler and evaluate its potential application as filler in NRLF made out of centrifuged latex	$3.5 \times 10^{-3} \text{ dm}^{-3}$ growth in crosslink density, 42 ibf rise in hardness and 281 % reduction of swelling index were experienced upto 10 Phr PMW in NRLF.
	[19]	A comparative study on mica waste-filled natural rubber foam composites made out of creamed and centrifuged latex.	To compare properties of PMW filled foam rubber made from centrifuged latex and creamed latex aiming on biodegradability, antibacterial activity, extractable protein	200 % biodegradability, 35 % compression set, $0.92 \text{ N mm}^{-1}$ tear strength, 0.25 MPa tensile strength, 0.16 MPa modulus at 100 %, $4^\circ \text{C}$ thermal stability improvements resulted with

Zhang et al.	[21]	Enhancement of the Antibacterial Activity of Natural Rubber Latex Foam by Blending It with Chitin. <i>Materials</i>	content and thermal degradation behavior together with some important physio-mechanical properties To identify the antibacterial activity and mechanical properties of chitin– natural rubber foam composites	75 mgml <sup>-1</sup> protein content, 250 % elongation at break reductions of NRLF with addition of processed waste mica up to 10 Phr. 283.1 % of antibacterial properties, 0.12 kgm <sup>-3</sup> foam density, 15 Moh hardness, 0.07 MPa compressive strength risings were reported with 170 % swelling, 0.09 MPa tensile strength, 290 % elongation at break reduction, from 0 to 5 Phr chitin loadings into NRLF composites.
Ramasamy et al.	[36,37]	Tensile and Morphological Properties of Rice Husk Powder Filled Natural Rubber Latex Foam	To investigate the potential and reinforcing effect of rice husk powder, as well as to diminish the overall compounding cost of the NRLF	0–10 Phr rice husk powder enhanced tensile strength by 0.04 MPa, modulus at 100 % by 0.03 MPa, hardness by 25 Mohs, density by 85 kgm <sup>-3</sup> with 200 % reduction of elongation at break
Mahathaningwon et al.	[38]	Morphology and properties of agarwood-waste-filled natural rubber latex foams	To examine the properties and morphology of agarwood-waste-filled NRLFs	Improvements of 55 kgm <sup>-3</sup> for density, 9 (Shore AO) for hardness, 19 % for compression set were observed while reducing of rubber filler interactions from 0 to 6.5 Phr agarwood content in NRLFs
Sallehuddin et al.	[39]	Effects of silane treatment on tensile properties and surface morphology of kenaf bast filled natural rubber latex foam	To assess the effect of silane treatment on tensile and morphology properties of kenaf bast-filled natural rubber latex foam (NRLF)	0.13 MPa and 0.08 MPa drops of tensile strengths were reported for silane untreated and treated kenaf bast filled NRLF. Smaller pore sizes and better cell structure was identified in silane treated kenaf bast filled NRLF as opposed to untreated filler.
Panploo et al.	[40]	Natural rubber latex foam with particulate fillers for carbon dioxide adsorption and regeneration	To develop an eco-friendly material for CO <sub>2</sub> adsorption at ambient temperature and pressure	Foams made by cake mixer had 268 μm larger cell diameters, higher cell density than that of from overhead stirrer. Modified silica filled foams had a 0.39 mg g <sup>-1</sup> higher CO <sub>2</sub> absorption than unmodified silica filled NRLF
Pinrat et al.	[41]	Fabrication of Natural Rubber Latex Foam Composite Filled with Pineapple-leaf Cellulose Fibers	to enhance the method to prepare NRLF with various cellulose loadings from 0 to 7 Phr using Dunlop method	1 MPa tensile strength reduction, 1 % compression set drop, 9 kgm <sup>-3</sup> density growth and pore size enlargement from 0 to 7 Phr Pineapple-leaf Cellulose Fibers into NRLF
Prasbdee et al.	[42]	Effect of Fillers on the Recovery of Rubber Foam: From Theory to Applications. <i>Polymers</i>	To improve and prepare NRF to examine its recoverability and other characteristics by the incorporation of charcoal and silica fillers	30 kgm <sup>-3</sup> density, 85 mol m <sup>-3</sup> crosslink density development with addition of fillers from 0 to 8 Phr loadings into NRLF Charcoal and silica loading has increased glass transition temperature of NRLF by 12.91 °C and 4.41 °C respectively.
Yang et al.	[43]	Natural rubber latex/MXene foam with robust and multifunctional properties	To provide a easy method to produce high-strength elastic electrical conductive foams with EMI shielding capabilities, which broaden the potential applications of natural latex foam materials	171 % and 157 % enhancements of tensile strength at 2 and 3 Phr Mxene loading in NRLFs, 0.8–1 °C Tg improvement, 6.18 × 10 <sup>-4</sup> Sm <sup>-1</sup> electrical conductivity rise peaked with the addition of Mxene
Hu et al.	[44]	Influence of l -quebrachitol on the properties of centrifuged natural rubber	To examine the effect of l -quebrachitol on the properties of NRLF	0.05 MPa and 2 °C reduction of Young's modulus and glass transition temperature up to 3rd centrifugation cycle
Boondamneon et al.	[45]	Recycling Waste Natural Rubber Latex by Blending with Polystyrene - Characterization of Mechanical Properties	To investigate the potential of re-use waste NRLF and characterize the mechanical property development	7.6 × 10 <sup>-5</sup> crosslink density, 4.7 MPa tensile strength improved waste NRLF filled blend foams than virgin blend
Keavalin et al.	[46]	Effects of Bamboo Leaf Fiber Content on Cushion Performance and Biodegradability of Natural Rubber Latex Foam Composites	To investigate the effects of bamboo leaf content on the mechanical properties, foam structure, cushion performance, and biodegradability.	21.27 MPa compressive strength, 1.8 times biodegradability, 3.5 % compression set rise from 0 to 10 Phr bamboo leaf content in NRLF composites

**Table 2**

An outlook of prior studies on mica as a filler in polymers, and their blends and composites.

Author	Ref.	Study	Objectives	Research findings
Andraschek et al.	[52]	Mica/Epoxy-Composites in the Electrical Industry: Applications, Composites for Insulation, and Investigations on Failure Mechanisms for Prospective Optimizations	To provide an overview of the properties of virgin components, the composite, and the possible occurring failure mechanisms which is used in insulation materials for high voltage rotating machines with prospective optimizations	Mica is a superior material for electrical insulation
Baurova et al.	[53]	A study of mica structure and strength properties of polymer materials based on it	To investigate the potentials of use of mica and mica pigments as the fillers during preparation of smart polymer materials.	1.3 MPa shear strength improvement up to 10 wt% mica loading
Xiaolong et al.	[54]	Study on the performance and mechanism of modified mica for improving polypropylene composites	To improve the mechanical properties of polypropylene composites by addition of modified mica	40.72 % and 35.66 % decrease of material flow rate of PP with mica and modified mica against virgin polypropylene
Sreekanth et al.	[55]	Effects of Mica and Fly Ash Concentration on the Properties of Polyester Thermoplastic Elastomer Composites	To investigate the influence of mica with altering concentration on the mechanical, thermal, electrical, rheological and morphological properties of polyester thermoplastic elastomer	7000 kVm <sup>-1</sup> dielectric strength, 560 pas viscosity and 1020 MPa flexural modulus improvements and doubled flexural strength peaked at 40 Phr
Kahraman et al.	[56]	Influence of mica mineral on flame retardancy and mechanical properties of intumescent flame-retardant polypropylene composites	To study the synergistic effect of mica mineral and enhancing the flame retardancy properties of PP in order to achieve cost competitive solution	285 MPa elastic modulus and 3.7 MPa tensile strength peaked at 32 Phr mica in polypropylene
Kuelpmann et al.	[57]	Influence of platelet aspect ratio and orientation on the storage and loss moduli of HDPE-mica composites	To investigate the rheological properties of HDPE-mica composites with the particles oriented parallel or perpendicular to the flat sample surface	Shear modulus was increased strongly with the aspect ratio in mica-HDPE polymer composite
Zhao et al.	[58]	Study of the mechanical properties of mica-filled polypropylene-based GMT composite	To study the effect of the addition of mica on the mechanical properties of GMT To reveal whether the addition of mica will influence the interfacial properties between the fiber and the matrix	Eith the reduction of mesh size of mica from 60 to 100 S, 1620 MPa tensile modulus, 21.6 MPa tensile strength, 1800 MPa flexural modulus enhanced with 81 Jm <sup>-1</sup> Izod impact strength.
Jamel et al.	[59]	Mechanical Properties and Dimensional Stability of Rigid PVC Foam Composites Filled with High Aspect Ratio Phlogopite Mica	To improve physical and mechanical properties of PVC using mica as a filler	44 %-dimensional stability improvement of PVC from 0 to 20 wt% mica content and ultimate tensile strength was developed by 2.78 MPa at 10 wt % mica.
Liang et al.	[60]	Mechanical, Thermal, and Flow Properties of HDPE –Mica Composites	To study the effects of mica loading on the flow, mechanical, and heat resistance properties of HDPE –mica composites	11 kJm <sup>-2</sup> reduction of impact strength and 9 °C heat distortion temperature enhancement up to 15 wt % mica content in HDPE composites
Souza et al.	[61]	Effect of synthetic mica on the thermal properties of poly(lactic acid)	To study the effect of the type and amount of filler on some important thermal properties of these PLA nanocomposites	10 °C decomposition temperature growth and 26.2 °C glass transition temperature reduction were resulted from 0 to 7.5 wt % synthetic mica filled PLA nanocomposites
Parvaiz et al.	[62]	Effect of Particle Size of Mica on the Properties of Poly [Ether Ether Ketone] Composites	To examine the effects of particle size of mica on the mechanical, thermal and morphological properties in Poly[Ether Ether Ketone] Composites	20 wt % mica filled composites exhibited about a 79 % increase in the storage modulus at 50 °C and about a 68 % increase at 250 °C
Deshmukh et al.	[63]	Mica-filled PVC composites: Effect of particle size, filler concentration, and surface treatment of the filler, on mechanical and electrical properties of the composites	To investigate the effect of the mica with a range of particle size and filler concentration on mechanical and electrical properties of PVC	300 % stiffness enhancement was peaked at 30 wt % mica in PVC, 120 MPa rise of Young's modulus and 8.5 × 10 <sup>14</sup> improvement of surface resistance experienced for 50 Phr loading of 44 μm mica
Verbeek et al.	[64]	Mica-Reinforced Polymer Composites.	To investigate the impact of mica on various properties of polymer matrices	Mica could improve thermo-mechanical properties of polymer composites and highly compatible material with polymer matrices
Yazdani et al.	[65]	Effects of silane coupling agent and maleic anhydride-grafted polypropylene on the morphology and viscoelastic properties of polypropylene–mica composites	To investigate the reinforcing effect of mica and maleic anhydride in polypropylene composites	4.1 MPa tensile strength, 3D hardness and 4.7 MPa flexural strength rise from 0 to 40 wt % mica in polypropylene composites
Somarathna et al.	[66]	Waste mica and carbon black filled natural rubber composites	To compare the properties of waste mica and carbon black in natural rubber vulcanizates	Waste mica has a potential to replace carbon black in natural rubber compounds at low filler contents
Fernandes et al.	[67]	Characteristics of Acrylic Rubber Composites with Mica and Carbon Black	To compare the cure, mechanical and swelling behaviors of mica and carbon black filled acrylic rubber composites	40 Phr mica filled composites were performed similar behavior to 20 Phr carbon black filled composites
Martinez et al.	[68]			

On the Combined Effect of Both the Reinforcement and a Waste Based Interfacial Modifier on the Matrix Glass Transition in iPP/a-PP- pPBMA/Mica Composites

Flexural behavior of PP/Mica composites interfacial modified by a p-phenylen-bis-maleamic acid grafted atactic polypropylene modifier obtained from industrial wastes

[69]

To study and predict the glass transition temperature ( $T_g$ ) for the identification and interpretation of the combined and synergistic effect of the mica reinforcement, and the p-phenylen-bis-maleamic acid grafted atactic polypropylene

To evaluate the influence of the interfacial modifications performed in the injection molded PP/Mica system

4 °C improvement of glass transition temperature from 10 to 25 Phr mica content in Polypropylene

18.68 MPa tensile strength and 14 MPa flexural strength growth from 0 to 35 Phr mica loading in PP

addition of 0.4 Phr sodium alginate using 10 % solution [71]. The creaming was carried out for 14 days and characterized using the standard test methods given in Table 3.

2.3. Mica waste preparation and characterization

Mica waste discarded from the mining industry was collected, processed and characterized as shown in Table 4. A flow chart indicating the steps involving the processing of mica filled NRLF is presented in Fig. 1. Processed mica was characterized for their morphology using scanning electron microscopy.

2.4. Preparation of natural rubber latex foams

NRLFs were manufactured following the Dunlop process using centrifuged and creamed latex [33,47]. For 100 parts of natural rubber centrifuged and creamed latex, both 50 % Potassium oleate soap and 50 % ZnO were added in 6 Phr levels. 50 % ZDEC, 50 % ZMBT and 50 % poly(dicyclopentadiene-co-P-cresol) were mixed in 1.5 Phr contents. 12.5 % SSF and 50 % SLS were added in 7 and 0.25 Phr values respectively. For both latex types, PMW was added in 0, 2, 4, 6, 8 and 10 Phr loading as an aqueous dispersion.

Table 3  
ISO methods for property evaluation of creamed latex.

Test	ISO Reference No.
Dry Rubber Content (DRC)	ISO 506: 1992(E)
Total Solid Content (TSC)	ISO 124: 1997 (E)
Volatile Fatty Acid number (VFA)	ISO 506: 1992(E)
Mechanical Stability Time (MST)	ISO 35: 2004(E)

Table 4  
Composition of waste mica.

Chemical Composition	Weight %
SiO <sub>2</sub>	37.4
Al <sub>2</sub> O <sub>3</sub>	27.6
MgO	19.1
K <sub>2</sub> O	10.2
Fe <sub>2</sub> O <sub>3</sub>	4.8
CaO	0.4
Na <sub>2</sub> O	0.4
Other	0.1

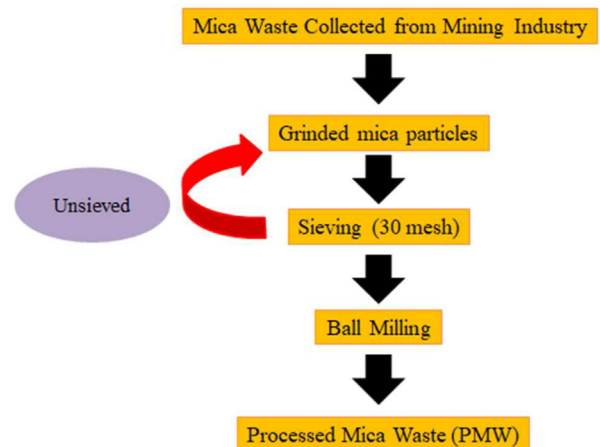


Fig. 1. Process flow chart of mica waste processing.

First, to de-ammoniate the latex, both type of NRL were stirred using a continuous air flow for 15 min using a lab-scale mechanical agitator at 50 rpm. Then, mixture was stirred for 20 min in the medium speed of magnetic stirrer after adding potassium oleate soap and Sodium Lauryl Sulfate (SLS) into the latex. Processed waste mica filler dispersions, sulfur, ZMBT, ZDEC and antioxidants (poly (dicyclopentadiene-co-P-cresol) in their dispersions with given concentrations were added into the latex mixture. Then mixture was mixed for another 20 min and allowed to mature for 8 h at room temperature (28 °C). Until volume reached 3 times the initial volume within 3 min, latex compound was beaten using the hand mixer (Philips HR-3740). Then gelling agents (DPG (secondary gelling agent) and SSF (primary gelling agent)) and ZnO were added at once to the mixture and mixed for another 30 s using hand mixer (speed level 3) at the same conditions. The latex compound was allowed to gell and transferred into an aluminum mold and placed in an oven for 2 h at 100 °C to complete the foaming. Then the foam was washed thoroughly with distilled water, squeezed well and dried in the oven at 70 °C for 8 h. For the preparation of control foam rubber samples, the same procedure was continued.

## 2.5. Morphology

Scanning electron microscope (FESEM: Zeiss Supra model 35 VP, Germany) is used to scan tensile fractured surface of mica filled NRLFs were scanned. The samples were stranded on aluminum stubs, with a thin layer of gold-palladium sputter-coated.

## 2.6. Gelling time

The time spent for gelling was measured by calculating the time become non sticky the latex compound, 30 s stirring after adding gelling agents [35].

## 2.7. Bulk density

Equation (1) was used to determine the densities of cuboid shape (5 cm × 5 cm × 5 cm in volume) PMW filled NRLF samples following the ASTM D1622-03.

$$\text{Density} = \frac{M}{(l \times w \times t)} \quad (1)$$

M, l, w and t are the mass (g), length (cm), width (cm) and thickness (cm) of the samples, respectively.

## 2.8. Hardness

According to ASTM D3574, hardness of the foam rubber samples (55 mm in length, 30 mm width and 25 mm thickness) were measured by an Indentation Load Deflection machine (Model HD-750, China).

## 2.9. Swelling index

Vulcanized test pieces (30 × 10 × 2 mm dimensions) were weighed using an analytical balance (Precisa, Model 228, Switzerland). Then the specimens were immersed in toluene until it reaches equilibrium swelling. After equilibrium swelling, the samples were taken out and the solvent was blotted from the surface of the vulcanizate using filter paper and weighed instantly. Percentage of swelling was calculated according to ASTM D 0471-0479 and Equation (2), where,  $M_1$  and  $M_2$  are initial mass of specimens (g) and mass of specimens after immersion in liquid (g) [21,28].

$$\text{Swelling \%} = \left( \frac{M_2 - M_1}{M_1} \right) \times 100\% \quad (2)$$

## 2.10. Water absorption

Water absorption of the NRLF were calculated according to ASTM C272 standard [72]. The specimens were 3" × 3" × 0.5" (l × w × t) in dimensions. First, the initial weight of the specimen was measured using an electronic balance. Secondly, the specimen was immersed in distilled water and allowed to drain it for 1 min until excess water is removed. Then using an electronic balance, the final weight was again measured. Water absorption percentage was calculated using Equation (3), where  $M_1$  and  $M_2$  are the initial mass and the water immersed mass of the specimen respectively.

$$\text{Water Absorption \%} = \left( \frac{M_2 - M_1}{M_1} \right) \times 100\% \quad (3)$$

## 2.11. Thermal properties

### 2.11.1. Dynamic scanning calorimetric studies (DSC)

The glass transition temperatures of the foam samples were measured by Netzsch DSC 214 F1, Germany with a continuous nitrogen gas flow. The scan was carried out from –70 °C to 300 °C at a scanning rate of 10 °C/min.

### 2.11.2. Thermal conductivity

The thermal conductivity test was conducted according to ASTM International (2010c) [2,4,5]. The Netzsch test apparatus was used to measure the thermal conductivity of the NRLF and the thermal conductivity was measured using the principles of heat transfer as shown in Equation (4), where k is the thermal conductivity, Q is the sample heat flow, s is the sample thickness, A is the sample area and  $\Delta T$  is the temperature difference across the plates.

$$k = (Q \times s) / (A \times \Delta T) \quad (4)$$

## 2.12. Electrical properties

Electrical conductivity of foam samples was measured using resistivity tester following ASTM D991 method. Sample size of the test specimens was 15 mm × 15 mm × 15 mm (l × w × t). Resistivity of the sample was calculated using Equation (5), where R,  $\rho$ , L and A are resistance, resistivity, length and area of samples respectively.

$$R = \frac{\rho L}{A} \quad (5)$$

## 3. Results and discussion

### 3.1. Physicochemical properties of centrifuged and creamed latex

As shown in Table 5, creamed latex has 11.96 % higher TSC value together with 11.73 % higher DRC than the centrifuged latex. Generally, higher DRC values were obtained for creamed latex as the creaming process could be continued until it reaches its highest achievable DRC value whereas centrifuging process limits it value to the standard DRC value (60 % ± 0.2). The study carried out on creamed rubber latex by Yumae et al. has reported this behavior of creamed latex [73]. In some of the studies, it could be found out that creamed latex accounts for lower NRC value and higher MST values



**Table 5**  
Characteristics of creamed latex and centrifuged latex.

Parameter	Creamed latex	Centrifuged latex <sup>a</sup>
Total Solid Content (TSC) % (w/w)	68.6	61.4
Dry Rubber Content (DRC) % (w/w)	67.4	60.2
Non-Rubber Substances (NRS) % (w/w)	1.2	1.2
VFA number	0.17	0.11
Alkalinity (w/w) %	0.70	0.67
MST (seconds)	798	800

<sup>a</sup> Reproduced for the purpose of comparison.

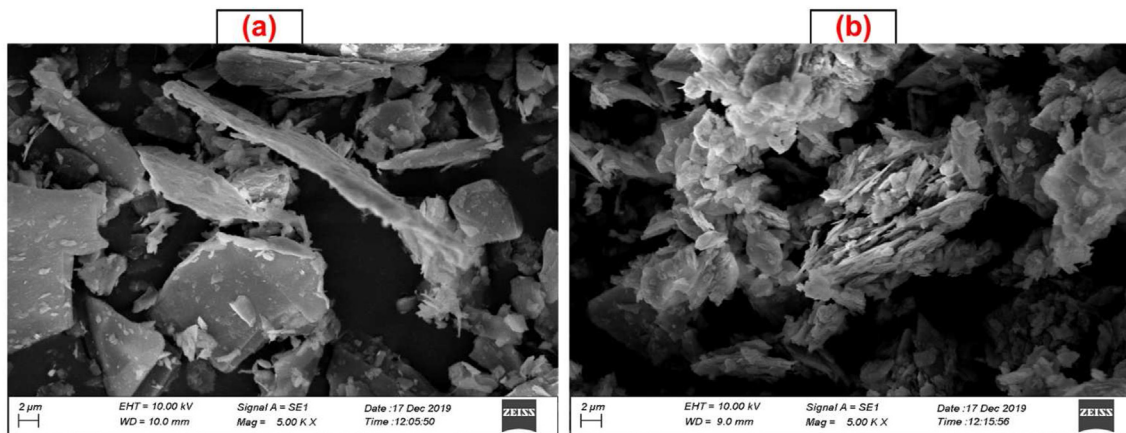
than the reference centrifuged latex, probably depend on the processing conditions used during manufacture. It could be seen that both types of latex used in this study have almost comparable latex properties, except their TSC and DRC values, which make the main difference between the two forms of latex. With regard to the non-rubber content (NRC) values, though values are similar, residuals of additionally added creaming ingredients should be partly accounted for the NRS value of the creamed rubber.

### 3.2. Morphological studies

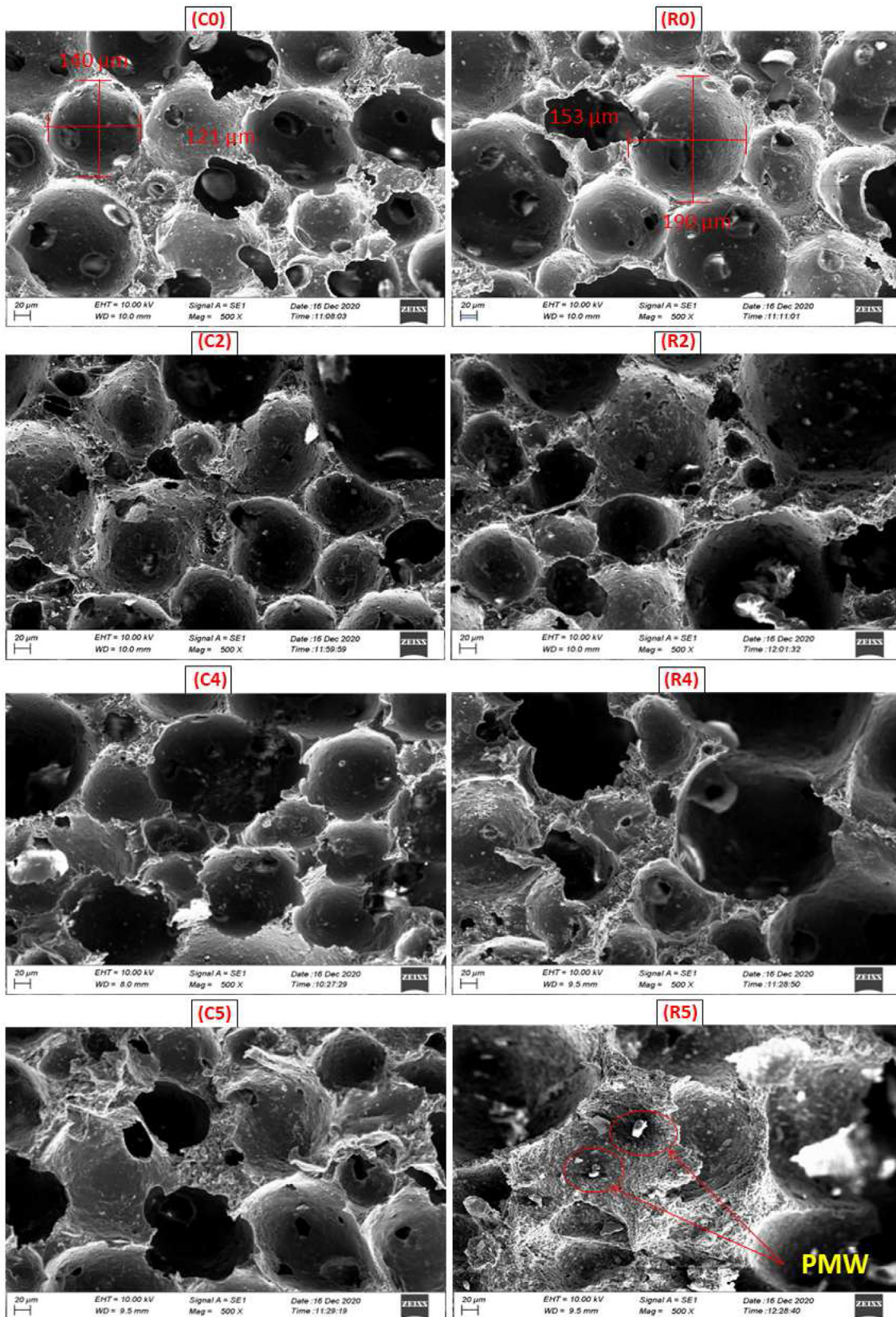
Particle size, their shape and distribution are the major factors that affect the filler performance in a polymer matrix. Smaller filler particles and regular shape fillers facilitate the reinforcement. Formation of agglomerates loses the reinforcing ability. Therefore, the particle size data of UPMW and PMW were carried out to examine the particle size and possible agglomeration. SEM micrographs of PMW loaded centrifuged and creamed NRLF are shown in Fig. 2. It shows that both unprocessed and processed mica particles are platelet like structures irregular in size and shape. Sharp edges could be seen in the particles indicating that they are crystalline in nature. It also provides visual evidence for the reduction of the particle sizes as the result of the grinding process used. Careful examination of both micrographs suggests that there is an improvement in the uniformity of particle sizes in processed mica in Fig. 2(b) when compared to the unprocessed stuff. Fig. 2(a) shows that unprocessed mica has clusters of mica particles having a wide range of aspect ratios. Processing has narrowed down the distribution of aspect ratio of the processed mica waste. Therefore, SEM study confirms the processing methodology used was capable in

reduction of the particle size and irregularity in shape of the mica particles.

Fig. 3 presents the scanning electron microscope images of PMW filled NRLFs made of both creamed and centrifuged latex at different PMW loadings. Both NRLF have a spherical shape open cell structure of different cell sizes. It could be seen that the cell diameter of creamed NRLF (153 μm in width and 190 μm in height) is larger than that of centrifuged NRLFs (121 μm in width and 143 μm in height). This may be ascribed to 11.73 % increase of DRC and 11.96 % rise of TSC of creamed latex compared to centrifuged latex [22]. Phomrak et al. [1] also reported the cell sizes of NRLF composites could be varied from 10 to 500 μm in his study on micro and nano cellulose powder filled NRLF. Here the cell sizes are in the range. Bayat et al. [74] found cell sizes in between 160 μm and 600 μm in silica filled NRLF composites. At the same filler loading, due to the presence of creaming agent, creamed latex density is higher than centrifuged latex density (Fig. 6), because the cell sizes of the NRLF made of creamed latex may become higher. A cell deformation is occurred in 10 Phr PMW loaded creamed latex foam composites. Similar observations were experienced at 10 Phr filler content by Kudori et al. [28] in his study on kenaf fiber loaded foams, Bashir et al. [47] in his study on egg shell powder filled NRLF, by Suksup et al., [23] who compared the unfilled NRLF prepared from both type of latex, and Ramasami et al. [75] in the study of rice husk powder as a filler in NRLFs. This could be observed due to their larger cell sizes of the creamed foams and reduced cell wall thickness. Images of the actual specimen and a model of NRLF composite structures at 0, 4, and 10 Phr PMW loading are illustrated in Fig. 4. Controlled specimen and 4 Phr PMW filled NRLF consist evenly distributed small air voids and rubber matrix compared to 10 Phr PMW filled NRLF. As presented in SEM images, the cross-sectional view of images in Fig. 4 depicts a certain amount of air voids generation and distractions of rubber cells at 10 Phr PMW filled NRLF with a discoloration effect. Air voids were generated inside the NRLF particularly due to the increase of mica loading in a unit volume, Therefore, the volume fraction of natural rubber has decreased and the mica has aggregated as evident from Fig. 3 – R5 (10 Phr PMW filled creamed NRLF). Thus, the cross-sectional views of NRLF presented in Fig. 4 have been confirmed by the SEM images in Fig. 3. Here, some discoloration condition was resulted by mica added into the latex foams. PMW has a pale brown color and the discoloration nature was increased with the addition of mica into the NRLF. Moreover, the model of NRLF at 0, 4, and 10 Phr PMW



**Fig. 2.** a. SEM image of UPMW, b. SEM images of PMW.



**Fig. 3.** SEM images of NRLFs. (C0):0 Phr PMW in centrifuged NRLF, (C2): 4 Phr PMW in centrifuged NRLF, (C4):8 Phr PMW in centrifuged NRLF, (C5) 10 Phr PMW in centrifuged NRLF, (R0):0 Phr PMW in creamed NRLF, (R2): 4 Phr PMW in creamed NRLF, (R4):2 Phr PMW in creamed NRLF, (R5) 10 Phr PMW in creamed NRLF.

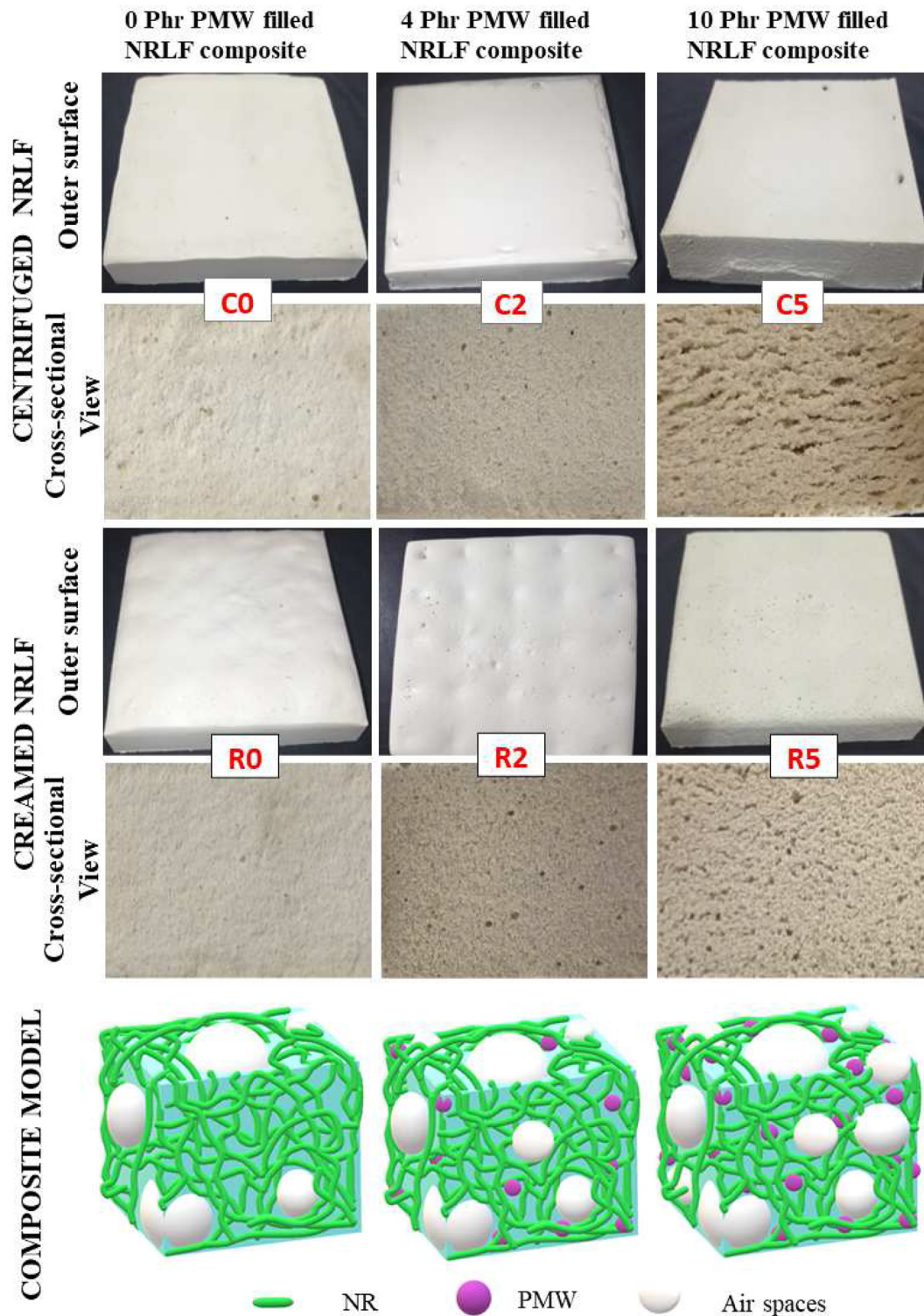


Fig. 4. Images of the outer and cross surfaces of the actual specimen (top), Schematic representation of 3D-models of creamed and centrifuged NRLF (bottom) at 0, 4 and 10 Phr PMW loadings.

filled NRLF shows the rise of air voids with the addition of PMW into the rubber matrix. Also, it introduces a larger amount of rubber-rubber and rubber-filler crosslinks at 10 Phr PMW loaded samples than the controlled samples.

### 3.3. Gelling time

With respect to the latex type, it is shown that gelling time of centrifuged latex-based foam rubber (5.5 min) is higher than that of

creamed latex (5.0 min), however, only by less than half a minute (Fig. 5). Additional stabilizing effect offered by the creaming agent may slightly retard the gelling process of creamed latex [76]. Zinc ions in the media reacted with ammonia present in the latex and and the zinc ammine complexes react with fatty acids and accelerate the gelling. According to the data presented in Table 2, the alkalinity that relates to ammonia content is 0.03 % higher in creamed latex than centrifuged latex. Therefore, the gelling time of creamed latex is 0.5 min lower than centrifuged latex. Hui mei et al. [77] has reported this mechanism in her study on natural rubber foam composites. However, gelling time of both types latex could be considered as comparable to each other and may not make a significant difference in the productivity in industrial applications as the maximum gelling time limits to 5 min. Irrespective to the latex type, gelling time showed 3 min declining of gelling time with the PMW loading. This could be explained by two ways. Gelling of natural rubber latex normally happens at 8.5 pH [78]. The pH of mica dispersion was 9.1 and 7.0 and 7.4 pH values were indicated for centrifuged and creamed natural rubber latex respectively. Adding mica dispersion into latex enhanced the pH value of latex and made closer to the gelling pH value. Hence it made the favorable conditions for gelling faster and reduced gelling time by 3 min. Processed waste mica reinforce natural rubber chains by mica-rubber bonds and entanglements of rubber chains according to the findings of Dananjaya et al. [35]. Further, this ability of mica was accelerated by numerous ions remaining in the mica filler dispersion could also support the sub interactions between rubber molecules through the multivalent ions. Addition filler increase the density and hence the viscosity of the latex phase and facilitates the gelation process. These factors contribute to the fall of gelling time with mica incorporation. Inert foreign particle addition into the latex could increase the nucleation ability of the macromolecules leading to reduction of gelling time as observed in gelation of agarwood waste loaded NRLFs at low filler loading [35,38,73].

### 3.4. Density

Fig. 6 presents the variation of density of NRLF against 0–10 Phr PMW loadings. Density of a foam rubber is one of the main parameters which influence the foam properties. Density of foam rubber composites studied in this study corresponds to the measurement of a weight to volume ratio of multi-phase system consist of filler (heavier phase), rubber (moderately heavier phase) and air (lighter phase). It could be approximately determined following the

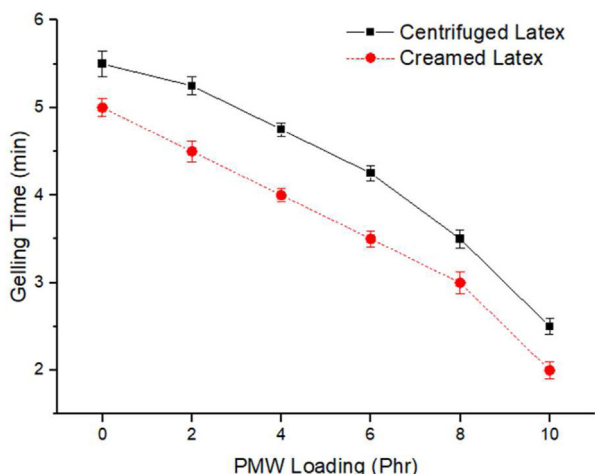


Fig. 5. Variation of Gelling time of composites against PMW loading and type of latex.

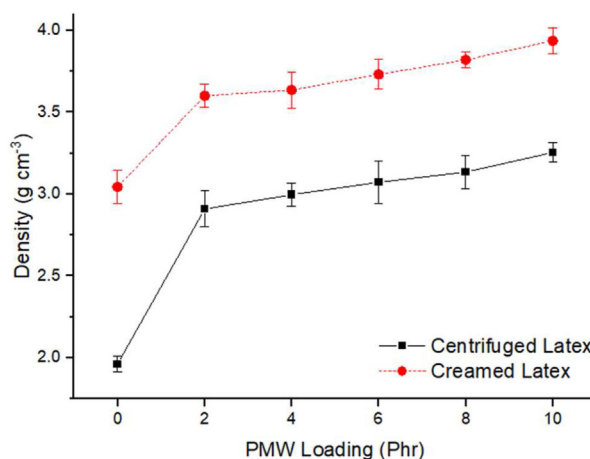


Fig. 6. Density variation against the mica loading and the type of latex.

additive rule using following Equation (6), where  $d_{cp}$  is density of rubber phase,  $d_{fil}$  is the density of filler,  $\phi_{cp}$  is the volume fraction of polymer and  $\phi_{fil}$  is the volume fraction of filler [79].

$$d_{fm} = d_{fil} \phi_{fil} + d_{rp} \phi_{rp} + d_{air} \phi_{air} \tag{6}$$

According to eq. (6), density of the filled foam rubber is governed by two parameters mainly density and volume fractions of rubber, filler and air. Thus, increase in volume fraction of filler or rubber phase increases the density of the foam composites. Creamed latex contains higher total solid content and additionally added sodium alginate (creamng agent) which has a higher density than rubber. Therefore, density of rubber phase formed from creamed latex becomes higher than that of the rubber phase formed from centrifuged latex. This may be attribute to the higher density of NRLF prepared from creamed latex than the counterpart at the same filler loading which is in consistent with the observation reported by Suksup for unfilled foam rubber made out of creamed latex and centrifuged NR latex [23]. In addition, the effect of difference of densities of rubber phases has predominant over the effect caused by the differences of air volume fractions. Similarly, incorporation of PMW of high density into latex also increases the density NRLFs of both types of latex. These trends are clearly shown in Fig. 6 and in consistent with observations reported in similar studies carried out on varying filler loading latex foam with varying filler loading [29,35,80].

### 3.5. Hardness

Fig. 7 shows the variation of hardness of controlled and mica loaded creamed and centrifuged latex foams. Hardness generally depends on rubber-filler interactions (reinforcement) and crosslink density. It could be seen that 22 ibf improvement of hardness of creamed latex foams is higher than the hardness of centrifuged latex foams. Presence of higher non-rubber contents including residual creaming agent in creamed latex may be responsible for higher hardness of creamed latex-based foam rubber. Presence of residual creaming agent in the rubber phase could act as reinforcing centers through trapping the rubber agglomerate [81]. The increase in the hardness by 25 and 42 ibf values for centrifuged and creamed NRLF with increasing PMW loading from 0 to 10 Phr in both foams were identified and the results are well in agreement with the similar studies already reported in the literature [27–29]. There are two reasons for this: the nature of filler and demobilizing effect of filler on the polymer chain. The mica filler is harder than

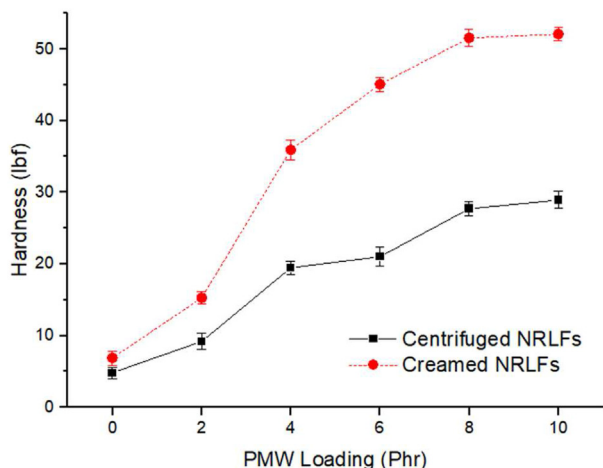


Fig. 7. Variation of hardness of 0–10 Phr PMW loaded creamed and centrifuged NRLFs.

the polymer matrix; thus, when the filler content increases, the hardness of the rubber phase also increases. Moreover, good dispersion of mica filler in the NRLF matrix causes the polymer chain to be dispersed on the filler surface, consequently growing the hardness and stiffness of the foams filled with mica. Also, as evidenced by Fig. 4, higher degree of reinforcement Velavan et al. [82] revealed that uniform dispersion of mica inside the composite structures boosted the hardness of cast hybrid Al mica composites by observing 17.19 % hardness enlargement at 3 wt % mica inside the composite structures.

### 3.6. Swelling index

Fig. 8 presents the effects of latex type and PMW loading on the swelling index of filled NRLFs. Swelling indices of creamed foam rubber are 281 % higher than the centrifuged based foam rubber. This suggests lower crosslink density of the rubber phase of the creamed foam rubber. Compared the creamed latex and centrifuged latex, former has the higher DRC and non-rubber substances including sodium alginate which is used as the crosslink agent [83]. Therefore, it could absorb the higher percentage of solvent molecules gaining enhanced swelling index of creamed latex-based foam rubber. As shown in Fig. 8, there is an initial rapid reduction followed by a gradual reduction in the swelling index by 550 % with the PMW loading. Increasing the filler loading, there is a drop of

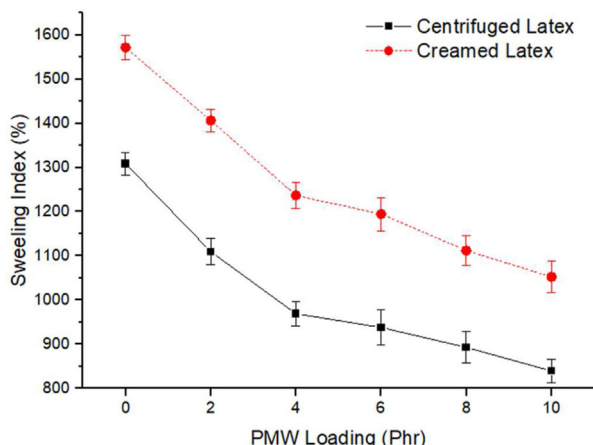


Fig. 8. Effect of mica loading and latex type on swelling index of NRLF.

rubber material amount in unit volume reduces the solvent absorption capacity. It has been reported by Ramasamy et al. [36] in his study of rice husk powder filled latex foams. He has affirmed that increasing number of crosslinks in rubber reduce the swelling index. In a similar study of eggshell powder incorporated NRLFs, Bashir et al. [47] identified that the higher interactions between rubber chains decrease the swelling. Similar observations made for those filled NRLFs are in consistent with the observations made for PMW loaded foams on this study. Dananjaya et al. [19] has discussed the development of 0.2 MPa tensile strength and 0.16 MPa modulus in waste mica filled NRLF up to 10 Phr. There, he claimed that the booming ability of reinforcement of mica in NRLF could be supported this behavior and therefore the results are well supported here as well.

### 3.7. Water absorption

The variation of water absorption of NRLF against latex type and PMW is shown in Fig. 9. In PMW filled foam rubber composites, water absorption capacity could be governed by both the water absorption ability of the filler and the nature of the rubber phase. NRLFs made out of creamed latex have more hydrophilic non-rubbers and a residual creaming agent. 7 % and 3 % higher water absorption capacity of creamed NRLFs than that of centrifuged latex-based foam rubber at 0 and 10 Phr PMW content respectively shown in Fig. 9 could therefore be elucidated by the presence of these higher content of hydrophilic substances in the rubber phase of the creamed latex-based foam composites. PMW contains silica which could absorb water through forming H-bonding between water molecules and OH groups of silica particles [84,85]. Therefore, 14 % and 10 % water absorption or water retention capacity increased with the increase in filler loading in centrifuged and creamed NRLF. This observation, however, cautions the selection of latex type and PMW for preparation of foam rubber for industrial applications due to the associated mold growth and discoloration issues.

### 3.8. Differential scanning calorimetry (DSC)

The differential scanning calorimeter (DSC) profiles, used to study the heat flow vs. the temperature from 198 to 773 K, of foam rubber composites are shown in Fig. 10. The glass transition temperatures derived from the DSC curves are tabulated in Table 6. Glass transition temperature ( $T_g$ ) gradually increases 6 °C for

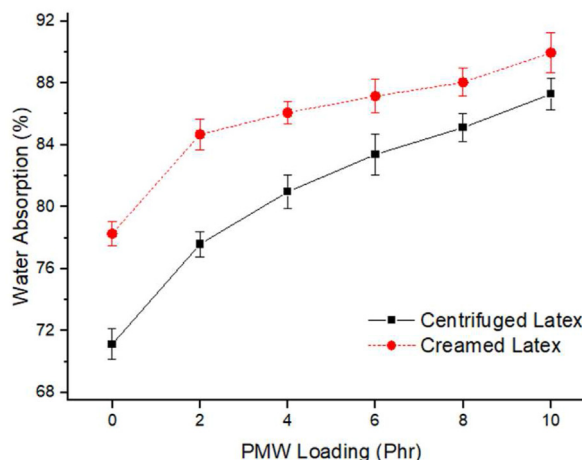


Fig. 9. Effect of PMW loading and latex type on water absorption.

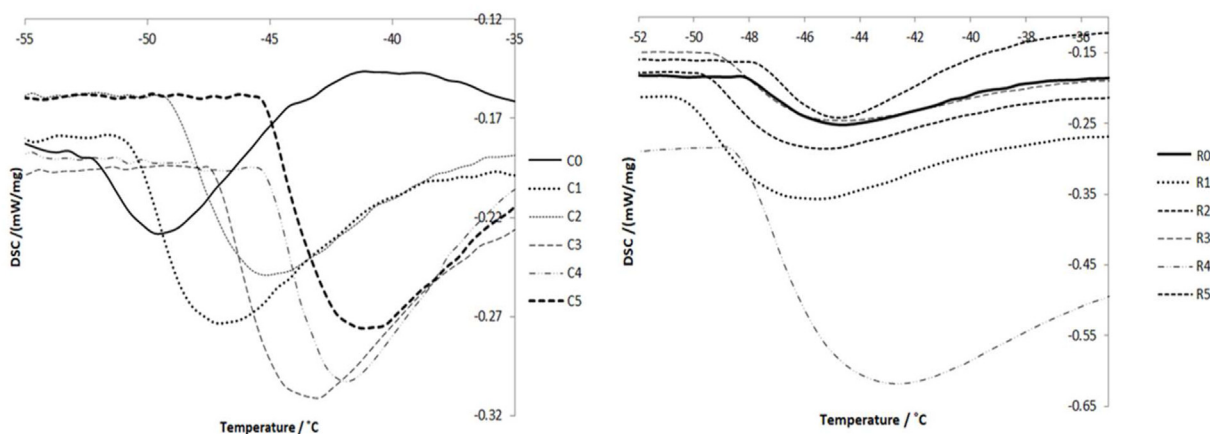


Fig. 10. DSC plots of PMW filled NRLFs.

Table 6  
DSC data of foam composites.

PMW loading/Phr	Centrifuged samples	Glass transition temperature/C	Creamed samples	Glass transition temperature/C
0	C0	-49.6	R0	-48.6
2	C1	-48.2	R1	-47.0
4	C2	-46.6	R2	-46.4
6	C3	-45.7	R3	-45.7
8	C4	-45.5	R4	-42.7
10	C5	-43.6	R5	-40.9

centrifuged NRLF and 7.7 °C for creamed NRLF with increasing filler loading confirming the reinforcing ability of mica [86]. This information supports the earlier observations made on increased density of the mica filled composites. Parvaiz et al. [62] has discussed 10.52 °C rise in  $T_g$  up to 10 Phr mica addition into poly(ether ether ketone) composites and concluded the heterogeneous nucleation effect of mica inside NRLF and as a result of that behavior of mica crystallinity increases by improving the  $T_g$ . 0.8 °C rise of  $T_g$  was identified by Martinez et al. [68] from 14.4 wt % to 25 wt % of mica in atactic polypropylene composites. He has shown the crystallinity of dense mica is comparatively higher than that of the polymer and it could uplift the  $T_g$  of entire composite structure for some extent. The slightly higher  $T_g$  value for foam rubber made out of creamed NRLFs probably may be owing to the presence of sodium alginate during the preparation of creamed latex [83]. A similar evaluation on the leaching method comparison and the effect of the leaching and NRS were studied by Ho et al. [87] and discussed the ability of leaching compounds and NRS to enhance the  $T_g$ .

### 3.9. Thermal conductivity

Fig. 11 shows the thermal conductivities of NRLFs with the increasing mica loading. Centrifuged NRLFs exhibit higher thermal conductivity than creamed NRLFs [4]. Larger cell sizes of creamed NRLFs as observed in SEM images suggest higher amount of free volume in the cell morphology reducing the thermal conductivity of creamed latex foams than its counterpart. In addition, higher mica to rubber ratio of centrifuged latex owing to its lower DRC values may also be contributed to this enhanced thermal conductivity of foam rubber prepared from centrifuged latex. It could be observed that the thermal conductivity increased in a range from  $0.10 \text{ Wm}^{-1}\text{K}^{-1}$  to  $0.16 \text{ Wm}^{-1}\text{K}^{-1}$  over the filler loading from 0 to 10 Phr. Heat transferring procedure inside a polymer material generally accomplished through polymer molecules. With the

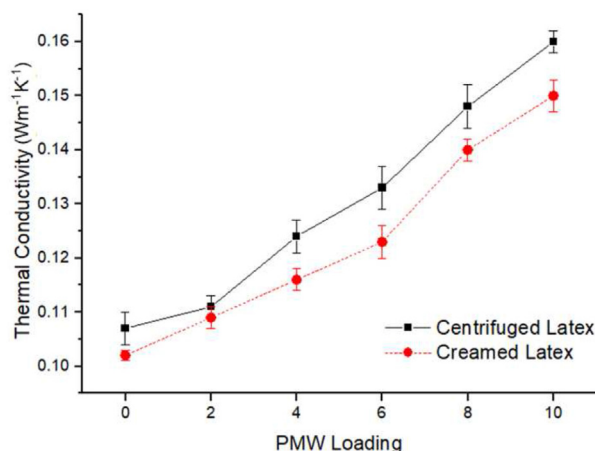


Fig. 11. Variation of thermal conductivity of controlled and mica filled creamed and centrifuged NRLFs.

incorporation of mica into the polymer matrix, micron level mica formed a thermal conductive network by its large surface area. Mica has relatively higher thermal conductivity ( $0.7 \text{ Wm}^{-1}\text{K}^{-1}$ ) than the thermal conductivity of natural rubber ( $0.5 \text{ Wm}^{-1}\text{K}^{-1}$ ) [4,88]. Therefore, with the addition of mica, thermal conductivities of mica incorporated foams were improved. A similar finding was reported by Tian et al. [89] experiencing  $0.2 \text{ Wm}^{-1}\text{K}^{-1}$  rise of thermal conductivity in brick mud mica composites from 0 to 80 wt % mica content. Zhang et al. [90] claimed  $0.19 \text{ Wm}^{-1}\text{K}^{-1}$  greater thermal conductivity in mica filled Poly ethylene glycol composites than that of virgin polymer. He concluded thermal release efficiency of the mica in the composites was relatively faster than the virgin due to its arrangements in polymer composites. However, it is reported that thermal conductivity of foam structures is considerably lower than thermal conductivity of dry natural rubber as a larger volume of air present in NRLF reduces the conductivity of heat [2,4,5]. Moreover, polymer materials consist of amorphous molecular arrangements and therefore, a uniform direction of heat transfer could not be clearly identified to the random orientation and greater degree of entanglements [91–94]. The heat transferring units in NR inside among polymer chains leads to relocation of heat and result a weak thermal conductivity as reported by Dong et al. [5] and Liu et al. [95]. Mica conducts heat in a great extent in composite structures and hence it is named as a heat conductive filler. Incorporation of heat conductive filler into natural rubber

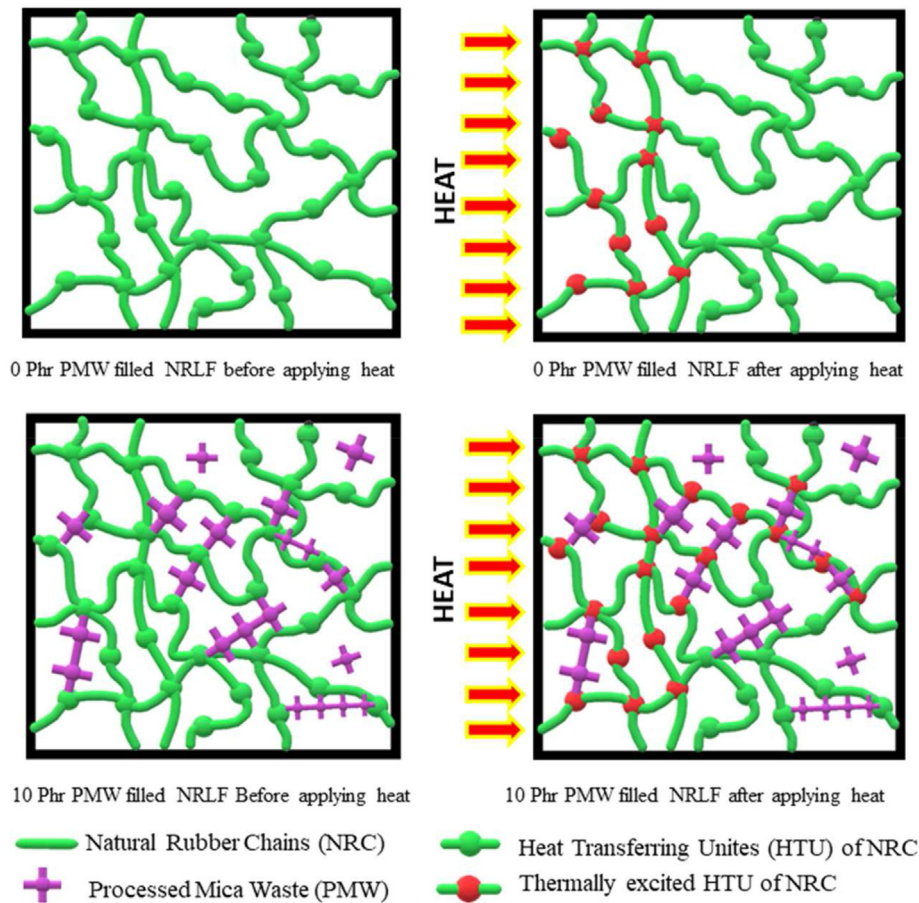


Fig. 12. Representation of thermal conductivity of 0 Phr and 10 Phr PMW filled NRLFs.

matrix with increasing filler contents accelerates formation of filler-rubber interactions and filler-filler interactions according to the Payne's Effect [95] as shown in Fig. 12. In this study also, mica acted as an agent of heat conductance among the polymer chains and enhanced the thermal conductivity with mica loading growth.

### 3.10. Electrical properties

Electrical conductivity was determined by the resistance between two points with a fixed distance a composite. It is interesting to note that both centrifuged and creamed NRLFs performed equally with respect to the electrical resistivity (Fig. 13). It was found that centrifuged latex contains higher non-rubber constituents. This observation implies that those non-rubber constituents would not be adequate enough to make a difference in the electrical properties of the corresponding composites. However, at 8 and 10 Phr mica loadings, creamed NRLFs shows a slight increasing trend in electrical resistivity. This could be explained by the morphology of the composites shown in SEM images. At higher filler loadings in creamed NRLFs, the cell structure has deformed and larger void spaces were formed with larger cell sizes causing an increase in electrical resistivity [43]. Saltas et al. [96] has reported the Fe and other transition metal (Mg and Ti) contents in mica also supports the electrical conductivity. Further, he has concluded biotite mica which is 17.1 % Fe<sub>2</sub>O<sub>3</sub>, 2.9 % TiO<sub>2</sub> and 7.5 % MgO in its composition performed 2–4 times magnitude of electrical conductivity than muscovite mica that consists of 5.3 % Fe<sub>2</sub>O<sub>3</sub>, 0.2 % TiO<sub>2</sub> and 0.0 % MgO. As shown in Table 4, here, waste consists of 4.8 % Fe<sub>2</sub>O<sub>3</sub>, 0.00 % TiO<sub>2</sub> and 19.1 % MgO. Therefore, results inferred that

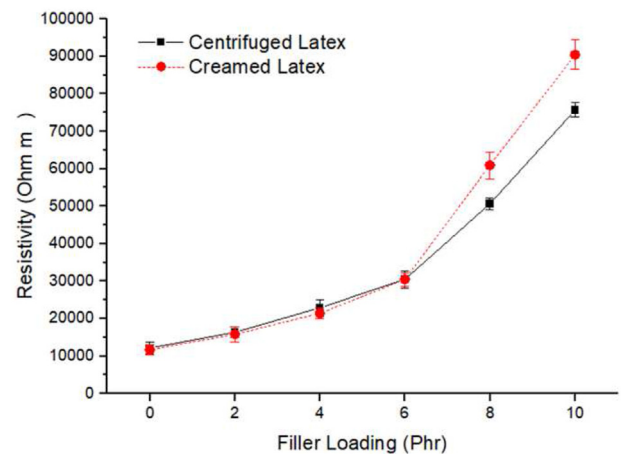


Fig. 13. Variation of electrical resistivity of controlled and mica filled creamed and centrifuged NRLFs.

waste mica also has considerable amounts of transitional metals along with high electrical conductivity. However, Khan et al. [97] has reported an reverse trend of electrical conductivity with mentioning 38.8 kV mm<sup>-1</sup> dielectric strength. He concluded the behavior due to surface and volume resistance of silica. Also, silica is a better material as an insulator due to its outer electrons are occupied in the covalent bonds of the diamond like framework [98]. The highest amount waste mica composition has been occupied by

SiO<sub>2</sub> (37.4 wt %) and approximately two times of MgO. Therefore, Zhao et al. [99] has observed a similar performance of electrical conductivity and he identified the electrical breakdown strength as 13 kV mm<sup>-1</sup>. This value of mica performed good electrical insulation property.

As shown in Fig. 13, electrical resistivity has enhanced by 8 times which means 8 times reduction of electrical conductivity up to 10 Phr PMW loading performing accelerating trend of electrical resistivity. As the filler loading increases, pore sizes and cell diameters are increased as shown in SEM images. Therefore, air voids percentage in the foam rubber increases with the increased filler loading yielding higher electrical resistivity [100]. Also, mica is good electrical insulating material [96] and its electrical resistivity has reported as 10<sup>13</sup> or 10<sup>14</sup> Ω m [63]. Deshmukh et al. [63] reported 3.21 × 10<sup>14</sup> Ω m increase of electrical resistivity in mica filled PVC composites peaked at 50 Phr mica content. Sukhmandan et al. [101] experienced 3.48 × 10<sup>3</sup> keV electric energy loss per micro meter in muscovite mica. Therefore, incorporation of electrical insulating material into the polymer matrix, the electrical conductivity property of mica filled composite structures became weakened.

#### 4. Conclusions

Overall, the processed mica waste improves most of the thermal and physical properties such as density, hardness, glass transition temperature, electrical resistivity and water absorption. However, mica loading reduces the gelling time and swelling index. Also, it was found that mica loading has a similar trend on the foam rubber properties investigated in this study for both types latex foam composites. NRLFs prepared using creamed latex exhibit 33 and 50 μm (width and height) cell diameter, 3 ibf hardness, 281 % swelling index improvements and 0.05 Wm<sup>-1</sup>K<sup>-1</sup> thermal conductivity, and 1 °C glass transition temperature (T<sub>g</sub>) reductions than centrifuged NRLF and indistinguishable electrical resistivity. With the addition of mica (0–10 Phr), both NRLF types showed a similar ascending trend in hardness (42 ibf), water absorption (16 %), T<sub>g</sub> (7 °C), thermal conductivity (0.54 Wm<sup>-1</sup>K<sup>-1</sup>), electrical resistivity (69 × 10<sup>3</sup> Ω m) with decreasing gel time (3 min) and swelling index (550 %). The 6 Phr PMW filled creamed NRLF (R3) is the most suitable NRLF for potential applications with a greater improvement of density (25 %), hardness (650 %), water absorption (11.53 %), glass transition temperature (4.3 %), thermal conductivity (21.57 %), electrical resistivity (200 %) and reduction in swelling index (23.57 %) than the controlled samples. Due to cell distractions and PMW agglomeration, 8 and 10 Phr PMW loaded samples are not that suitable for practical applications. It could be concluded that mica could be used as an effective filler for the preparation of NRLF using either centrifuged latex or creamed latex, where the latter showed its potential as a good competitor for high energy-consuming and costly centrifuged latex for some of the selected industrial applications namely green rubber toys, mattresses and pillows along with other cushion manufacturing industries.

#### Conflicts of interest

The authors declare that there is no conflicts of interest.

#### Data availability

Data will be made available on request.

#### CRediT authorship contribution statement

**Vimukthi Dananjaya:** Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Resources, Writing – original

draft, Writing – review & editing. **Yashoda Somarathna:** Data curation, Investigation, Writing – review & editing. **Susantha Siriwardena:** Conceptualization, Formal analysis, Project administration, Supervision, Writing – review & editing. **Narayana Sirimuthu:** Formal analysis, Supervision, Writing – review & editing. **Laleen Karunanayake:** Formal analysis, Project administration, Supervision, Writing – review & editing. **Chamil Abeykoon:** Data curation, Formal analysis, Investigation, Supervision, Validation, Visualization, Writing – review & editing.

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