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Microwave depolymerisation of PMMA: Power density, char management, and reactor design for high-purity monomer recovery

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ABSTRACT

Poly(methyl methacrylate) (PMMA) is a versatile polymer valued for its optical and mechanical properties, durability and biocompatibility. However, its production carries a high carbon footprint, primarily due to the energy-intensive synthesis of its monomer, methyl methacrylate (MMA), while recycling rates remain below 10 %, largely due to limitations in current conventionally heated technologies. Microwave depolymerisation offers a promising route to address these challenges by enabling greener, more energy-efficient recovery of the monomer. However, due to the unique microwave heating mechanisms, innovation in microwave reactor design is required to realise this at industrial scale. This study defines the key processing parameters required for successful scale-up of microwave depolymerisation of PMMA into high-purity MMA. A semi-continuous microwave reactor was developed to explore these parameters under controlled conditions. Electromagnetic simulations were employed to optimise reactor performance and provide insight into the observed process behaviours. For the first time, the roles of power density and char accumulation in determining product quality during microwave depolymerisation of PMMA are quantitatively established, highlighting the need for reactor designs that enable high power density and effective char removal. These findings define key design principles for microwave reactor scale-up and represent a critical step toward economically and environmentally viable PMMA recycling within circular plastic systems.

1. Introduction

Plastics have become an essential part of modern life, permeating nearly every sector of society. However, their widespread use has resulted in significant environmental concerns, driven by large-scale production and inadequate waste management. These environmental challenges include air pollution from open burning, threats to marine life due to ocean pollution, and the ubiquitous spread of microplastics [1].

Poly(methyl methacrylate) (PMMA), commonly known as acrylic, acrylic glass, Perspex and Plexiglas, and its *co*-polymers are known for their excellent optical and mechanical properties, durability and biocompatibility. These characteristics make them attractive choices for applications in the automotive, construction, healthcare, and electronics industries [2]. Global PMMA production is estimated at 3.9 million tonnes per year, with a projected compound annual growth rate (CAGR) of 3.4 % [3]. However, only 10 % of PMMA waste is currently recycled [3]. Beyond the wider environmental consequences of poor plastic waste management, PMMA is also among the polymers with a notably high carbon footprint associated with its production. The global warming potential (GWP) of primary PMMA products in Europe is estimated to range from 3.9 to 5.1 kg CO2eq/kg PMMA [4]. The majority of this GWP is attributed to monomer production, which accounts for 3.7 kg CO2eq/kg MMA [5]. Other studies have reported even higher GWP values for PMMA production [6]. Adopting a circular approach by maximising recycling rates is viewed as a key strategy for addressing these environmental challenges while enabling the continued use of such versatile materials. To achieve this, the development of advanced recycling

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technologies is urgently required.

Mechanical recycling, which involves processing plastic waste into new products by melting and reforming the plastic, is the most widely used method for recycling plastics, accounting for 99 % of recycled plastics in Europe [7]. However, the repeated heat and pressure cycles involved in the process have been shown to degrade the polymer's structure. This limits the recycled plastic to low-value applications and reduces the ultimate number of reprocessing cycles it can undergo. For PMMA, mechanical recycling has a particularly negative impact on its optical properties, one of its key features. Moreover, mechanical recycling is unsuitable for thermosetting plastics, as their highly cross-linked polymer structures prevent them from being re-melted and reshaped [8]. Some PMMA products, such as dental resins [9], are highly cross-linked, which restricts their recyclability through mechanical methods. Collectively, these limitations constrain the potential of mechanical recycling to substantially increase recycling rates.

Meanwhile, chemical recycling has emerged as a promising alternative with the potential to significantly enhance recycling rates. It involves breaking down the polymer into its oligomers, monomers, and other basic chemicals, which can then be used to produce the original polymer or other high-value products. The process usually involves heating the plastic waste, with or without the addition of chemicals/catalysts.

PMMA can be broken down to its constituent monomer, methyl methacrylate (MMA), by heating to temperatures up to 450 °C without the addition of other chemicals or catalysts. This is known as thermal depolymerisation or pyrolysis and can recover >95 % MMA compared with ≤ 85 % monomer recovery for polystyrene and $\leq$ 45 % for polypropylene and polyethylene at lab scale [10]. The resulting monomer can be used to produce virgin quality polymer products. Additionally, PMMA made from recycled MMA has been shown to have a lower carbon footprint than that produced from virgin MMA [11,12].

At industrial scale, various thermal processing methods have been explored for the depolymerisation of PMMA, including molten metal baths, dry distillation, rotating drums, fluidised beds, and extruders [2,13]. Among these, the molten lead bath process is the most widely used industrial technology; however, it raises significant environmental concerns due to lead contamination of the solid residues produced during processing [14]. A major challenge with all these heat transfer-based methods is the low thermal conductivity of plastics, which along with the presence of voids between particles or films, leads to slow heating rates. This often necessitates larger equipment. Furthermore, longer residence times during PMMA depolymerisation has been linked to lower monomer yields and diminished product purity [15,16]. While increasing the temperature of the heating medium can typically speed up heating, higher temperatures are also linked to secondary decomposition and side reactions, which lower both liquid yield and purity [16–18].

Microwave heating has emerged as an attractive technique for delivering the energy required to break the polymer chains during thermal decomposition [19–22], including some progress toward industrial implementation [23–25]. Unlike conventional heat transfer methods, microwave heating works through the direct interaction of electromagnetic fields with the material at molecular and sub-molecular levels, bypassing the limitations of heat transfer. As a result, microwave heating is considered an energy transfer technique rather than a heat transfer method. This allows for faster heating, more compact equipment, and the potential to enhance product quality and energy efficiency. Microwave heating is also regarded as a promising technology for decarbonising process industries, as it uses electricity, which can be supplied from renewable and green energy sources.

Polymers are generally considered poor microwave absorbers at room temperature due to their complex molecular structure and lack of mobile electrons. However, polymers containing polar segments are believed to exhibit increased microwave susceptibility, especially at elevated temperatures [26]. In such cases, heating occurs through the local movement of these polar segments in response to the alternating electromagnetic field. This heating mechanism, known as dipolar dispersion or displacement, differs from the dipolar loss mechanism commonly associated with polar liquids in that the latter involves rotation of the whole molecule. PMMA is believed to interact with microwaves through its polar ester group [26,27]. In a recent study, Adam et al. demonstrated that PMMA's dielectric loss factor – a measure of its microwave absorption ability – increased significantly at temperatures above its glass transition temperature (T_g), reaching a peak that is over an order of magnitude higher than its room temperature value [27]. This increase was attributed to the greater mobility of the polymer chains above its T_g, which allows the polar segments more freedom to interact with the electromagnetic field. The findings affirmed that polymers with polar segments, such as PMMA, can be effectively heated with microwaves without the need for additional microwave absorbent materials (susceptors). This marked a significant departure from the common practice in studies exploring microwave heating for plastic pyrolysis, as highlighted in several review papers [19–22], where the addition of susceptors such as carbon and silicon carbide (SiC) are often considered essential. The inclusion of susceptors in the pyrolysis process typically adds capital and operational costs due to the extra equipment required to recover and recycle the susceptor back into the reactor.

Despite considerable research on PMMA depolymerisation and the growing interest in microwave technology for plastic recycling, there is a lack of studies specifically addressing the microwave depolymerisation of PMMA. One of the few works in this area is a patent by AECI Limited [28] and more recently a patent by Microwave Chemical CO., LTD [29]. Although both, the patent includes some experimental examples of PMMA depolymerisation without a microwave susceptor, neither provide details on how process parameters affect product yield, quality, or overall process efficiency. Such analysis is essential to identify the optimal reactor designs and conditions required for successful scale-up.

The overarching aim of this study is to establish how key processing parameters govern the heating behaviour, product yield, and liquid purity during microwave depolymerisation of PMMA. To achieve this, the following objectives were set: (i) to set up an experimental system capable of investigating microwave depolymerisation of PMMA under controlled laboratory conditions; (ii) to develop an electromagnetic simulation model to predict the electric field configuration within the system, thereby assisting in optimisation of the setup and providing insight into the associated heating behaviours; (iii) to investigate the relationship between the dielectric properties of the reaction bed and the resulting heating profile; and (iv) to quantify the influence of key processing parameters on product yield and quality.

2. Materials and methods

2.1. Materials

Clear cast PMMA (Perspex®), supplied by Mitsubishi Chemical UK Ltd., was used in this study. According to information provided by the supplier, this grade contains no pigments or co-monomers. The PMMA samples were initially granulated in a rotary mill and then sieved to achieve the desired particle size. The PMMA grade used has a glass transition temperature (T_g) of 102 °C, determined by Differential Scanning Calorimetry (DSC), as shown in Fig. A1 in the Supporting Information document. Thermogravimetric analysis (TGA) showed that this PMMA grade starts to decompose at around 180 °C, with the main decomposition phase occurring between 300 and 450 °C, as illustrated in Fig. A2 in the Supporting Information document. The dielectric constant and loss factor of the sample, measured using the cavity perturbation technique, are also presented in Fig. A3 in the Supporting Information document. The dielectric constant (ε') is a measure of the material's ability to store electrical energy, while the loss factor (ε'') represents the material's ability to convert electromagnetic energy into heat.

2.2. Microwave depolymerisation setup

A lab-scale, semi-continuous microwave heating system (Fig. 1) was developed to carry out the PMMA depolymerisation tests outlined in this paper. The system comprised a 2.45 GHz microwave generator (GU020, IMS Ltd., Buckinghamshire, UK) with a maximum power output of 2.0 kW. The microwave power was delivered to the applicator/cavity via standard WR340 and WR430 waveguides. A sliding short-circuit and a three-stub motorised Homer auto-tuner (STHT 2.45 GHz, S-TEAM, Bratislava, Slovakia) were employed for impedance matching to improve the efficiency of microwave power delivery. The Homer tuner measured the forward and reflected power which allowed the determination of the microwave power absorbed by the heated material by difference.

An IR pyrometer (Optris CT 3MH, Berlin, Germany) was used to measure the temperature during microwave heating. Operating at a wavelength of 2.3 μm, the pyrometer measures the surface temperature of the polymer sample inside the quartz tube, rather than the external surface of the quartz tube itself. This model has an effective temperature measurement range of 100 °C to 600 °C and, from its mounted position, measures a spot diameter of approximately 15 mm on the reactor wall. The recorded temperature represents the average value across the measured spot size.

The samples were heated in a 34 mm (i.d.) quartz tube/reactor placed inside a single-mode cavity. The quartz tube was secured by a clamp and positioned such that the sample is centred within the cavity as displayed in Fig. 1. A condensation system consisting of a water-cooled coiled condenser followed by a wet ice trap and then a liquid nitrogen trap was used to recover condensable decomposition products. Samples of the non-condensable gases were collected at the outlet of the liquid nitrogen trap. The cavity's top choke and the glass assembly between the choke and the coiled condenser were trace-heated and thermally insulated to minimise early condensation of the depolymerisation vapours and prevent liquid reflux into the reactor.

The system was operated in semi-continuous mode, with PMMA particles continuously fed from an overhead feeder (LAMBDA DOSER, LAMBDA CZ, s.r.o., Czech Republic) as displayed in Fig. 1. The feeder comprised a 1-l glass vessel with a side arm, a distributor, and a programmable controller. The side arm was used to load the sample into the feeding vessel before the experiments and later to introduce nitrogen into the reactor during the process. The feeding rate was regulated by adjusting the motor speed via the controller. An optical camera (Microsoft LifeCam Studio Webcam) was employed to monitor and record the depolymerisation process through a side port in the reactor cavity. Approximately 18 g of PMMA particles, with a size range of 0.5–3 mm, were loaded into the quartz tube, and 40–100 g were placed into the feeder vessel before the experiment began. Fresh polymer feeding from the overhead vessel commenced after decomposition in the reactor had started. The depolymerisation experiment can be divided into three distinct stages. Stage 1 covers the period from the onset of microwave heating of the initial polymer charge until just before the introduction of fresh polymer. Stage 2 corresponds to the period of continuous feeding. Stage 3 begins once fresh polymer feeding stops and continues until microwave heating is terminated.

After the experiments, the liquid recovered in the condensers was collected and weighed. Any remaining PMMA in the feeding vessel was also collected and weighed to determine the amount of PMMA fed into the reactor during the experiment. The solid residue in the reactor was quantified by subtracting the weight of the empty tube, measured before the experiments, from the combined weight of the tube and its contents after the experiment. The liquid and solid residue yields were calculated accordingly. The char content was determined by heating the reactor, with the solid residue still inside, in an electrically heated tube furnace at 600 °C for 1 h under a nitrogen atmosphere and measuring the remaining solid. This step was carried out to ensure complete depolymerisation of any polymeric residue left in the reactor.

2.3. Characterisation

Gas samples were collected in gas bags (Supel™-Inert Multi-Layer Foil, Sigma-Aldrich Co. LLC) and analysed on the same day on a Clarus 580 gas chromatograph (PerkinElmer, Inc., US) equipped with a flame ionisation detector (FID) for hydrocarbon gases and a thermal conductivity detector (TCD) for non-hydrocarbon gases. The method used for GC analysis was based on that reported in Uguna et al. [30]. Individual hydrocarbon gas fractions were identified and quantified in

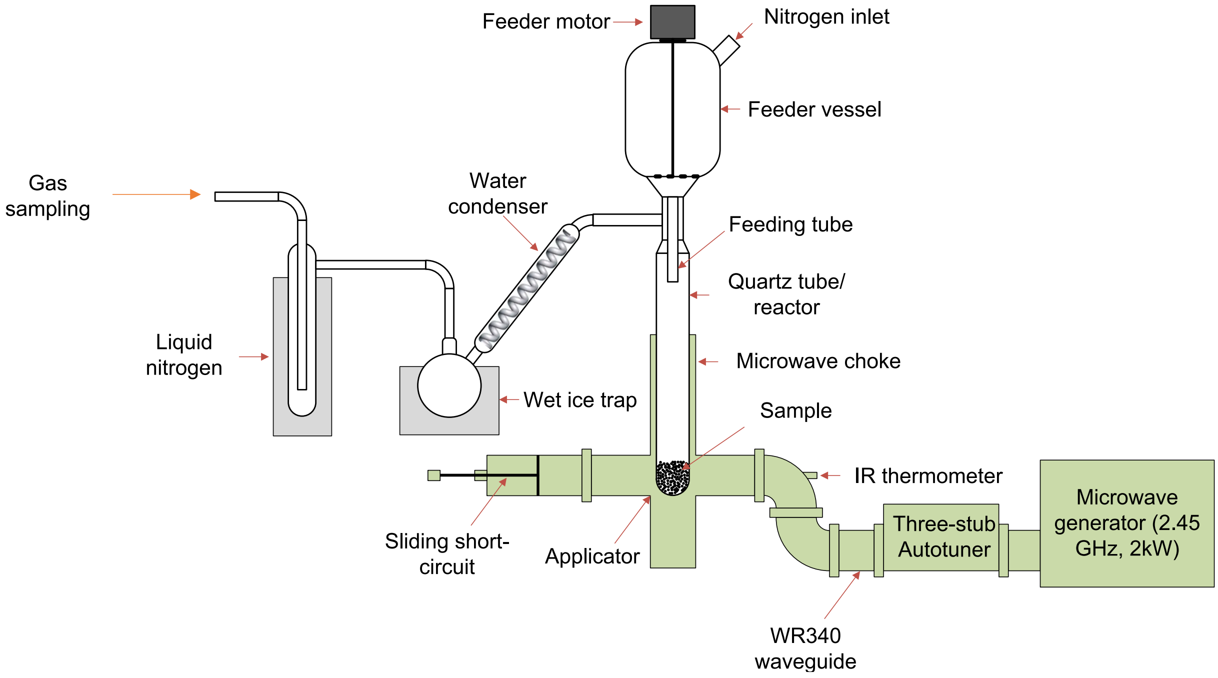

Fig. 1. Schematic of the semi-continuous apparatus developed for the microwave depolymerisation experiments.

relation to C1-C5 gases (injected separately) as an external gas standard. The non-hydrocarbon gases were also quantified relative to a mixture of H_2, H_2S, CO, and CO_2 as external gas standards. The calculations for the quantification of hydrocarbon (C1–C5) and non-hydrocarbon (H2, H2S, CO, and CO2) gases were based on ASTM D7833–14 [31].

Liquid samples were collected in glass vials, and approximately 100 ppm of Topanol A stabiliser was added to inhibit the activity of any potential free radicals. The composition of the liquid was then analysed using a gas chromatography apparatus equipped with mass spectroscopy and flame ionisation detectors (GC–MS/GC-FID), with the analysis method developed by Mitsubishi Chemical Group Ltd. (See Appendix B in the Supporting Information document).

3. Results and discussion

3.1. Electromagnetic modelling

Electromagnetic simulations using COMSOL Multiphysics® V6.1 were conducted to model the electric field configuration in the cavity and waveguides, as well as the resulting power dissipation density within the workload. The aim was to help determine the optimal position for the sliding short-circuit and the initial positions of the tuning stubs. It was previously shown that using electromagnetic simulations to set the sliding short position results in better heating homogeneity compared to techniques such as cold-matching with a network analyser [32]. Further details on setup of the simulation model, including geometry, material properties, and boundary conditions, can be found in Appendix C in the Supporting Information document.

Simulations were initially run to set the sliding short at a position that placed the PMMA sample in the cavity within a region of maximum electric field intensity (i.e. an electric field 'hotspot'). This was performed while leaving all three tuning stubs outside the waveguide. Once the sliding short position was set, simulations were then run to optimise the position of the tuning stubs with the aim of achieving the highest microwave power delivery efficiency (absorbed power as a fraction of input power). This was achieved by iteratively adjusting stubs #1 & #2 or stubs #2 & #3 between 0 and 20 mm (see Fig. 2). The latter is the maximum distance into the waveguide that each stub can travel. The highest power delivery efficiency of 42 % was achieved at stub positions of 0, 10, and 20 mm for stubs #1, #2, and #3, respectively.

Fig. 2 shows the electric field configuration within the cavity and waveguide, as well as the resulting power dissipation density (i.e. absorbed power per unit volume) within the reactor at the optimised positions of the sliding short and tuning stubs. These settings were used as the initial configuration for the microwave depolymerisation experiments (results presented in Section 3.2 onwards). During the depolymerisation experiments, automatic tuning consistently maintained stub #1 at 0 mm and stub #3 at 20 mm. The position of stub #2 varied between 9 mm and 13 mm in response to changes in the dielectric properties of the bed. Additionally, during the early stages of steady feeding and heating, and before significant quantities of char formed and accumulated in the reactor, the power delivery efficiency ranged from 45 % to 50 % (see Section 3.2). These values are very close to the 42 % predicted by the electromagnetic simulations. These findings demonstrate the robustness of the simulations in both predicting the electric field configuration and in in guiding the selection of the initial sliding-short and tuning-stub positions used throughout the depolymerisation experiments.

3.2. Heating profile

Fig. 3 shows typical absorbed power and temperature profiles during the microwave depolymerisation of the studied PMMA grade, carried out using the setup described in Section 2.2 at a feeding rate of 8.8 g/min and an input power of 800 W. The profile can be divided into three stages consistent with those described in Section 2.2. Stage 1 covers the period from the start of microwave heating of the pre-loaded polymer in the reactor until just before feeding of fresh polymer begins. Stage 2 corresponds to the period of constant feeding, and it is the closest region to steady-state processing. Stage 3 starts when the feeding of fresh polymer stops and continues until microwave heating is halted.

The relatively low absorbed power at the beginning of Stage 1 was attributed to the polymer's low dielectric loss at lower temperatures, particularly below its glass transition temperature as discussed in detail by Adam et al. [27]. After approximately one minute of heating, the absorbed power began to increase rapidly due to the rise in the loss factor above the glass transition temperature (see Fig. A1 in the Supporting Information document).

The sample surface temperature recorded by the IR sensor during Stage 1 and the beginning of Stage 2 was likely significantly lower than the temperature at the centre of the reactor. This is evidenced by the presence of an undecomposed polymer layer on the reactor walls at the start of Stage 2, despite most of the polymer at the centre having already decomposed, as shown in Fig. 4. This significant temperature gradient is believed to result from a combination of electromagnetic and heat transfer factors. The electromagnetic factor is related to the higher power dissipation density at the middle of the reactor compared to near the walls, as illustrated in Fig. 2, leading to a greater heating rate in the middle. The heat transfer effect is the heat loss to the cold surroundings of the uninsulated reactor, causing a substantial temperature gradient across the reactor's width. This suggests that even if the power dissipation density was uniform across the reactor bed, a temperature gradient would still exist, with heat flowing from the reactor to its colder surroundings. It is important to note that the temperature gradient observed in this laboratory-scale system is partly a result of limited thermal insulation. In a properly insulated systems, such gradients

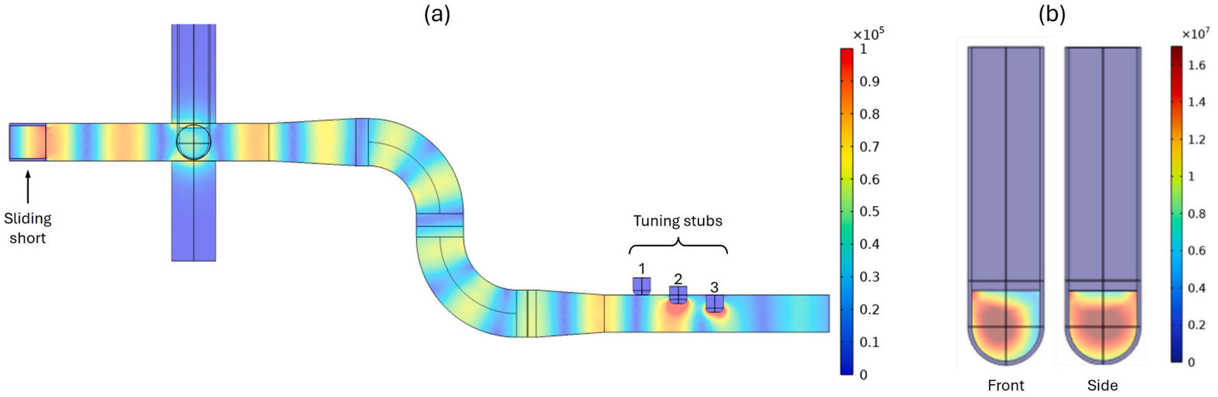

Fig. 2. Electromagnetic simulation results showing: (a) the electric field distribution, in V/m, within the cavity and waveguides; and (b) the power dissipation density, in W/m^3, in the reactor and workload.

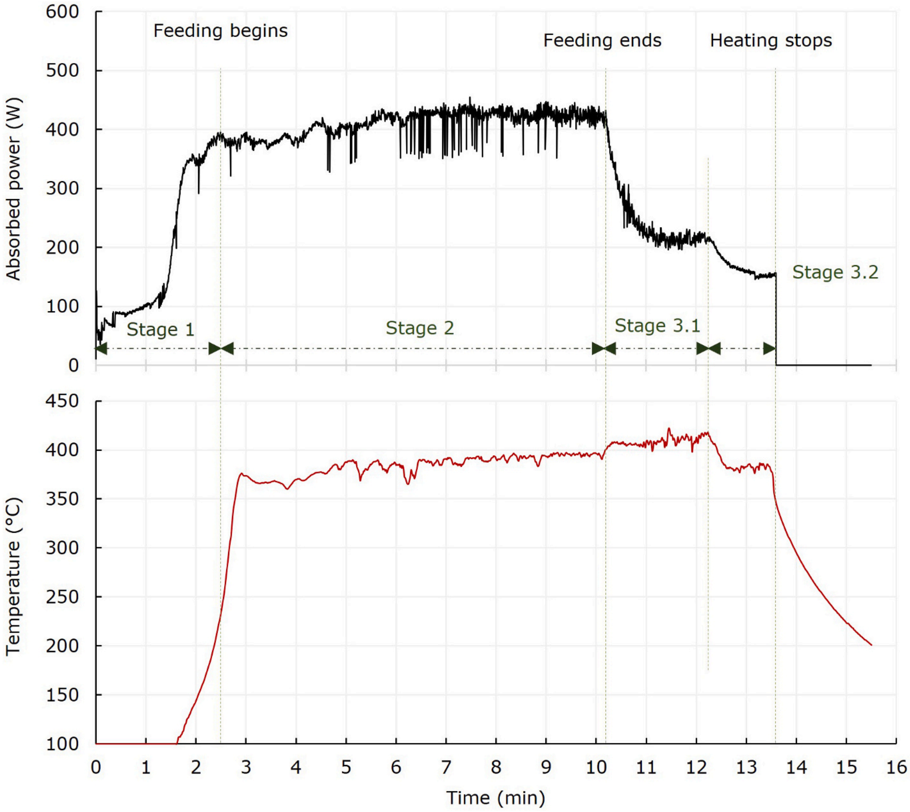

Fig. 3. Experimentally measured power and temperature profiles during microwave depolymerisation of clear cast PMMA at a feeding rate of 8.8 g/min and an input power of 800 W.

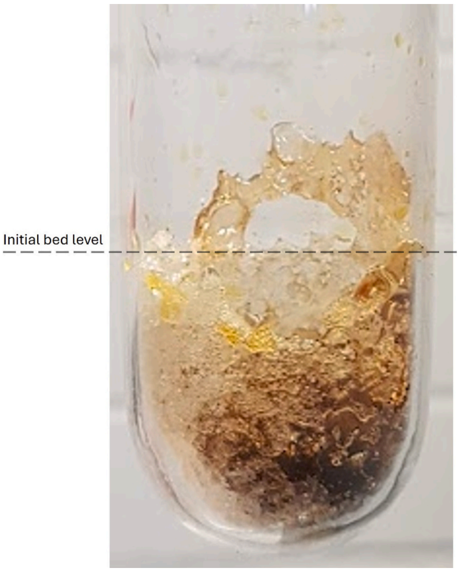

Fig. 4. Solid residue (21 %) left inside the reactor after the experiment was stopped at the end of Stage 1. The liquid yield from this test was 77 %. Most of the polymer at the reactor's centre had decomposed, leaving a layer of mostly un-depolymerised material on and near the reactor wall.

would be significantly reduced.

A gradual increase in absorbed power and temperature was observed during Stage 2, likely caused by the formation and accumulation of char in the reactor. Char is a good microwave susceptor, and its presence probably caused the increase in absorbed power, which subsequently led to a rise in the temperature of the reaction bed. The reduction in absorbed power during Stage 3 is due to the decreasing amount of polymer in the reactor after the feeding stops. The slightly faster temperature rise at the beginning of Stage 3 compared with Stage 2 can be attributed to the higher char-to-polymer ratio in the reactor once fresh polymer is no longer being added.

The apparent temperature drop toward the end of Stage 3 is thought to be due to the bed level falling below the spot diameter measured by the IR pyrometer. It is possible that the bed temperature continued to rise during Stage 3, even though the pyrometer showed a decrease.

Typical product yields and compositions, based on the conditions used to produce the profiles shown in Fig. 3, are summarised in Table D1 in the Supporting Information document. Under these conditions, the process consistently achieved a liquid yield of approximately 96 %, a solid yield of 1.7 %, and a gas yield of less than 3 %. The liquid product consisted predominantly of MMA (~95.0 wt%), with minor impurities. The total amount of char deposited in the reactor was approximately 0.47 % of the total processed PMMA. The gas phase was primarily composed of carbon dioxide and carbon monoxide, alongside small amounts of light hydrocarbons.

These results serve as a reference for assessing the effects of processing time, feeding rate, and power density on product yield and quality, as discussed in Section 3.3.

3.3. *Effect of processing conditions on product yield and quality*

3.3.1. *Processing time and char accumulation*

Experiments were carried out over varying durations, but with a constant feeding rate, to examine the impact of gradual char accumulation in the reactor and the associated increase in bed temperature over time as illustrated by Fig. 3 on the product quality. Fig. 5 shows the concentrations of MMA and methyl isobutyrate (MIB) in the liquid product as a function of feeding duration, which is directly proportional to the amount of polymer fed during the steady feeding period. MIB was

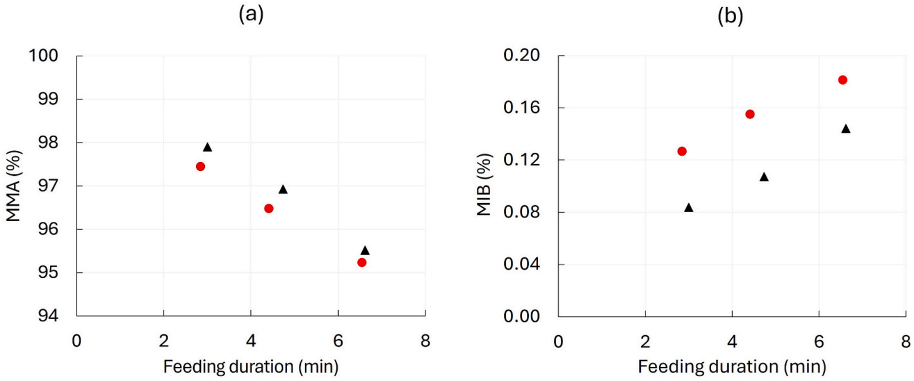

Fig. 5. Concentrations of MMA and MIB in the liquid product after microwave depolymerisation of clear cast PMMA at a feeding rate of 8.8 g/min and an input power of 800 W. The black triangles represent experiments where the microwave power was stopped simultaneously with the end of dosing (end of Stage 2 as shown in Fig. 3). The red circles represent experiments where the microwave power remained on after dosing stopped, continuing into Stage 3.

specifically highlighted among the impurities in the liquid product, as it is a side product whose formation is favoured at high temperatures [9,16]. Its generation has also been previously linked to char formation, where hydrogen released during the charring process can hydrogenate MMA to form MIB [15]. As the char dehydrogenation is itself accelerated at high temperatures [33], this further reinforces the strong temperature dependence of MIB formation. The presence of MIB in the liquid product at significant concentrations increases the cost and complexity of downstream purification by distillation, as its boiling point (92 °C) is close to that of MMA (100 °C) [34].

Fig. 5 shows that as the processing duration increased, the concentration of MMA in the liquid product decreased, while the concentration of MIB increased. The significantly higher concentration of MIB when heating continued after feeding had stopped (indicated by the red circles in Fig. 5) compared to when both heating and feeding were halted simultaneously (indicated by the black triangles) can be attributed to the elevated temperatures during Stage 3 (see Fig. 3), which favour MIB formation.

The effects of temperature and char accumulation on the gas quality were also investigated. Gas samples were collected at various intervals during the microwave depolymerisation experiments. Fig. 6 presents the concentrations of carbon dioxide (CO_2), carbon monoxide (CO), and methane (CH_4) over time from two separate experiments. The formation of these gases during PMMA depolymerisation has previously been linked to the decomposition of radicals formed by random side group scission and/or MMA decomposition [2]. The results in Fig. 6 indicate that the CO concentration increased over time. A sharp rise in both CO and CH_4 concentrations, along with a decrease in CO_2 concentration, was observed after feeding stopped (Stage 3, as shown in Fig. 3). This shift in gas composition is likely due to changes in reaction pathways resulting from char formation and the subsequent increase in bed temperature. One possible explanation for the rise in CO concentration and the corresponding decrease in CO_2 levels is the Boudouard reaction, a reversible process in which carbon reacts with carbon dioxide to form carbon monoxide ($C + CO_2 \rightleftharpoons 2CO$). The forward reaction becomes thermodynamically favourable at higher temperatures [35]. As the experiments progressed, particularly during Stage 3, the reactor conditions – characterised by increased char content and elevated temperatures – likely favoured greater CO production and a corresponding reduction in CO_2 level.

3.3.2. *Feeding rate*

The impact of feeding rate on liquid quality further illustrated how char accumulation and subsequent temperature increases affect product quality. As shown in Fig. 7 (a) and (b), increasing the feeding rate resulted in a rise in MMA concentration, while the concentration of MIB decreased. This trend can be attributed to the corresponding decrease in reaction bed temperature with increased feeding rates, as presented in Fig. 7 (c). The variation in bed temperature is likely linked to both the amount of polymer present in the reaction bed and the char-to-polymer ratio. At lower feeding rates, the available microwave power is higher than the minimum required for complete polymer decomposition. The excess power input results in an increase in bed temperature and a reduction in the amount of polymer present in the reactor, due to faster decomposition. As a result, the char-to-polymer ratio increases. Since char is a more effective microwave absorber than PMMA, the higher char content further enhances microwave absorption, leading to an additional rise in bed temperature. In contrast, at a higher feeding rate, the greater influx of fresh polymer maintains a lower char-to-polymer ratio and a higher overall polymer content in the bed. This reduces

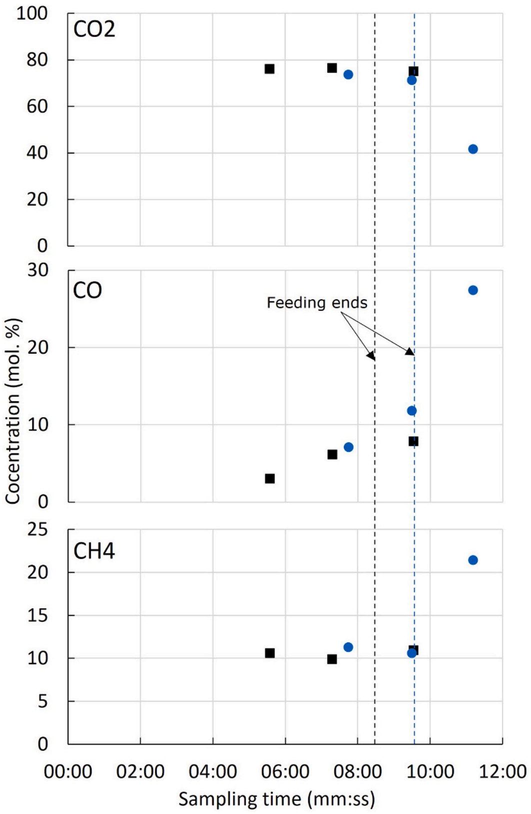

Fig. 6. Concentration of CO, CO_2 and CH_4 in gas product from clear cast as a function of sampling time. The results are from two separate experiments: one represented by black squares and the other by blue circles.

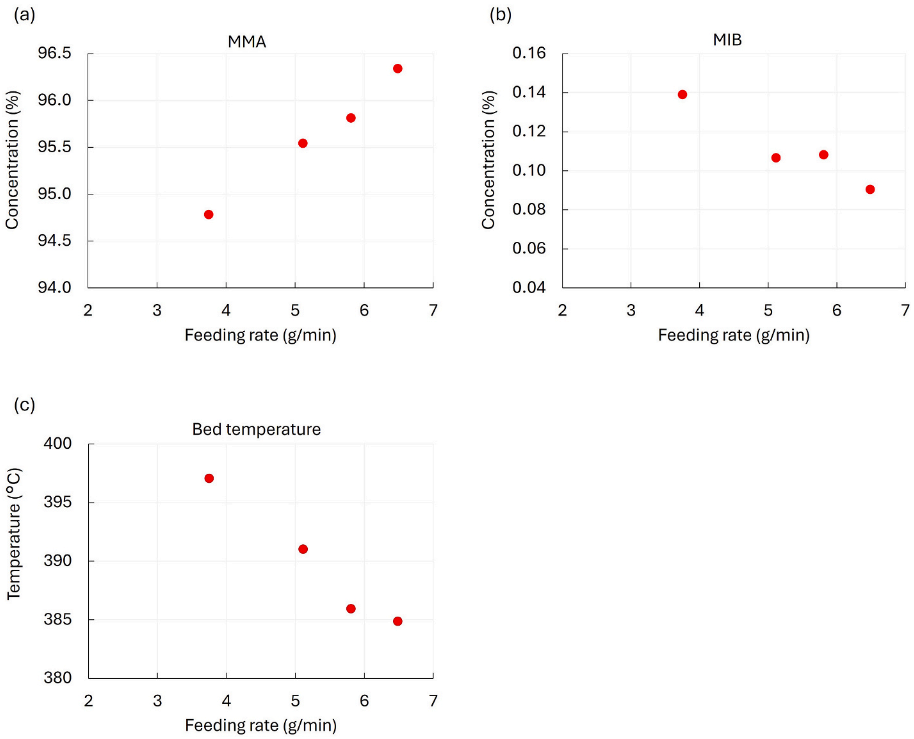

Fig. 7. Effect of feeding rate on the liquid quality (a and b) and bed temperature (c) during microwave depolymerisation of clear cast PMMA. All experiments were conducted at 600 W input power with a total processed quantity of PMMA of 78 ± 3 g. Liquid and solid yields were within 95.5 ± 0.5 % and 2.8 ± 0.9 %, respectively in all experiments.

the extent of microwave absorption by char, resulting in lower bed temperatures.

3.3.3. *Power density*

The effect of power density on product yield and liquid quality was investigated within a range of 0.61×10^7 to 3.4×10^7 W/m^3, corresponding to input power values of 200 W and 1600 W, respectively. The power density was calculated as the average absorbed power during the steady-state feeding period (Stage 2, as described in Section 3.2) divided by the bed volume, which was approximately 27 cm^3. The apparent reactant residence time[2] ranged from 4 min at the lowest power density to 62 s at the highest. At the maximum power density, the heating rate of the incoming fresh polymer was estimated to be 380 °C/min, which is notably higher than what conventional heat transfer mechanisms can typically deliver. Heating rates on the order of 10s °C/min (<100 °C/min) have been reported during plastic pyrolysis in conventionally-heated pilot-scale screw reactor systems [36].

A number of measures were taken to isolate the effect of power density on product yield and quality. The total processed polymer was kept constant at approximately 78 g to eliminate the impact of char accumulation associated with differences in the total amount of material processed, as discussed in Section 3.3.1. Similarly, the ratio of power input to feeding rate was maintained at 93 ± 1 W per g/min across all experiments to enable variation of the power density while keeping the available energy input per gram of polymer effectively constant.

Fig. 8 (a) shows that low power densities resulted in low liquid yields, primarily due to incomplete depolymerisation near the reactor walls. This was likely caused by a combination of low bed temperatures and high temperature gradients, with the underlying factor being the relatively high heat loss, as discussed in Section 3.2. The liquid yield remained consistently high (≥96 %) at power densities above 1.5×10^7 W/m^3. As shown in Fig. 8 (b), the bed temperature increased with rising power density but began to plateau above 1.5×10^7 W/m^3 which corresponds to the point where both liquid and solid yields also started to level off.

Unlike depolymerisation in conventional heat transfer-based reactors, where the bed temperature is controlled by the heating medium or surface, microwave depolymerisation is constrained by the maximum temperature the polymer can reach before fully decomposing and exiting the reaction bed. Therefore, unless significant quantities of char accumulate in the reactor vessel to act as an additional heat source, microwave depolymerisation of PMMA can be considered potentially self-limiting in temperature.

The results shown in Fig. 8 (b) indicate that the maximum achievable temperature within the studied power density range and char accumulation levels is approximately 410 °C. Excessively high temperatures, often necessary to enhance heat flux and heating rates in conventional heat transfer systems, have previously been shown to reduce both liquid yield and purity due to secondary side reactions [16–18]. In contrast, microwave depolymerisation demonstrated improved liquid purity as power density, which drives the heating rate, increased. As shown in Fig. 9 higher power densities resulted in an increase in MMA concentration in the liquid product, along with a reduction in impurities such as MIB. This enhancement in product quality at higher power densities is likely due to the shorter residence time, which is believed to limit side reactions. Previous studies have also shown that longer residence times promote the formation of side products including MIB [9,15]. Additionally, the aforementioned self-regulating capability of microwave heating during PMMA depolymerisation allowed for a higher heating rate while avoiding the excessive temperatures that also trigger side reactions.

The specific energy, defined as the average absorbed power during the continuous feeding period (Stage 2, as described in Section 3.2)

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[2] The apparent residence time was calculated by dividing the bed volume by the volumetric feeding rate. The latter was determined from the weight-based feeding rate and the initial bed packing density, which was approximately 0.67 g/cm^3.

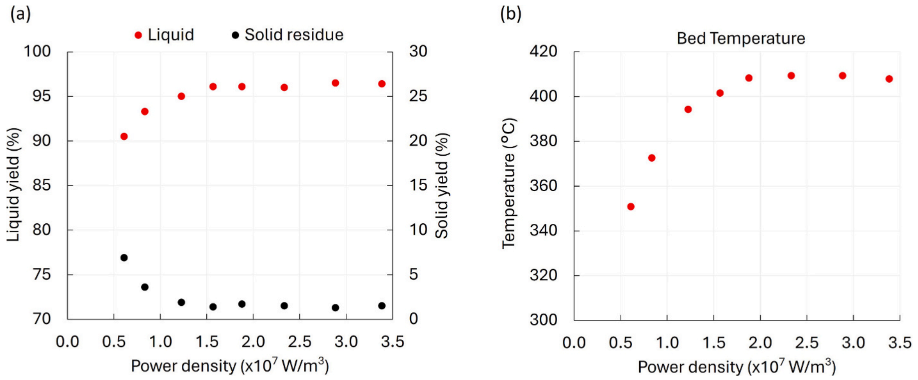

Fig. 8. Effect of power density on (a) product yield, and (b) bed temperature. The total processed feed quantity was kept constant at about 78 g, with the ratio of power input to feeding rate maintained at 93 ± 1 W per g/min across all experiments.

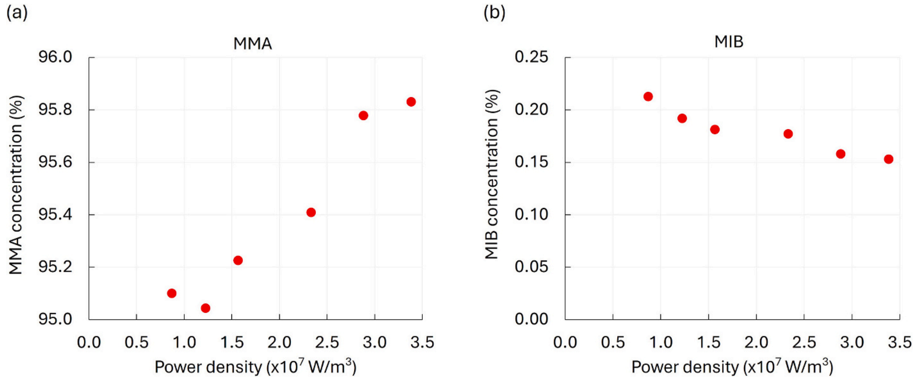

Fig. 9. Effect of power density on liquid purity.

divided by the feeding rate, ranged from 2.8 kJ/g to 5 kJ/g across the studied power density range of 0.61 × 10^7 to 3.4 × 10^7 W/m^3. It is important to note that, since the section of the reactor within the cavity is not thermally insulated, significant heat loss through the reactor wall is expected. A heat loss analysis, detailed in Appendix E, indicates that the specific energy requirement would range between 2.4 kJ/g and 3.4 kJ/g. This remains a conservative estimate, as it accounts only for heat losses within the microwave-heated section of the reactor. Comparatively, energy consumption in the range of 0.92 to 1.50 kWh/kg (3.31 to 5.40 kJ/g) has been reported in the literature for PMMA pyrolysis in conventionally heated fluidised bed systems [37]. Lower energy consumption levels (1.9–3.1 kJ/g) have also been reported for a twin-screw depolymerisation process [38]. In practice, the energy requirement of a properly insulated microwave reactor would be expected to approach the theoretical values (1.7–1.8 MJ/kg [13]), since microwave heating selectively delivers energy directly to the polymer, thereby avoiding excessive bulk heating of the reactor.

3.4. Implications of the findings on potential scale-up routes

The findings presented in this paper provide a robust foundation for microwave reactor design and carry significant implications for scale-up, enabling a step change in PMMA recycling through the microwave technology. The results have identified the key design features required for successful scale-up. High power density (i.e. a faster heating rate) and shorter residence times are required to achieve high monomer yield and purity. Additionally, char accumulation during the process, along with the resulting increase in bed temperature, negatively impacts product purity and can lead to the formation of undesirable side products, such as MIB.

Enhancing liquid product purity by increasing MMA concentration and reducing impurities can significantly lower downstream processing costs. Effective removal of these impurities is essential to achieve virgin-quality PMMA from recycled MMA [39]. MIB is particularly challenging to separate due to its similar boiling point and solubility to MMA, often requiring complex and energy-intensive purification steps [34]. That said, Boutros et al. [39] have reported a two-stage distillation process in which the azeotrope formed between MIB and water is exploited in the first stage to facilitate its separation from MMA. Nevertheless, suppressing impurities formation during microwave depolymerisation remains the most effective approach to simplify purification, reduces energy use, and improves the economic viability of recycled PMMA.

Effective char management is therefore essential for sustaining both high yield and liquid quality. Reactor configurations that enable high power density and incorporate mechanisms for either minimising char formation or actively removing it during processing are, therefore, likely to offer a significant performance advantage. Continuous systems, such as conveyor-based reactors, inherently facilitate the rapid transport of material and may support continuous char removal, making them attractive candidates for scale-up. However, this does not preclude the viability of other reactor systems. Ultimately, the findings from this study provide a framework for evaluating different scale-up pathways based on their ability to support key operating conditions: high power density, short residence time, and robust char management.

4. Conclusions

This study investigated the microwave depolymerisation of PMMA in a semi-continuous setup, specifically examining the impact of process conditions on product yield and quality. The results indicated that char accumulation within the reactor led to a gradual increase in absorbed power and bed temperature, which adversely affected the purity of the liquid product. Additionally, both char accumulation and bed temperature influenced the quality of the gas product.

Higher microwave power densities, which resulted in heating rates of up to 380 °C/min and shorter polymer residence times, significantly enhancing liquid purity while maintaining a consistently high liquid yield. The inherent self-regulating capability of microwave heating during PMMA depolymerisation facilitated these higher heating rates while avoiding excessive temperatures that can trigger side reactions, a combination that is challenging to achieve in conventional heat transfer-based systems.

The findings reported in this paper suggest that future scale-up efforts should prioritise reactor designs capable of supporting high power density while actively removing char from the system. These conditions are more likely to be realised in continuous conveyor-type processes rather than in fed-batch systems.

Moreover, electromagnetic simulations were employed in this study to optimise the lab-scale system for heating homogeneity and power delivery efficiency. The predicted fraction of absorbed power and the optimal positioning of tuning stubs closely aligned with experimental values, demonstrating the reliability of these simulation tools. Such tools are invaluable during the design phase for scaling up, as well as for optimisation and troubleshooting during operation.

These findings lay the groundwork for a new generation of microwave depolymerisation reactors, underpinned by detailed understanding of microwave–material interactions and the key roles of power density, residence time, and char management. By highlighting the importance of reactor designs that maximise monomer recovery and suppress impurity formation, this work is essential to unlock a substantial increase in PMMA recycling rates. The use of renewable electricity to power microwave systems offers a practical route to reducing the global warming potential of PMMA production. More broadly, the combined benefits demonstrated in this study represent a meaningful step toward the development of low-carbon, circular plastic systems.

CRediT authorship contribution statement

Mohamed Adam: Writing – review & editing, Writing – original draft, Visualization, Resources, Project administration, Methodology, Investigation, Formal analysis, Conceptualization. **Nicklas Hjalmarsson:** Writing – review & editing, Resources, Methodology, Conceptualization. **Chai Siah Lee:** Writing – review & editing, Investigation. **Nidia Diaz Perez:** Investigation. **Mark Fields:** Methodology, Investigation. **Adam J. Clarke:** Supervision, Project administration, Conceptualization. **John Runnacles:** Writing – review & editing, Project administration, Funding acquisition, Conceptualization. **Derek J. Irvine:** Conceptualization. **John Robinson:** Supervision, Project administration, Funding acquisition, Conceptualization. **Eleanor Binner:** Writing – review & editing, Supervision, Project administration, Funding acquisition, Conceptualization.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: (Mohamed Adam, Nicklas Hjalmarsson, Adam J. Clarke, John Runnacles, John Robinson, and Eleanor Binner has patent #A method of microwave depolymerisation of (co)polymers of methyl methacrylate (GB2501009.1) pending to Mitsubishi Chemical UK Limited. If there are other authors, they 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.susmat.2025.e01807.

Data availability

Data will be made available on request.

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