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Development of a continuous-flow system for microwave-assisted extraction of pectin-derived oligosaccharides from food waste

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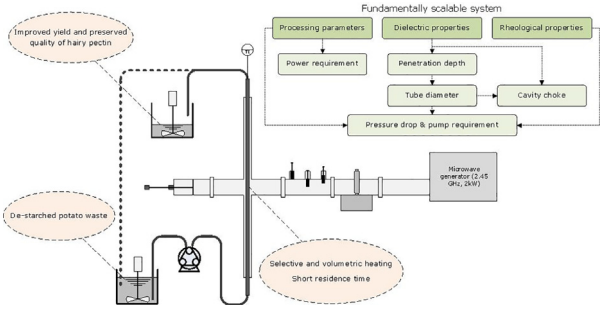
HIGHLIGHTS

- A scalable, continuous, and "green" hairy pectin extraction process developed.
- Microwave-Assisted Extraction successfully implemented in continuous operation.
- Improved starch reduction achieved by sieving procedure.

GRAPHICAL ABSTRACT



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ABSTRACT

This paper addresses the current lack of a scalable process for the extraction of "hairy" pectins to upcycle biomasses, by describing the design methodology, building and testing of a continuous microwave-assisted process for potato waste pectin extraction. Hairy pectins have been shown to present prebiotic activity. Conventional pectin extraction methods are not suitable to produce them, as they lead to degradation of hairy regions, in part due to long heating times. Microwave heating is considered an alternative due to its selective and rapid heating. The 2 kW single mode system developed in this study achieved good temperature control of ± 2.5 °C, and a stable target temperature in ≈1 min processing time at a feed flow rate of 250 mL min^{-1}. Pectin yields of 40–45% (indicated by the galacturonic acid content) were achieved, with a feed residence time of only 0.81 s followed by 20 min cooling-down under stirring. These yields were 59.75% higher on average than control experiments in batch conditions or under continuous heating. This indicates that a short heating time is sufficient to allow pectin hydrolysis, after which the rate limiting diffusion step can proceed during cool-down, minimising pectin degradation caused by prolonged exposure to high temperatures. The presence of hairy pectin regions in the extract was confirmed by neutral sugar analysis. The high starch content in potato necessitated a de-starching procedure. A sieving procedure was implemented, which removed more starch than enzymatic de-starching, resulting in a higher purity pectin extract and the ability to collect starch as a separate value stream.

Abbreviations: Y_E, extract yield; Y_{GalA}, galacturonic acid yield; Rha, rhamnose; Glu, glucose; Gal, galactose; Ara, arabinose; Xyl, xylose; D-GalA, D-galacturonic acid; HG, Homogalacturonan; RG-I, rhamnogalacturonan-I; RG-II, rhamnogalacturonan-II; MAE, Microwave Assisted Extraction; IC, ionic chromatography; ε′, dielectric constant; ε″, loss factor; Dp, penetration depth; SP, single-pass route; RE, recirculation route; CWE, conventional water extraction; POS, pectin-derived oligosaccharides

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This work proves the suitability of microwaves to be used as a fast heating method to extract pectin from biomass, avoiding degradation, using a scalable continuous mode of operation.

1. Introduction

The lack of a clean, low environmental impact method to extract "hairy" pectins at scale is a major bottleneck in the development of novel products for food and pharmaceutical applications from plant-based food processing co-products. The work presented in this paper demonstrates, for the first time, a scalable process to extract hairy pectin from potato pulp. This will enable testing of the extracts for a wide range of bioactivities and a route to scale-up, information that is vital to attract investment in the commercialisation of this process.

Around $1x10^6$ tonnes of potato pulp are produced every year in Europe as a by-product of the potato starch industry [1]. Currently, potato pulp mainly goes to anaerobic digestion or is used as animal feed, at a price that barely covers the processing and transport expenses [1,2]. If valuable compounds could be isolated from this material, they could present an additional revenue stream for potato businesses.

Pectins are a group of complex heteropolysaccharides found in the primary cell wall and middle lamella of terrestrial plants. D-galacturonic acid (D-GalA) has been described as the basic constituent of all pectins. There is still no consensus about pectin structure, but D-GalA is traditionally described as present in three polymeric forms: homogalacturonan (HG), a linear homopolymer of α-1,4-linked-D-GalA, which can be methyl-esterified and acetylated; rhamnogalacturonan-I (RG-I), a repeating disaccharide of D-GalA and rhamnose (Rha), which frequently has neutral sugar side-chains associated with it, and rhamnogalacturonan-II (RG-II), consisting of a HG backbone with numerous complex side chains containing Rha and other neutral sugars [3]. Due to their highly branched nature, RG-I and II regions are often referred to as "hairy" pectin [4], while HG-rich regions are known as "smooth" pectin. The physicochemical properties and hence potential applications of pectin are heavily influenced by these variations in structural features.

Commercially available pectin is rich in smooth pectin, widely used in the food and pharmaceutical industries for its excellent hydrocolloidal properties. It is produced mainly from citrus peels, apple pomace and sugar beet [5]. Potatoes have traditionally been considered a poor source of pectin, since it is highly branched and therefore has poor gelling properties. However, recent work has linked hairy pectin to prebiotic applications, as it might have beneficial effects on the human gut microbiome [6,7]. It has been shown to promote growth of bifidobacteria *in vitro*, inhibit adhesion of pathogenic bacteria and toxins and stimulate apoptosis of human colonic adenocarcinoma cells. There is an increased interest in the identification, evaluation and commercialisation of new products with improved functional properties [8], especially prebiotics. The global prebiotics market size was over U$ 2.90 billion in 2015 and is expected to grow around 12.7% by 2025 [9].

Conventional industrial pectin extraction is carried out in stirred batch reactors using mineral acids at pH values of around 1–3, temperatures of around 80 to 100 °C, and residence times of around one hour [10]. This process is not suitable for hairy pectin extraction, since the side chains are hydrolysed [11,12]. Pectins experience degradation (depolymerisation and demethoxylation reactions) when undergoing thermal treatment. Longer holding times and high temperatures increase the extent of degradation. Depolymerisation mechanisms are acid hydrolysis, which occurs at pH values of 2 or below, and β-elimination, which is favoured at pH values between 3 and 7 [12,13]. It is therefore clear that for hairy pectin to become a commercially viable product, a new industrial-scale pectin extraction process is required. This would need to operate under neutral or alkaline conditions (to preserve the neutral sugar side chains) and ideally not operate at elevated temperatures for prolonged periods (to minimise degradation via β-elimination). Over the last decade, a variety of alternative pectin extraction techniques and novel sources of pectin have been explored, and these efforts have been reviewed elsewhere [14,15]. The overall conclusion about these emerging techniques (acid extraction aided by chelating agents, alkaline extraction and enzymatic extraction) is that, even though many studies report products with improved functional properties, few considered the effect of extraction method on pectin structure [16], and scale-up is not addressed. Scale-up is vital to progress in this field, as without sufficient samples to allow further bioactivity testing and *in-vivo* trials, commercial partners will not commit investment.

Microwave-Assisted Extraction (MAE) is emerging as a potentially scalable extraction process. Among its advantages, shorter processing times, significant increase in yield, lower energy consumption and decreased use of solvents have been cited [17]. Microwaves can penetrate uniformly throughout the volume of material instantaneously (known as volumetric heating), which allows microwave processes to be fast, continuous, compact and flexible in operation [18]. Microwaves also heat different components of heterogeneous systems at different rates (selective heating). Volumetric and selective heating mechanisms, which are unique to electromagnetic heating, can also lead to temperature differences between the solvent and the biomass, and this can also lead to enhanced extraction through temperature-induced diffusion [19].

Some authors have recently been successful in extracting pectin by using MAE [20–24], employing sugar beet pulp and orange peel as pectin-containing sources. However, little attention has been paid to the extraction of potato pectin. The few recent works published report acid, alkaline and enzymatic extractions [1,25–27]. One study by Khodaei, Karboune and Orsat [2] did investigate MAE of potato pectin, although in combination with alkaline extraction. However, their purpose was to optimize the extraction by varying several reaction parameters simultaneously, with a view to laboratory-scale optimisation rather than process scale-up.

The aim of this study was to develop a scalable and environmentally friendly process for extracting hairy pectin from potato pulp, valorising this co-product. The process was designed to overcome the three main limitations identified in pectin extraction, namely scalability, hairy pectin degradation and environmental concerns. The main challenge in scaling-up the batch processes described above is that as the reactor size increases, the residence time increases, leading to increased pectin degradation. Microwave heating was therefore selected for its ability to rapidly heat process streams, enabling a continuous process to be designed, vastly reducing processing times and hence preserving hairy pectins' neutral sugar side chains. Water was selected as the solvent, as opposed to alkali. The environmental credentials of the process could therefore be considered good from the point of view of avoiding the production of caustic waste streams and also the ability of microwaves to be powered via renewable electricity sources (rather than fossil fuels in conventional heating processes). The work was carried out in two phases. First, small-scale batch exploratory experiments were run to investigate the feasibility of the water-only microwave-assisted extraction of hairy pectin and to evaluate the influence of processing parameters on the extraction yield. The outcome was used to inform the design and development of a continuous-flow pectin extraction system. Different operational modes were studied and the presence of hairy pectin in the extracts was evaluated using analytical techniques. Using this design methodology, the outcome of this work is a scalable process for the extraction of hairy pectin from potato pulp, which will feed into

the design of other liquid-based biomass valorisation processes.

2. Materials and methods

2.1. Materials

Industrial potato pulp, obtained as a co-product of a protein extraction process, was kindly provided by B-Hive Innovations (Lincoln, UK). Table 1 summarises potato pulp average chemical composition (analytical data provided by B-Hive Innovations). Isopropanol (IPA) was from Fisher Scientific (Loughborough, UK). α-Amylase was from Sigma (Gillingham, UK, EC 3.2.1.1, *Bacillus licheniformis*). Galacturonic acid (GalA) was from Sigma. Sugar standards were all analytical grade.

2.2. Overview of pectin extraction and recovery process

Fig. 1 shows the flow of unit operations that was developed to extract and recover pectin from potato pulp. The feasibility of water-based microwave-assisted extractions for the intended purpose, along with up- and downstream processing requirements, were evaluated in small-scale batch systems. These exploratory experiments were used to develop pectin extraction and recovery protocols and to evaluate the influence of processing parameters (heating rate, holding times, input power and feed concentration), in order to provide a basis of design for the continuous-flow experiments. A continuous-flow microwave process was then designed based on the outcomes of the batch-scale experiments. The following sections explain these unit operations in more detail.

2.3. Reduction of the starch content

Table 1 shows that the potato pulp was rich in starch. Available carbohydrates are sugars and starch, and sugars content is < 0.1%. During experimentation, it was found that a reduction of the starch content was required to address two challenges. The first one was gel-formation as a result of the temperature increase during microwave-assisted extraction; a feed in gel form did not flow across the continuous system due to the high viscosity [28]. The second issue pertained to the analytical characterisation. A high content of starch and thus of glucose (Glu) when analysing the monosaccharide composition interfered with the identification of the pectin-derived sugars during chromatographic analyses (see Supporting Document S.4). Therefore, the suitability of using a starch removal procedure prior to or during the MAE was tested. Two different alternatives were evaluated. As a first approach, an enzymatic hydrolysis using α-amylase (enzyme-treated extracts) at an E/S ratio of 0.04 (v/w) was used, as it was found to be the most common laboratory-scale procedure reported in the literature [2,25]. As a second alternative, a sieving process (sieved extracts) was developed taking into account the size of the potato starch granules [29], using a 150 μm mesh size. The degree of success of the de-starching procedure was tested by analysing the ionic chromatography (IC) glucose peak in the enzyme-treated extracts, the sieved extracts and the sieved permeate (see Supporting Document S.4).

2.4. Batch Microwave-Assisted Extraction (MAE) experiments

The aim of the batch exploratory experiments were to investigate the feasibility of the water-only microwave-assisted extraction of hairy pectin from potato pulp and to evaluate the influence of processing parameters on the extract yield (Y_E) to inform the design of the continuous-flows system. The batch MAE experiments were performed in an Anton Paar Monowave 400 (St. Albans, UK), a single mode cavity operating at 2.45 GHz. This piece of equipment does not show the reflected power, and thus the absorbed power cannot be calculated. All batch exploratory experiments started with the same initial temperature of 37 °C, and were performed in 15 mL Monowave glass vessels under constant stirring at 600 rpm. The processing parameters studied in this set of experiments include holding time, feed concentration, and input power. The thermal treatment consisted of a heating ramp, during which the temperature was increased to 90 °C, and then held for required time as illustrated in Table 2. The target temperature of 90 °C was chosen based on results from previous work [22]. Feed concentration was assayed at 25 and 50% w/w, and input power at 400 and 800 W. Finally, the holding of the target temperature for 20 min (holding) was compared with heating for 1 min, and then allowing 19 min to elapse without applying any input power to the sample (flash), so that the temperature was reduced to 75–80 °C. The purpose of this was to simulate and evaluate the extraction under fast heating with short residence time, which is expected from a continuous-flow microwave heating system. For clarity, Table 2 summarises the different batch experiments, which were all performed in triplicate.

2.5. Design of the continuous-flow microwave system

In order to provide the rapid and controlled heating desired to minimise pectin degradation, a single mode cavity design was selected. This cavity type enables the application of an even and predictable electric field to the sample, thereby achieving volumetric heating of the whole sample as it passes through the cavity. In order to achieve an effective design, knowledge of how the material interacts with microwaves is required. The interactions between the electromagnetic field at certain frequency and a nonmagnetic material are defined by the dielectric properties of the material; namely its dielectric constant and loss factor. The dielectric constant, ε', defines the material's ability to be polarised and to store energy, while the loss factor, ε'', defines its ability to dissipate the electromagnetic energy into heat [30,31].

A systematic approach was followed for the process design, taking into account the dielectric properties and their effect on the penetration depth and selection of the flow diameter, rheological properties and their effect on the pressure drop and pumping requirement, and the choke design to avoid microwave leakage. Fig. 2 summarises the systematic approach followed for the design of the continuous-flow system. The design was based on a feed flow rate of 250 mL min^{-1}, feed initial temperature of 37 °C and a target outlet temperature of 90 °C.

2.5.1. Flow diameter

The choice of the flow diameter is restricted by the penetration depth of the electromagnetic field into the material, which is determined by the dielectric properties of the material. The dielectric properties of the potato pulp solution were measured using a dielectric probe kit (Keysight 85070E) connected to an N5232A PNA-L network analyser. The dielectric constant and loss factor of the potato pulp as functions of temperature are displayed in Fig. 3. Both the dielectric constant (ε') and loss factor (ε'') increase with temperature up to 90 °C.

Fig. 3 also displays the penetration depth (D_p), which was calculated using the measured dielectric properties according to the equation presented in Section S.1 in the supporting document. It is defined as the distance from the surface of the material at which the microwave power flux drops to 1/e (~0.368) of its surface value [32]. It can be seen that

Table 1
Industrial (B-Hive Innovations) potato pulp chemical composition (analytical data provided by B-Hive Innovations).

Analysis	Result (g/100 g)
Moisture	54.9
Crude protein (Dumas)	0.9
Ash	0.8
Available carbohydrates	39.70
Total sugars	< 0.1
Total fat	< 0.5
Total dietary fibre	3.7

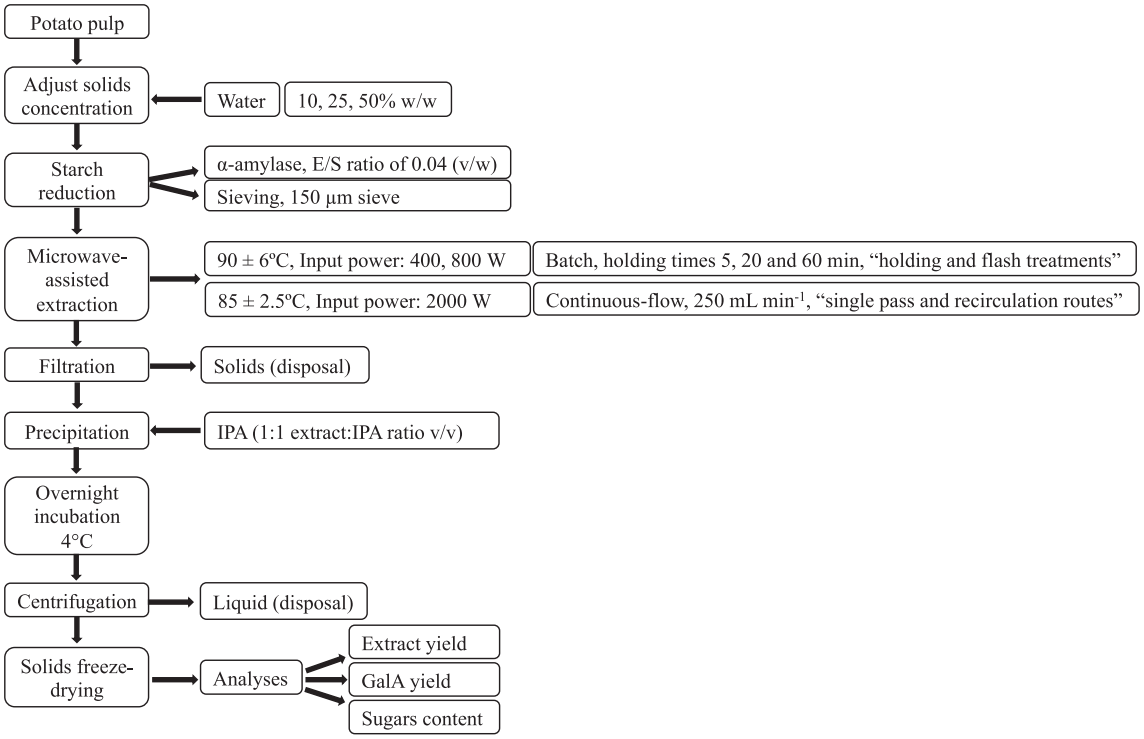

Fig. 1. Overview of pectin extraction and recovery process.

Table 2
Experimental conditions during batch exploratory experiments.

Parameter varied	Potato pulp concentration (% w/w)	Microwave power (W)	Holding time (min)
Holding time (min)	25	400	5/20/60
Pulp concentration (% w/w)	25/50	400	5
Microwave power (W)	25	400/800	5
Holding/flash (min)	25	400	20/1 + 19

the D_p of the microwaves in the potato pulp mixture varies between 13 and 16 mm over the temperature range between 20 and 90 °C. Based on this, a flow diameter of 10 mm was chosen. A flow diameter larger than the D_p would lead to significant heating heterogeneity across the radial direction. Based on this diameter and the 250 mL min^{-1} flow rate, the average velocity of the potato pulp mixture inside the cavity is 0.053 m s^{-1}.

In order to calculate the pressure drop, the viscosity of the potato

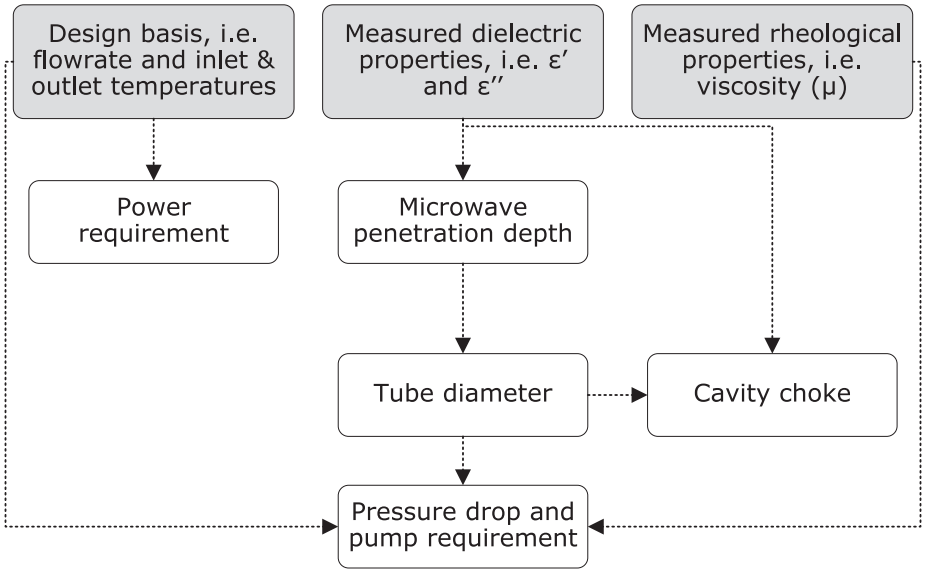

Fig. 2. Approach followed for the design of the continuous-flow system.

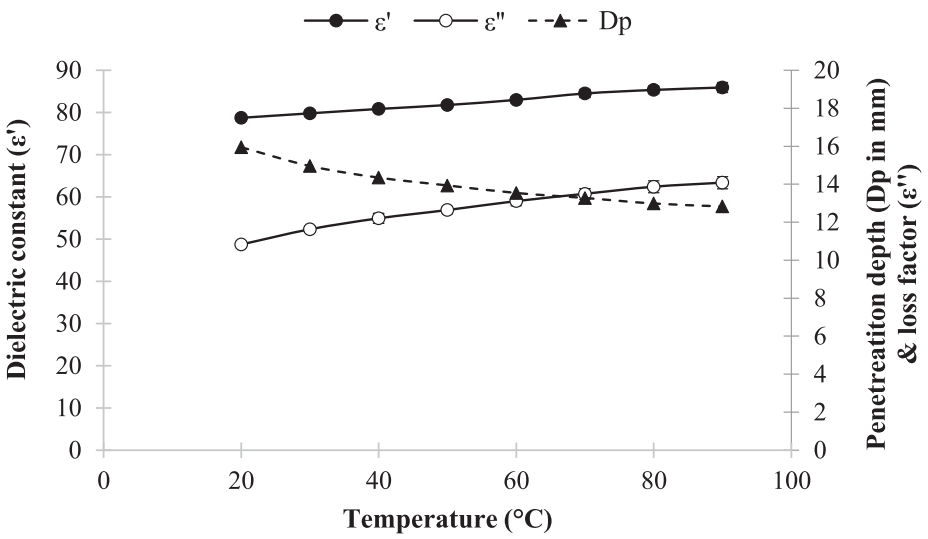

Fig. 3. Dielectric properties of the 10% w/w potato pulp mixture as functions of temperature.

pulp solution was measured using a cone and plate viscometer (Anton Paar Modular Compact Rheometer MCR 302). The results of the viscosity measurement, which can be found in Section S.2 in the supporting document, show that the potato pulp slurry exhibits a non-Newtonian shear-thinning behaviour. The pressure drop was calculated accordingly, and found to be 0.103 bar m^{-1}. The calculations are detailed in Section S.2 of the supporting document.

2.5.2. Choke design

One of the challenges when designing continuous-flow microwave-heated processes involving fluids with high permittivity is electromagnetic choking at the inlet and outlet. The aim of the choke is to keep the microwave power flux at or below the maximum permitted leakage level, which is 5 mW cm^{-2} [33]{Regulations, 2016 #84}. The simplest form of choking is the 'cut-off attenuation choke', which involves using a conductive tube long enough for the electromagnetic field to decay to the permitted level. However, such a choke requires the bore diameter to be lower than a cut-off diameter. The cut-off diameter was calculated at the inlet ($\varepsilon' = 79$) and outlet ($\varepsilon' = 86$) conditions and found to be 8.1 and 7.7 mm respectively as detailed in Section S.3 in the supporting information document. Both are lower than the chosen flow diameter, which means that the cut-off choke would not be suitable for the current system.

Choking could, alternatively, be based on the attenuation within the load due to the energy conversion into heat. The attenuation factor due to this 'load-loss' was calculated based on dielectric properties at the inlet and outlet conditions as detailed in Section S.3 in the supporting information document, and was found to be 22.1 and 17 cm at the inlet

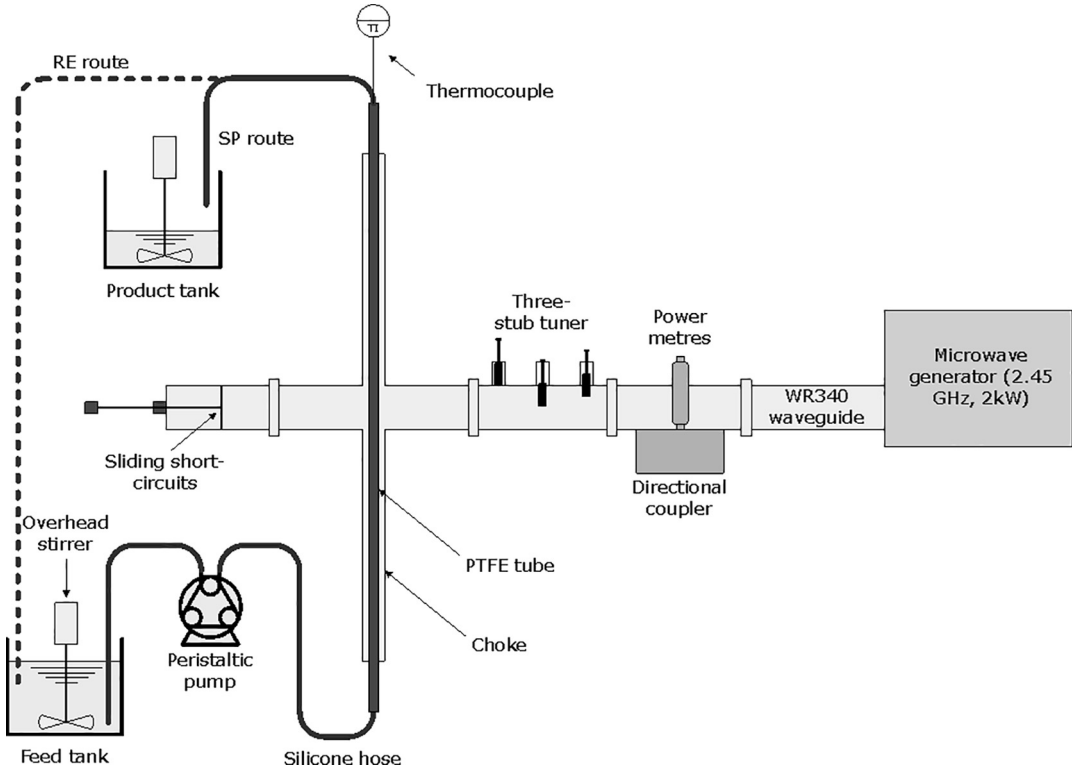

Fig. 4. Continuous-flow microwave processing rig assembly and components.

and outlet, respectively. Therefore, a length of 25 cm was used for both the inlet and outlet choke in the current study.

2.5.3. Power requirement

The microwave power requirement was estimated using Eq. (1).

$$P = \dot{m} c_p (T_{out} - T_{in}) \quad (1)$$

where P is the required microwave power (kW), $\dot{m}$, is the mass flowrate of the feed (4.17×10^{-3} kg s^{-1}), c_p is the specific heat capacity of the slurry (4.18 kJ kg^{-1} $°C^{-1}$), and T_{in} and T_{out} are the inlet (37 °C) and outlet (90 °C) temperature respectively (°C). Substituting these values into Eq. (1) gives a power requirement of 0.92 kW.

It should be noted that Eq. (1) assumes a constant specific heat capacity over the study's range of temperature and that the potato pulp mixture absorbs all the microwave power. Given that the mixture used in the continuous-flow experiments was 90% water, as will be shown later, it is also assumed that specific heat capacity of the mixture is same as of pure water. Given that dry potato has a specific heat capacity lower than that of water (~1.8 kJ kg^{-1} $°C^{-1}$ for dry potato [34]), the overall specific heat capacity of the mixture and the power requirement would be up to 6% lower than that of pure water. Furthermore, Eq. (1) does not account for heat losses to the surroundings. Therefore, the calculated power can be considered as the minimum power requirement. A microwave generator with an input power in the range 0.2 to 2 kW was used for this system.

2.5.4. Full system

Fig. 4 depicts the continuous microwave processing rig assembly, naming the main components. Microwaves were supplied using a 2 kW, 2.45 GHz microwave generator (Sairem, Neyron, France). Power sensors (Agilent, Santa Clara, CA, USA) connected to a directional coupler were used for measuring forward and reflected power and frequency. Standard WR340 waveguides were used for transmitting the microwave power. The cavity is a single-mode applicator with circular chokes (20 mm ID and 25 cm length each) attached to the top and bottom faces. A 3-stub tuner and sliding short-circuits were used for impedance matching, which helps to improve the microwave power delivery efficiency. The 3-stub tuner allows for amplifying the electric field intensity in the cavity by establishing a state of resonance. The sliding short-circuits enables the standing wave configuration within the waveguide to be shifted to maximise power delivery to the load. The net absorbed power was determined by subtracting the reflected power from the input power. It was passed across the cavity in a PTFE tube (10 mm ID, 20 mm OD and 60 cm length) connected to flexible silicon pipes at the inlet and outlet. The mixture was pumped through the system using a peristaltic pump (Watson - Marlow 604 U/R, Wilmington, MA, USA) with silicone tubing. The feed temperature was measured using a thermometer, while the outlet temperature was measured using a K-type thermocouple (RS components, Corby, UK) inserted at the outlet of the choke. A microwave leakage detector was used to monitor and avoid any microwave leakage above the maximum permitted level during the experiments.

2.5.5. Continuous-flow experiments

The operating parameters of the continuous-flow system were selected based on the outcomes of the batch exploratory experiments. The target temperature was lowered from 90 °C to 85 °C, in order to prevent overheating, considering that the thermocouple was not inserted immediately after the applicator, but after the choke. Also, the feed concentration had to be lowered to 10% w/w due to downstream processing issues (as explained in Section 3.2). A total volume of 4.5 L of feed was used in each experiment. The potato pulp mixture was continuously stirred in the feed tank using an overhead stirrer, in order to ensure homogeneity. The temperature was controlled manually using the input microwave power. The residence time of the potato pulp within the cavity (heating region) was only 0.81 s.

Two different operation modes were tested (see Fig. 4), resembling the "holding" and "flash" batch experiments:

(a) Single-pass route (SP): After microwave-assisted heating inside the cavity, the treated pulp was collected in a beaker. When all the feed had passed through the cavity, the treated pulp in the beaker was allowed to cool down under stirring for 20 min, reaching a temperature of ≈70 °C. After that, it was further cooled-down to 50 °C in cold water by immersion, and immediately subjected to the downstream processing.
(b) Recirculation route (RE): Once the feed reached the target temperature, the sample was sent back to the feed tank and reprocessed like this for 20 min, maintaining the target temperature. Then, all the treated pulp was cooled-down to 50 °C and subjected to downstream processing.

Continuous-flow experiments were performed in duplicate. The continuous-flow experiments were performed with both enzyme-treated and sieved potato pulp, to compare the effect of starch removal method on the results.

2.6. Conventional water extraction experiments

In order to have a reference with which compare the quality of the extracts (analyses explained in Section 2.8), conventional water extractions (CWE) using a water bath were also performed with the potato pulp, keeping the same parameters than in the MAE, but with a 1 h treatment time, which was selected to be consistent with reports in the literature and industrial practice [10]. Also, because of available equipment limitations, the total volume per experiment was of 1 L. CWE experiments were performed with both de-starching procedures and in duplicate.

It is noted that the purpose of the CWE was not to compare the fundamental difference between microwave heating and conventional heating in pectin extraction (which has been done elsewhere [22]), but to compare the results of the fast heating rate of the continuous process with those of a process with a comparable heating time to an industrial-scale batch process.

2.7. Pectin recovery

The downstream processing to isolate the pectin-containing extracts after the MAE (or the CWE) was performed as elsewhere in the literature [29,35]. Briefly, the thermally treated pulp was filtered with Fisherbrand qualitative filter paper QL 100 (Fisher Scientific, Loughborough, UK) using partial vacuum, in order to remove the solids. Next, the liquid extract was treated with IPA at a 1:1 ratio (v/v) and incubation at 4 °C overnight, so as to precipitate pectin oligosaccharides. Finally, the pectin-containing extract was recovered by centrifugation (Sorvall ST 40R, Thermo Scientific, Waltham, MA, USA) at 3900 rpm during 20 min, and freeze-dried (LycoDry compact, Mechatech, Bristol, UK) for further analyses.

2.8. Analytical procedures

Three different parameters were used to evaluate the extraction process (see Fig. 1), namely the extract yield (Y_E), the sugar composition and content, and the GalA yield (Y_{GalA}).

Y_E was calculated using Eq. (2):

$$Y_E(\%) = \frac{\text{Weight of dry extract}}{\text{Weight of dry feed}} \times 100\% \quad (2)$$

In order to assess the quality of the extracts, the GalA content and the sugar composition were tested. The GalA content of the extracts was measured as an approach to quantify pectin, following the method

developed by Filisetti-Cozzi and Carpita [36], with some variations. Freeze-dried potato pulp extracts (20 mg) were dissolved in deionised water at a concentration of 1 mg/mL, using Pyrex boiling tubes, after which 40 µL sulphamate and 2.5 mL of concentrated sulphuric acid were added, and the mixture heated at 99 °C in a water bath for 20 min. The solution was then cooled under running water and 80 µL of an m-hydroxydiphenyl solution (0.15% m-hydroxydiphenyl in 0.5% NaOH solution) was added, and the samples left to develop a pink colour over 5–10 min. The absorbance of the solutions was then determined by UV–Vis chromatography at wavelength 525 nm (Jenway, Cole Palmer, Staffordshire, UK). The zero absorbance reference was set with deionised water, and the standard solution was made with GalA concentrations ranging from 0 to 97 mg L^{-1}.

The sugar analysis was used to evaluate the content in neutral sugars, indicator of pectin hairy regions (see Section 1). It was performed by ionic chromatography (IC) with a Dionex ICS-3000 system (Thermo Fisher, Loughborough, UK). The freeze-dried pectin extract was hydrolysed with concentrated sulphuric acid using the same method as the GalA assay. After the pre-hydrolysis, 100 µL of supernatant were pipetted into 50 mL Pyrex boiling tubes and then diluted with 10 mL of 10 mM NaOH. 1 mL of the hydrolysed solution was added into Dionex vials for sugar analysis. The Dionex setting used a CarboPac PA20 column (3 × 150 mm, BioLC, Thermo Fisher) and software Chromeleon. 10 mM NaOH was used as eluent and 200 mM NaOH as the mobile phase; retention gradient and time as −10 ~ −5, 0.5 mL/min, < 3000 psi, 100% eluent; −5 ~ 14, 0.4 mL/min, < 2300 psi, 100% mobile phase; stand-by, 0.1 mL/min, 700 psi. Mixtures of sugar standards (L-rhamnose, L-arabinose, D-galactose, D-glucose and D-xylose) at various concentrations (1–20 mg/L) were used as external standards for identification and quantification. Sugar analysis was also used to verify that only the starch was removed during the sieving procedure. For that, the permeate solids from the sieving process were collected and freeze-dried and analysed by IC as well (supporting document S.4).

Finally, Y_{GalA} was calculated with Eq. (3):

$$YGalA(\%) = \frac{\text{GalA content in extract}}{\text{GalA content in feed pulp}} \times 100\% \quad (3)$$

3. Results and discussion

3.1. Temperature regulation

To assess the temperature control during batch experiments, the highest yield experiment (holding for 20 min, 25% w/w concentration, 400 W input power) was monitored for the temperature variation. More detailed results are shown in Supporting Document S.5. The target temperature was 90 °C. The batch microwave equipment took 4.5 ± 0.5 s to reach the target temperature. After the heating ramp, there was some overheating of the sample, reaching 94 °C. During the first 1.5 min of holding time, the temperature fluctuated by ± 6 °C, but stabilised to 90 °C ± 2 °C from that point onwards. Once the target temperature was achieved, the input power never exceeded 85 W.

Fig. 5 shows the potato pulp temperature measured with the thermocouple and the power recordings during the continuous-flow SP and RE experiments, for both enzyme-treated and sieved potato pulp. The absorbed power was calculated by subtracting the reflected power from the input power (see Section 2.4). For both operational modes (SP and RE), it took on average 1.05 ± 0.15 min to achieve a stable target temperature, at a feed flow rate of 250 mL min^{-1}. From the moment the target temperature was achieved, the temperature was controlled manually by adjusting the input power. It resulted in good control, varying only ± 2.5 °C maximum.

The initial input power was set to 2000 W. Once the target temperature was reached, for SP experiments the power was lowered down

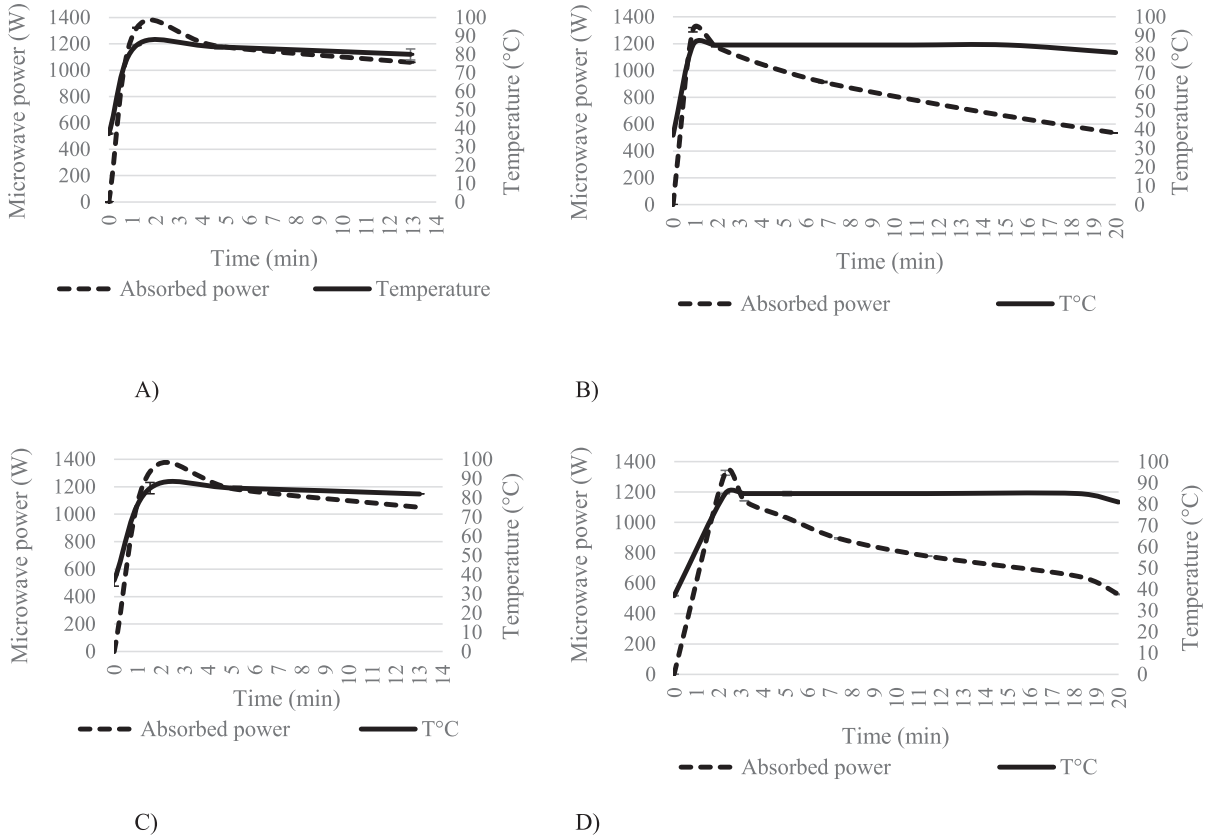

Fig. 5. Absorbed power and temperature recordings against treatment time during continuous-flow MAE experiments. A) Enzyme-treated pulp, single pass (SP). B) Enzyme-treated pulp, recirculation (RE). C) Sieved pulp, SP. D) Sieved pulp, RE. Error bars show differences between replicates.

to 1600 W to maintain the set temperature, while 800 W were reached during RE experiments. In all the experiments, a good degree of temperature control was achieved using these settings. Once steady state was achieved, the heating rate and residence time were calculated to be 65.4 °C s^{-1} and 0.81 s respectively.

These results confirm that the continuous process developed here is able to achieve (in fact exceed) the fast heating rate, short residence time and good temperature control of the batch experiments, and this is promising for the overall scalability of the process.

3.2. Extract yield

Fig. 6 summarises the extract yields (Y_E) obtained for the different conditions assayed during batch exploratory experiments.

As it can be seen in Fig. 6, both the holding time and the microwave input power had significant effect on Y_E. The intermediate holding time (20 min) worked better than shorter or longer treatment, and the yield was higher for an applied power of 800 W compared with 400 W. These results are consistent with the literature in terms of general trends, although direct comparison is not possible as this is the first study (to our knowledge) that has extracted pectin from potato pulp using only water as the solvent and applying microwaves. It is generally accepted that there is an optimal duration for the thermal extraction of pectin, beyond which thermal degradation occurs. Pectin has a thermolabile nature and therefore extended exposure to high temperatures results in depolymerisation and de-esterification reactions [37]. The small polymers do not precipitate with the IPA treatment [38] and thus the yield appears decreased. Increasing the microwave power from 400 W to 800 W increased the yield. This could be attributed to the shorter heating time [39,40] or the effects of microwave selective and volumetric heating on mass transfer [41,42]. Assayed feed concentrations did not have a great impact on Y_E. These concentrations are considerably higher than those used in other works about MAE of pectin, of around 8% w/v [21,43]. The reasoning behind it was to assay as high solids concentration as possible, thinking about industrial application. Increasing the feed concentration from 25 to 50% resulted in limited reduction in Y_E, a result in line with general trends. It is generally accepted that low solvent:solid ratios can reduce extraction yield through limitations in solubility, viscosity and a reduction in osmostic potential between the plant matrix and the solvent. Nevertheless, low solids concentration and high solvent content negatively affect the economics of the process and mean higher wastewater disposal or drying costs [38,44]. For the continuous-flow experiments, the effect of feed concentration on yield could not be tested. The viscosity of the solution (supporting document S.2) at concentrations higher than 10% w/w affected the downstream processing. The potato slurry was so thick that it could not be processed for filtration with the equipment available, and also stirring was not possible. Therefore, the feed concentration in a continuous process will be determined by processability of the slurry rather than mass transfer limitations.

Very similar yields were obtained for experiments with a holding time of 20 min at 90 °C and a thermal treatment of 1 min holding time followed by 19 min without applying any input power to the sample (holding and flash treatments in Fig. 6). Pectin extraction can be understood as a two-step process of hydrolysis and solubilisation [38]. First, pectin is hydrolysed from the cell wall. Second, it gets solubilised and is diffused through the biomass pores to the surface and into the bulk solution. Diffusion-controlled processes are considered slower than hydrolysis reactions, and therefore the second event is assumed as the rate-limiting step. The common approach to perform a thermal extraction treatment is to hold a target temperature for a certain amount of time [22,39,40], after which the solution is cooled-down and the extracted pectin precipitated. With this flash experiment, it was proven that there is no need to hold the target temperature for the whole duration of the experiment. The only requirement is to allow enough time to elapse so that diffusion can take place, but not necessarily at the target temperature. This is shown in Fig. 6, where higher extract yields are obtained for 1 min at the target temperature followed by 19 min gradual cool-down, than for a 20 min hold-time. This finding would not only allow the implementation of a continuous process and decreased energy consumption, but also reduce pectin exposure to high temperatures and thus potentially reduce degradation reactions. The pectin compositional analysis will be discussed in Section 3.3.

Fig. 7 displays the extract yields obtained for both routes assayed (SP and RE) during continuous-flow experiments using the two types of de-starched potato pulp (enzyme-treated and sieved), and comparing them with the corresponding batch ones (conducted at 400 W and 25% feed concentration). The 20 min of constant heating at the target temperature in full recirculation (RE) operational mode corresponded to the holding batch experiment, whereas the heating to target temperature plus 20 min of cooling down time (SP) operational mode corresponded to the flash batch experiment.

Continuous-flow extraction experiments had, on average, smaller yields than the batch ones, achieving as little as half the extract yield in the case of the sieved SP mode. This result was unexpected, as the lower feed concentration and higher heating rate could have been expected to increase the yield. The slightly lower processing temperature (85 compared with 90 °C) may have contributed to the reduced yield. However, it is noted that optimisation was not within the scope of this feasibility study, and so it is likely that yields could be improved with further experimentation. Comparing the continuous operational modes, the yields were comparable for both sieved samples, at around 1%. This

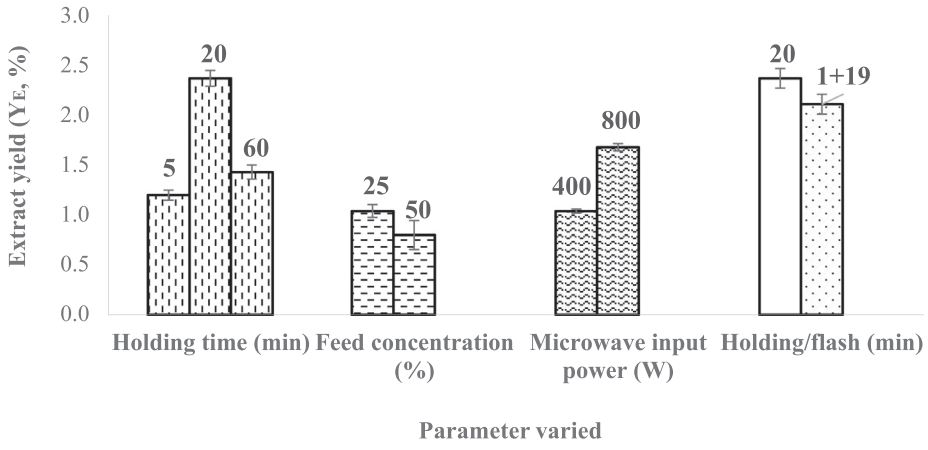

Fig. 6. Extract yields (Y_E, %) obtained during batch exploratory experiments according to the parameter varied. Refer to Table 2 for further details on the experimental parameters. Error bars show differences between replicates.

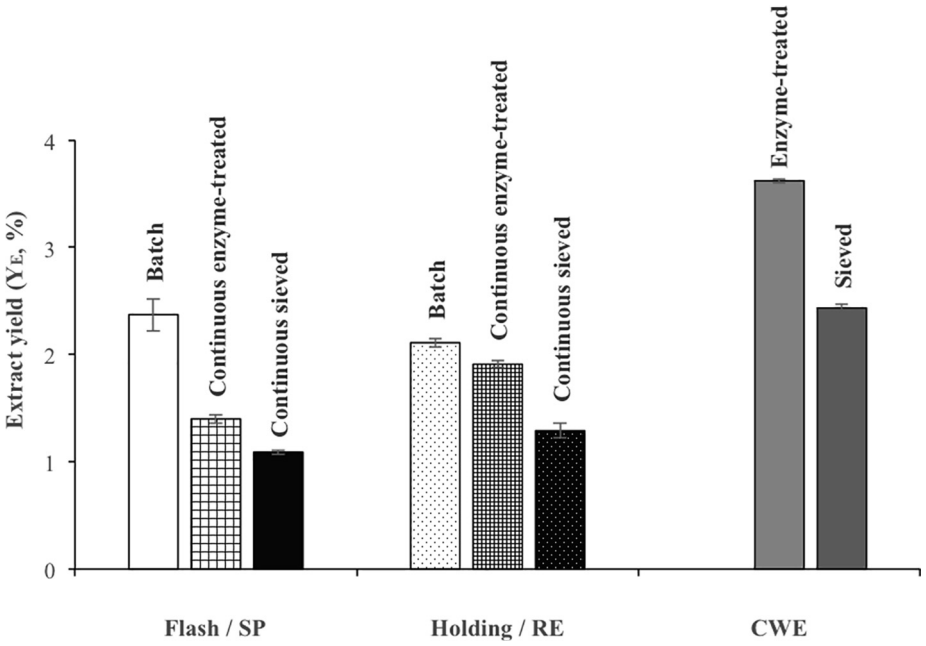

Fig. 7. Continuous-flow MAE experiments extract yield (Y_E, %), compared to the corresponding batch ones. The continuous-flow experiments were performed with both the enzyme-treated pulp and the sieved pulp in both operation modes (SP and RE). Batch experiments were performed with enzyme-treated pulp. Error bars show differences between replicates.

favours the hypothesis that a single pass continuous process can be achieved through a very fast hydrolysis step during heating followed by solubilisation and diffusion during the cool-down phase, and that this is equivalent, or potentially better than, a continuous thermal treatment. The enzyme-treated extracts were higher in yield than the sieved ones. This was expected since some starch precipitated in the extract during the IPA treatment (see Section 3.3.2), thus causing an overestimation of the quantity of pectin.

It should be noted that these results refer to the total extract yield, this is the amount of extract obtained in relation with the initial raw material used. In potato tubers, the average pectic substances content is of around 0.05–0.35% DM [45], and thus low extract yield values were expected.

3.3. Extracts composition

3.3.1. GalA content and yield

Fig. 8 depicts the GalA yields obtained in all the continuous-flow experiments. For both de-starching procedures, the SP route yields were more than double both the RE and the conventional extraction (CWE), achieving a GalA extraction yield of over 40%, compared with 15–20% for all of the other runs. The starch removal method doesn't appear to have affected the GalA yield, being slightly higher for the sieved samples in the SP runs and slightly lower in RE mode, but very close to within the error bars in both cases.

The fact that the SP operation mode allowed the extraction of significantly more pectin than longer treatments is related with the hypothesis proposed in Section 3.2. A thermal treatment of only 0.81 s but followed by cooling-down for 20 min under stirring was enough to allow both pectin hydrolysis and diffusion; and at the same time prevented pectin degradation, thus greatly enhancing the total amount recovered by the alcohol precipitation. This result highlights the potential impact of continuous processing on the wider pectin extraction industry.

3.3.2. Neutral sugars content

Pectin-derived oligosaccharides (POS) have been identified as emerging prebiotics, and the prebiotic activity is related with the hairy regions [6]. Hairy regions contain neutral sugar chains [46]. Therefore, the neutral sugar content was analysed by ionic chromatography (IC) as an indicator of hairy pectin regions in the extracts. For the enzyme-treated extracts, the Glu content was too high to obtain chromatograms in which every monosaccharide would have an isolated peak after method optimisation (see supporting document S.4). Glu content is an indicator of the extract starch content. Starch molecules are composed of Glu monomers, arranged in either linear or branched structures. Starch is stored in plants as semi-crystalline granules, which have a density of around 1.5 g cm^{-3}. Due to the high density, some starch co-

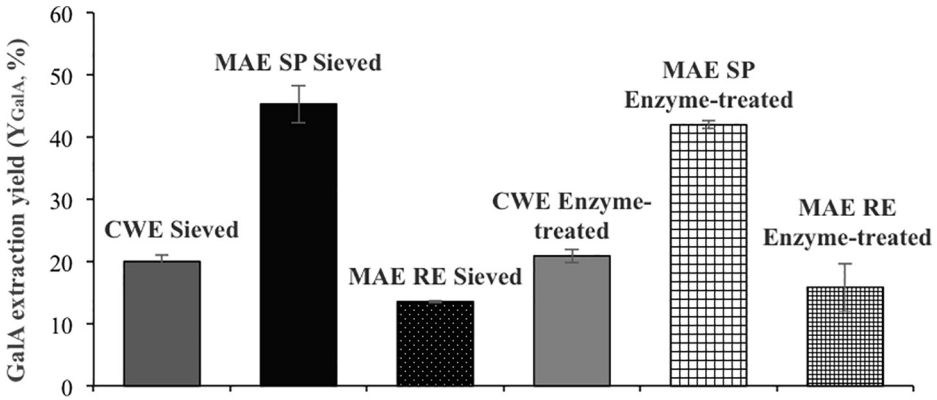

Fig. 8. GalA yield (Y_{GalA}, %) measured in the continuous-flow extracts compared to conventional extraction yields. Error bars show differences between replicates.

precipitates with pectin during downstream processing [47], and that is why Glu is present in the extracts. The poor de-starching performance of the enzymatic treatment is caused by the presence of resistant starch in potatoes [28,48]. As a result of this, the Glu peak was wide enough to hinder/affect the neutral sugars peaks at all the concentrations assayed, and thus could not be integrated. However, for the sieved extracts, the degree of de-starching was sufficient to obtain good peak resolution, and therefore, the sugar analysis results (Fig. 9) correspond only to the sieved continuous-flow extracts, as well as the raw sieved pulp and the extract obtained with CWE.

IC was carried out not only on the extracts, but also on the permeate of the sieving process. This was performed to evidence that only starch and not hairy pectin regions were removed. As expected, only a Glu peak and no other neutral sugars peaks were observed (see Section S.4 of the supporting document). This confirms that the sieving process allows starch to be isolated and collected as a co-product of the process, rather than being reduced to glucose in the enzyme process. It also offers the advantage of being more economic compared with the enzymatic procedure, and it would be straightforward to integrate it as a continuous upstream process to pectin extraction.

Fig. 9 shows that hairy pectin regions were extracted, as demonstrated by the presence of neutral sugars. Main neutral sugars were Rha, galactose (Gal) and arabinose (Ara), which agrees with other works on potato pectin extraction [1,27,46]. Extracts containing Gal, Ara and also Rha have been showed to increase beneficial bacteria populations in the colon, confirming their potential as prebiotic ingredients [6].

Glu yield was significantly higher in CWE extracts (66.11%) compared with MAE extracts (41.10 and 36.07% for RE and SP respectively). Conversely, Rha, Ara, Gal and xylose (Xyl) yields were higher in MAE extracts than in CWE extracts in all cases. Additionally, the SP operational mode gave higher yields than the RE mode. This means that neutral sugars were preferentially extracted with MAE rather than with CWE, and that the SP route allowed higher yields of neutral sugars and lower yield of Glu than both MAE RE and CWE. Therefore, MAE, especially under short heating times, improved the quality of the extracts compared with CWE. This, along with the GalA yields presented in Fig. 8, confirm the hypothesis that the shorter heating time afforded by continuous process maximises yield by minimising hairy pectin thermal degradation.

3.4. Scaling-up routes

This study has demonstrated the feasibility of continuous-flow microwave-assisted extraction of hairy pectin from potato pulp. This represents an important step forward in the efforts towards scaling-up microwave assisted extraction processes. The systematic approach that was followed for the development of the lab-scale continuous-flow process, as illustrated in Fig. 2, would be a good base for any attempt at scaling up the process.

The simplest strategy for scaling up the process would be to move to a lower frequency. This would not require special cavity design, and a simple single-mode cavity similar to the one used in the current study could be used. The use of a lower frequency would bring two important merits for scaling-up. The first is the larger penetration depth due to the longer wavelength at lower frequencies. For example, a frequency of ~0.9 GHz would result in a penetration depth of about 10 cm based on Equation S.1 in the supporting document. The second advantage of using a lower frequency is the larger single-mode applicator that comes with it. The standard WR340 waveguide, which was used in the current study, has a height of 4.3 cm, whereas a standard WR975 which covers frequencies from 0.75 GHz to 1.15 GHz has a height of 12.4 cm. Applying the same residence time used in the current study (i.e. 0.81 m s^{-1}) for a flow in a 10 cm diameter tube through a standard WR975 waveguide would allow for processing 1.2 lit s^{-1} (4.3 ton hr^{-1}). The choke design, pressure drop and energy requirements can be determined using the same procedures followed in the current study.

4. Conclusions

This work experimentally demonstrates the feasibility of using a scalable continuous-flow microwave-assisted thermal treatment to extract hairy pectin regions from industrial potato pulp, using only water as the extraction medium. The system allowed good temperature control of ± 2.5 °C and achieved a stable target temperature in ≈1 min processing time at a feed flow rate of 250 mL min^{-1}. The best results of 40–45% Y_{GalA} were achieved when treating the potato pulp in a single pass with as residence time of less than 1 s in the microwave cavity, which more than doubles the yields obtained using recirculating mode or batch conventional extraction. Neutral sugars, predominantly Rha, Gal and Ara, were detected in the extracts, also at higher concentration in single pass operational mode. This proves the suitability of microwaves to be used as a fast heating method to extract pectin avoiding degradation.

The high starch content in potato pulp necessitated the inclusion of a starch removal step to prevent gel formation during processing and reduce the glucose content in the extract. This work demonstrated that starch removal via a sieving step (as opposed to the enzymatic treatment commonly reported in the literature) affords superior starch removal, and also the opportunity to isolate starch as a separate value stream in the process.

The work presented in this paper demonstrates, for the first time, a scalable process to extract “hairy” pectin from potato pulp. This will enable testing of the extracts for a wide range of bioactivities and a route to scale-up, information that is vital to attract investment in the commercialisation of this process. The design methodology presented here will contribute to the development of microwave-based continuous systems, which have huge potential application in the sustainable valorisation of distributed biomass feedstocks.

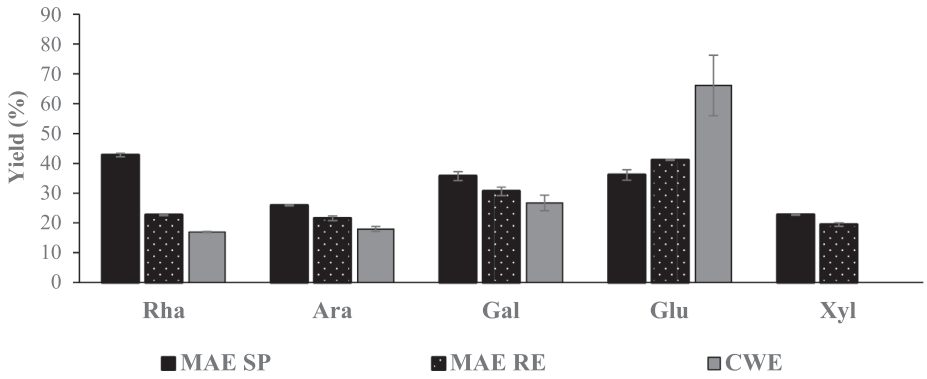

Fig. 9. Sugar yields (calculated as percent of sieved raw pulp contents) of the sieved CWE and sieved continuous-flow extracts. Error bars show differences between replicates. Rha = Rhamnose, Ara = Arabinose, Gal = Galactose, Glu = Glucose and Xyl = Xylose.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.cej.2020.125056.

References

[1] I. Byg, J. Diaz, L.H. Øgendal, J. Harholt, B. Jørgensen, C. Rolin, R. Svava, P. Ulvskov, Large-scale extraction of rhamnogalacturonan I from industrial potato waste, Food Chem. 131 (2012) 1207–1216.
[2] N. Khodaei, S. Karboune, V. Orsat, Microwave-assisted alkaline extraction of galactan-rich rhamnogalacturonan I from potato cell wall by-product, Food Chem. 190 (2016) 495–505.
[3] B.M. Yapo, Pectic substances: From simple pectic polysaccharides to complex pectins—A new hypothetical model, Carbohydr. Polym. 86 (2011) 373–385.
[4] W.G.T. Willats, L. McCartney, W. Mackie, J.P. Knox, Pectin: cell biology and prospects for functional analysis, Plant Mol. Biol. 47 (2001) 9–27.
[5] R. Ciriminna, A. Fidalgo, R. Delisi, L.M. Ilharco, M. Pagliaro, Pectin production and global market, AgroFOOD Ind. Hi-tech 27 (2016) 17–20.
[6] B. Gómez, B. Gullón, R. Yáñez, H. Schols, J.L. Alonso, Prebiotic potential of pectins and pectic oligosaccharides derived from lemon peel wastes and sugar beet pulp: a comparative evaluation, J. Funct. Foods 20 (2016) 108–121.
[7] C. Onumpai, S. Kolida, E. Bonnin, R.A. Rastall, Microbial utilization and selectivity of pectin fractions with various structures, Appl. Environ. Microbiol. 77 (2011) 5747–5754.
[8] B. Gullón, B. Gómez, M. Martínez-Sabajanes, R. Yáñez, J.C. Parajó, J.L. Alonso, Pectic oligosaccharides: Manufacture and functional properties, Trends Food Sci. Technol. 30 (2013) 153–161.
[9] M.C.R. Mano, I.A. Neri-Numa, J.B. da Silva, B.N. Paulino, M.G. Pessoa, G.M. Pastore, Oligosaccharide biotechnology: an approach of prebiotic revolution on the industry, Appl. Microbiol. Biotechnol. 102 (2018) 17–37.
[10] M. Marić, A.N. Grassino, Z. Zhu, F.J. Barba, M. Brnčić, S. Rimac Brnčić, An overview of the traditional and innovative approaches for pectin extraction from plant food wastes and by-products: ultrasound-, microwaves-, and enzyme-assisted extraction, Trends Food Sci. Technol. 76 (2018) 28–37.
[11] S.-Y. Chan, W.-S. Choo, Effect of extraction conditions on the yield and chemical properties of pectin from cocoa husks, Food Chem. 141 (2013) 3752–3758.
[12] I. Fraeye, A. De Roeck, T. Duvetter, I. Verlent, M. Hendrickx, A. Van Loey, Influence of pectin properties and processing conditions on thermal pectin degradation, Food Chem. 105 (2007) 555–563.
[13] A. De Roeck, T. Duvetter, I. Fraeye, I.V.d. Plancken, D.N. Sila, A.V. Loey, M. Hendrickx, Effect of high-pressure/high-temperature processing on chemical pectin conversions in relation to fruit and vegetable texture, Food Chem. 115 (2009) 207–213.
[14] S. Bhatia, H. Sharma, M. Alam, Pectin from agricultural by-products: structure, methods of extraction, physiological benefits and applications, Indian J. Agric. Biochem. 29 (2016) 1.
[15] P. Srivastava, R. Malviya, Sources of pectin, extraction and its applications in pharmaceutical industry – An overview, Indian J. Natural Products Resour. 2 (2011) 10–18.
[16] G. Mao, D. Wu, C. Wei, W. Tao, X. Ye, R.J. Linhardt, C. Orfila, S. Chen, Reconsidering conventional and innovative methods for pectin extraction from fruit and vegetable waste: Targeting rhamnogalacturonan I, Trends Food Sci. Technol. 94 (2019) 65–78.
[17] F. Chemat, N. Rombaut, A. Meullemiestre, M. Turk, S. Perino, A.-S. Fabiano-Tixier, M. Abert-Vian, Review of Green Food Processing techniques, preservation, transformation, and extraction, Innovative Food Sci. Emerging Technol. 41 (2017) 357–377.
[18] Y. Li, A.S. Fabiano-Tixier, M.A. Vian, F. Chemat, Solvent-free microwave extraction of bioactive compounds provides a tool for green analytical chemistry, TrAC, Trends Anal. Chem. 47 (2013) 1–11.
[19] A. Taqi, E. Farcot, J.P. Robinson, E.R. Binner, Understanding microwave heating in biomass-solvent systems, Chem. Eng. J. 393 (2020) 124741.
[20] M.L. Fishman, H.K. Chau, P.H. Cooke, M.P. Yadav, A.T. Hotchkiss, Physico-chemical characterization of alkaline soluble polysaccharides from sugar beet pulp, Food Hydrocolloids 23 (2009) 1554–1562.
[21] S. Yeoh, J. Shi, T.A.G. Langrish, Comparisons between different techniques for water-based extraction of pectin from orange peels, Desalination 218 (2008) 229–237.
[22] Y. Mao, R. Lei, J. Ryan, F. Arrutia Rodriguez, B. Rastall, A. Chatzifragkou, C. Winkworth-Smith, S.E. Harding, R. Ibbett, E. Binner, Understanding the influence of processing conditions on the extraction of rhamnogalacturonan-I “hairy” pectin from sugar beet pulp, Food Chem.: X 2 (2019) 100026.
[23] A.S. Matharu, J.A. Houghton, C. Lucas-Torres, A. Moreno, Acid-free microwave-assisted hydrothermal extraction of pectin and porous cellulose from mango peel waste – towards a zero waste mango biorefinery, Green Chem. 18 (2016) 5280–5287.
[24] V.L. Budarin, P.S. Shuttleworth, M. De bruyn, T.J. Farmer, M.J. Gronnow, L. Pfaltzgraff, D.J. Macquarrie, J.H. Clark, The potential of microwave technology for the recovery, synthesis and manufacturing of chemicals from bio-wastes, Catal. Today 239 (2015) 80–89.
[25] J.-S. Yang, T.-H. Mu, M.-M. Ma, Extraction, structure, and emulsifying properties of pectin from potato pulp, Food Chem. 244 (2018) 197–205.
[26] H. Pastukh, J. Grytsay, Y. Hrabovskyi, Research on the ways of the pectin extraction from the potatoes, Ukrainian J. Food Sci. (2013) 199–203.
[27] N. Khodaei, S. Karboune, Extraction and structural characterisation of rhamnogalacturonan I-type pectic polysaccharides from potato cell wall, Food Chem. 139 (2013) 617–623.
[28] Y.-D. Li, T.-C. Xu, J.-X. Xiao, A.-Z. Zong, B. Qiu, M. Jia, L.-N. Liu, W. Liu, Efficacy of potato resistant starch prepared by microwave–toughening treatment, Carbohydr. Polym. 192 (2018) 299–307.
[29] Y.-Y. Zhang, T.-H. Mu, M. Zhang, Optimisation of acid extraction of pectin from sweet potato residues by response surface methodology and its antiproliferation effect on cancer cells, Int. J. Food Sci. Technol. 48 (2013) 778–785.
[30] A.C. Metaxas, R.J. Meredith, Industrial Microwave Heating, Peter Peregrinus Ltd., London, 1983.
[31] C. Gabriel, S. Gabriel, E.H. Grant, E.H. Grant, B.S.J. Halstead, D. Michael, P. Mingos, Dielectric parameters relevant to microwave dielectric heating, Chem. Soc. Rev. 27 (1998) 213–224.
[32] R. Meredith, Engineers' Handbook of Industrial Microwave Heating, The Institution of Engineering and Technology, London, 1998.
[33] HSE, The Control of Electromagnetic Fields at Work Regulations, 2016.
[34] N. Wang, J.G. Brennan, The influence of moisture content and temperature on the specific heat of potato measured by differential scanning calorimetry, J. Food Eng. 19 (1993) 303–310.
[35] X. Guo, D. Han, H. Xi, L. Rao, X. Liao, X. Hu, J. Wu, Extraction of pectin from navel orange peel assisted by ultra-high pressure, microwave or traditional heating: a comparison, Carbohydr. Polym. 88 (2012) 441–448.
[36] T.M. Filisetti-Cozzi, N.C. Carpita, Measurement of uronic acids without interference from neutral sugars, Anal. Biochem. 197 (1991) 157–162.
[37] S. Christiaens, S. Van Buggenhout, K. Houben, Z. Jamsazzadeh Kermani, K.R. Moelants, E.D. Ngouemazong, A. Van Loey, M.E. Hendrickx, Process-structure-function relations of pectin in food, Crit. Rev. Food Sci. Nutr. 56 (2016) 1021–1042.
[38] L.R. Adetunji, A. Adekunle, V. Orsat, V. Raghavan, Advances in the pectin production process using novel extraction techniques: a review, Food Hydrocolloids 62 (2017) 239–250.
[39] H. Bagherian, F. Zokaee Ashtiani, A. Fouladitajar, M. Mohtashamy, Comparisons between conventional, microwave- and ultrasound-assisted methods for extraction of pectin from grapefruit, Chem. Eng. Process. Process Intensif. 50 (2011) 1237–1243.
[40] J.P. Maran, K. Swathi, P. Jeevitha, J. Jayalakshmi, G. Ashvini, Microwave-assisted extraction of pectic polysaccharide from waste mango peel, Carbohydr. Polym. 123 (2015) 67–71.
[41] C.S. Lee, E. Binner, C. Winkworth-Smith, R. John, R. Gomes, J. Robinson, Enhancing natural product extraction and mass transfer using selective microwave heating, Chem. Eng. Sci. 149 (2016) 97–103.
[42] J. Robinson, J. Meehan, A. Taqi, E. Binner, B. Tokay, Water desalination using a temperature gradient, Desalination 464 (2019) 1–7.
[43] Y. Liu, J. Shi, T.A.G. Langrish, Water-based extraction of pectin from flavedo and albedo of orange peels, Chem. Eng. J. 120 (2006) 203–209.
[44] N. Flórez, E. Conde, H. Domínguez, Microwave assisted water extraction of plant compounds, J. Chem. Technol. Biotechnol. 90 (2015) 590–607.
[45] G. Lisinska, W. Leszczynski, Potato Science and Technology, Springer, Netherlands, 1989.
[46] F. Xie, W. Zhang, X. Lan, S. Gong, J. Wu, Z. Wang, Effects of high hydrostatic pressure and high pressure homogenization processing on characteristics of potato peel waste pectin, Carbohydr. Polym. 196 (2018) 474–482.
[47] M. Sjöö, L. Nilsson, Starch in Food: Structure, Function and Applications, Elsevier Science, 2017.
[48] R. Hoover, Y. Zhou, In vitro and in vivo hydrolysis of legume starches by α-amylase and resistant starch formation in legumes—a review, Carbohydr. Polym. 54 (2003) 401–417.