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The process for making low density LAS surfactant detergent agglomerates using microwave heating

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Abstract

The internal structure of detergent surfactant agglomerates is modified using electromagnetic heating to produce a lower bulk density final product. The agglomerates are heated in a 1.8 KW microwave waveguide heating applicator operating at license-free ISM frequency band of 2450 MHz. The density of the agglomerates reduces significantly when exposed to high power electromagnetic fields. The effect of number of process variables such as input power initial moisture content over bulk density of detergent particles is studied. Three different input power levels (500 W, 1 KW & 1.6 KW) are applied to the agglomerates and it has been observed that higher input powers raise the temperature of the agglomerates very quickly, hence reducing the required residence time of the sample. Agglomerates exposed to the highest input power (1.6 KW) had the lowest bulk density. The temperature profile and residence time of the agglomerates during heating was continuously recorded. Experimental results obtained in the lab-based process will be used to design a full scale continuous mode applicator.

Keywords: Microwave heating, bulk density, porosity, microwave puffing, detergent agglomerates

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1. Introduction

Microwave drying of the materials has gained considerable attention in food, chemical and pharmaceutical industries and academia [1-6]. It has many benefits including short heating times, instantaneous on/off control, selective nature of heating etc. The rapid heating rates achievable with dielectric heating can result in higher yields in production, better quality and improved reproducibility which make it perfect candidate for some industrial drying applications. Besides drying applications, electromagnetic energy can be used to modify the internal structure (porosity) of the material being heated. Microwave "puffing" is the process of getting a lower density material by generating higher internal pressures within a particle by means of rapid heating [7]. The high internal pressure is generated due to formation of water vapour inside the material and this can result in significant deformation of the material. Microwave energy is well suited for reducing the bulk density of the materials due to its instantaneous rapid heating directly from inside the material[8]. This generation of pressure uniformly throughout the particle can alter the internal structure in a different manner compared to other, more conventional drying/heating techniques such as spray-drying. The generation of internal pressure inside the material helps to create a lower density material. It can also help the solubility of the material by creating internal pores that are better connected to the external environment. In granular detergent materials this can allow for the easier ingress of water and improved solubility.

This paper presents a study of change in the bulk density of LAS (Linear Alkylbenzene Sulphonate) surfactant detergent agglomerates using electromagnetic energy. Various process variables such as input power, residence time and initial moisture content of the agglomerate samples are discussed in detail to optimize the bulk density reduction.

2. Microwave Equipment

A lab-scale 1.8 KW continuous wave microwave waveguide heating experimental setup has been designed as shown in Fig 1. The microwave power generated by the SM845 (MKS instruments) microwave magnetron head is supplied to a waveguide applicator using WR-340 standard waveguides. A 6 kW isolator is placed between the magnetron

and the waveguide applicator to protect the magnetron from any strong reflections. A GA6004A universal waveguide applicator from Gerling Engineering is used as the waveguide applicator (see Fig 2). This is a standard WR-340 waveguide section with a 5cm diameter removable adapter ports on the top and bottom walls. This set-up allows for the use of this applicator either in batch mode or continuous flow applications. There are a number of 3mm diameter holes in one side wall of the applicator. These are used to insert fibre-optic temperature probes in the sample. The holes also allow for the videoing of the sample heating and the removal of water vapours during heating. The sample is contained within an Acrylic holder when placed inside the waveguide applicator.

A four channel fibre optic thermometer (FOB-100 from Omega Engineering) is used to continuously monitor the temperature of the samples during exposure to microwave radiation. The fibre-optic probe is placed within the sample via one of the 3mm holes in the side wall of the applicator as shown in Fig 2(b). A digital video recording borescope from Maplin is used to record the visual changes in the volume of the sample.

Fig 1 Waveguide heating experimental setup

Fig 2 Waveguide applicator (a) Top view with Acrylic sample holder & HLAS sample (b) Side view- 3mm perforations for fibre optic probes

3. Materials

The starting material used in this study was a detergent agglomerate containing 33% LAS surfactant, 63.5% inorganics (sodium carbonate, sodium sulphate, zeolite) and ~ 3.5% water and miscellaneous.

4. Experiment procedure

Separate samples of the above agglomerate were conditioned to different moisture levels by exposure to a range of ambient humidities. Following conditioning, the dielectric properties of the different agglomerate samples were measured and the samples then exposed to electromagnetic fields. Heating time and temperature profile of the samples were recorded over time. The bulk density of each sample was measured before and after microwave heating. The density values reported here are for a bulk density measurement based on filling a sample of known volume in a consistent manner and measuring the weight of powder used. The bulk density was used because it is the most helpful in an industrial and consumer context. The bulk density is a result of the packing arrangement, the particle size distribution and the envelope density and how the materials are handled. Bulk density is a crude

combination of factors but is what is important in terms of how a consumer handles and experiences detergent products. Since the sample had been placed inside a cylindrical sample holder, it could only expand in a vertical direction. The following section discusses the results in detail.

5. Results

I. Dielectric loss measurement

Besides other factors, microwave heating strongly depends upon the dielectric properties of the material of interest[9]. At microwave frequencies, the dielectric heating is caused by molecular dipole rotation within the material when placed in electromagnetic fields. The polarisation of a permanently polarized dipolar material arises from the finite displacement of charges or rotation of dipoles within the material when influenced by an external electromagnetic field. This colliding of dipoles creates thermal agitation and heating takes place[10]. Dielectric properties of the material define its capability to absorb electromagnetic energy and convert it to heat. The electromagnetic power absorbed per unit volume in a dielectric can be expressed as [11]

$$p_v = \frac{1}{2}\{\sigma(\omega) + \omega\varepsilon_0\varepsilon''(\omega)\}|E|^2\ Wm^{-3} \qquad (1)$$

Where p_v is the total power absorbed per unit volume, ε'' is loss factor of the material and $|E|$ is the magnitude of electric field. The materials with higher loss factors absorb more electromagnetic energy and heat up more quickly.

The loss factors and dielectric constants of the surfactant agglomerates were measured at room temperature using an open circuit microstrip stub partly loaded with the powder material [12]. The complex dielectric properties were determined by measuring the scattering parameters of a two port microstrip bandstop resonator and then replicating these results by simulation using the electromagnetic simulation tool Ansoft HFSS (High Frequency Structure Simulator). Dielectric loading of the powder changes the resonant frequency and absorption of the circuit. Relative dielectric constants and loss factors were calculated from the relative shifts in frequency and dielectric absorption [12]. The dielectric properties of the agglomerate samples varied with the moisture level. Therefore, agglomerate samples with three different initial moisture contents were measured. Table 1 summarises the dielectric properties data of three samples conditioned at three different ambient equilibrium Relative Humidities (eRHs). Since, only the free water in the sample agglomerates contributes the dielectric loss factor of the material, therefore,

eRH method of moisture measurement is used in this study. eRH is the direct measure of the free water in the sample.

Table 1 : Dielectric properties of LAS samples

II. Frequency/ penetration depth

The amplitude of the electromagnetic wave decreases as it travels down in the dielectric material due to the absorption of power by the material. The penetration depth is defined as the depth at which electromagnetic wave strength decreases to 1/e times of the strength at the surface of the material. It can be calculated as [13]

$$d_p = \frac{0.225\lambda}{\sqrt{\varepsilon_r\sqrt{1+\tan^2\delta}-1}} \qquad (2)$$

Where λ is the wavelength and $\tan\delta = \frac{\varepsilon''}{\varepsilon'}$ represents loss tangent of the material. The rate of decay of the power dissipation is a function of both dielectric properties of the material and frequency of the incident wave. It is an important parameter to define the total depth of the powder bed.

III. Variable Input Power

Incident input power has a major effect on the heating from the dielectric workload. Increasing the input power increases the power density (electric field per unit area) and therefore reduces the required residence time [13]. The 64% eRH (~8 % water) agglomerate sample was exposed to three different input power levels to study the effect of variable input power on their bulk density. Fig 3 shows the comparison of the LAS agglomerate samples heated with 3 different input powers. Increasing input power decreases the residence time of the agglomerate sample to reach a certain temperature. For example, it took only 13s to raise the agglomerate sample temperature to 100 ^{0}C with 1.6 KW as compared to 45s at an input power level of 500 W. We can also expect that the bulk density of an agglomerate sample exposed to higher input power will be reduced further. This is due to the fact that higher input powers will raise the internal temperature of the agglomerate sample more quickly. This can generate steam more rapidly, which can then become trapped internally as it is harder for it to escape. However, at lower power levels it

takes longer to raise the temperature of the agglomerate sample. Any generation of steam is more gradual thereby increasing the chances of steam to escape without puffing the sample. Therefore, samples can just dry without much puffing. The different shapes of the curves are most likely due to the rate of breakdown of hydrate structures in the samples being tested. The more gradual absorption of energy by the sample at 500 W means that here breakdown in hydrate structures happens more gradually and hence increase in ability to absorb microwave energy is slower. There is a positive feedback mechanism here which causes the curve behaviour at 500W. At the higher powers tested this mechanism is very fast and the properties of the agglomerate do not change in the same way during the heating, hence the more linear response. Table 2 presents the bulk density data of the all three samples puffed at variable input powers. It was observed that the bulk density of the sample heated with 1600 W input power was reduced the most as compared to the lower input powers. Although the 500W sample was exposed to microwave radiation for a longer period of time, it had less change in the bulk density of the powder due to a slower heating rate.

Fig 03 Temperature vs time graph for 3 different input power levels

Table-2 Bulk density & residence time for samples exposed to different input powers

It should be noted that only a fraction of incident power is being absorbed by the sample due to smaller sample size and rest of the microwave energy is consumed in a dummy dielectric water load.

IV. Initial moisture content

Water can be in a range of different states within surfactant agglomerates. It can be “free” and strongly responsive to microwave radiation or chemically bound (hydrated) to other molecules and less responsive. The nature of the water within agglomerates has a major impact on the physical, chemical and dielectric properties of the agglomerates. The free water contributes most to the dielectric properties of the material[14]. The dielectric properties of the agglomerates can change significantly over time during microwave heating. This can be due to drying and a reduction in the free water level or an increase in the free water level due to a temperature induced break down of hydrates within the agglomerate. Although most bound/hydrated water will become “free” during microwaving (due to temperature-dependent hydrate decomposition) there should be a minimum amount of free water present at the start of the experiment to absorb EM energy and accelerate the experiment. Initial free moisture content is the key parameter for optimal bulk density reduction. If too much moisture is present in the sample: it

takes longer to form vapours and needs more effort to be removed later. Also the agglomerates may become too soft and deformable to handle. If too little water is present, the material may dry completely before it has any chance to puff [7]. LAS agglomerate samples conditioned to 18.4 % (~1.8 % water) and 65.3 % eRH (~8 % water) were prepared. Both of the samples were heated with 1 KW input microwave power to raise their temperature to 150°C. Fig 4 shows a comparison of the temperature profile for both samples during heating. It is evident that the sample with the higher moisture content heated up much more quickly than the other one due to the higher energy absorption by free water molecules having a higher loss factor. Water vapour started to be generated as soon as temperature of the sample rose to water boiling point. The water vapour collecting at the interior of the top surface of the waveguide was removed using a vacuum cleaner.

Fig 04 Temperature vs time graph for 2 different initial moisture content samples heated with 1KW input power

If the sample is completely dry then it will not puff at all due to absence of microwave energy absorbing water molecules. The dry LAS sample has a very low loss factor. Therefore, it behaves as a transparent material to microwave energy. A LAS agglomerate sample with 3% (wt) moisture was heated in an open container in hot air oven for 30 minutes at 65 ^{0}C to make it completely dry. The sample was then placed inside the microwave applicator to find out its interaction with microwave energy. It is observed that the sample temperature doesn't increase at all as shown in Fig 5. This concludes that the loss factor of the dry LAS agglomerate sample is too low to absorb any electromagnetic energy.

Fig 5 Temperature vs time graph for pre-heated (dry) sample at 500 W

V. Residence time

The temperature profile of the microwave heated sample vs residence time could be divided into three phases;

(i) Initially, the temperature increased exponentially up to 100°C. The reason being more and more bound water became "free" due to hydrate decomposition as the temperature increased. (ii) Then, beyond 100°C the temperature slope increased at a lower rate due to water evaporation. (iii) Finally, if we kept applying microwave energy to the sample then either (a) its' temperature started decreasing due to the fact that sample stopped absorbing electromagnetic energy because it had already become completely dry. Therefore, sample started to cool down (b) if

the sample was exposed to higher input powers then it occasionally became too hot after a very short period of time and ended up decomposing due to localised overheating.

Therefore, low input power can be used for longer period of time for drying applications. The objective of reducing the bulk density of materials can be achieved by exposing the samples to higher input powers for shorter period of times. This quick higher input energy helps to form water vapours inside the material which produce pressure inside the material required for puffing. Fig 6 shows the temperature versus time graph of four LAS agglomerate samples (64% eRH) exposed to two different power input levels (470 W & 1410 W) for different residence times. The bulk density data measured before and after puffing of the samples is presented in Table 3. It is observed that the higher input powers and longer residence times both help to reduce the bulk density of the samples more. Understanding the time dependent behaviour of the heating process is crucial to this work. Conditions which require a very extended period of microwave heating will not be industrially interesting. This is simply due to the size of the equipment necessary to ensure a longer residence time. The hypothesis for the reduction in density is that the particles need to be heated to 100 C to ensure the generation of steam internally but it was uncertain how long the particles needed to be maintained at elevated temperature to cause puffing. The different residence times were chosen to give an indication of whether the change in density happened once the particles reached 100 C or whether they needed to be maintained at these elevated temperatures for extended periods of time. The data implies that there is both a rapid, puffing effect and a more gradual drying effect.

Fig 6 Temperature vs time graph for different residence time

Table-3 Bulk density & residence time for sample 1, 4, 5 & 6

6. Micro_CT Analysis

Fig. 7 shows the reconstructed Micro_CT images of untreated and microwave puffed LAS sample with 1.6 kW power. The increased porosity and volume expansion could be clearly observed from the images. Refer to fig 7, the images shows significant increment in the number of closed and open pores of microwaved powder. Direct observation of imags comparison is supported by 3D anlaysis data, as the puffed sample had increased percentage differences in open; 2.75% and closed pores; 1.373% as compared to unpuffed sample. Table 4 presents the 3D

analysis of sample volume and porosity using the CTAn Micro-CT Software. The data shows an increament of 4.04% in the total porosity of the sample after it have been microwaved with 1.6 kW power. The detailed 2D and 3D analysis of the all of the discussed samples would be presented in a separate publication.

Fig 7 Micro_CT images of (a) untreated and (b) Microwave puffed LAS sample

Table-4 3D volume and porosity of untreated and microwave puffed LAS sample

7. Conclusion

The bulk density of the LAS detergent powder agglomerates is reduced using high power microwave heating to produce puffed (fluffy) product. It has been observed that by applying microwave heating, the porosity of the materials can be increased significantly without affecting its chemical properties. The key factors affecting microwave puffing such as input power, residence time and moisture level are discussed in detail. The high microwave power applied for short period of time introduces microwave puffing of the material while as lower power levels tend to simply dry the particles. For example, 1600W applied to sample 1 for only 29 sec reduces its bulk density from 959 g/l to 823 g/l while as the same sample treated with 500 W input power for the period of 116 sec brings bulk density of the sample from 959 g/l to 902 g/l. The data generated through this lab based microwave puffing experiments will be used to design a full scale industrial microwave heating unit for a continuous mode powder processing.

8. Acknowledgement

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9. References

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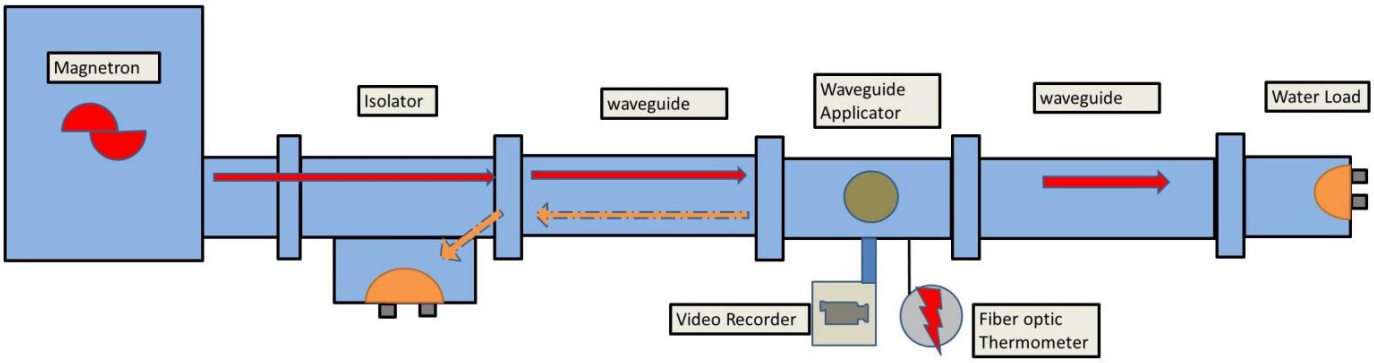
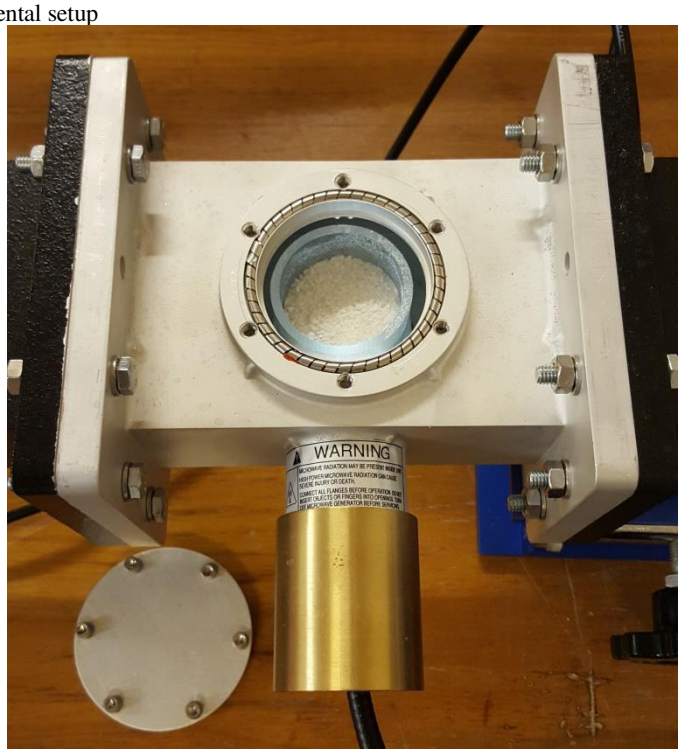
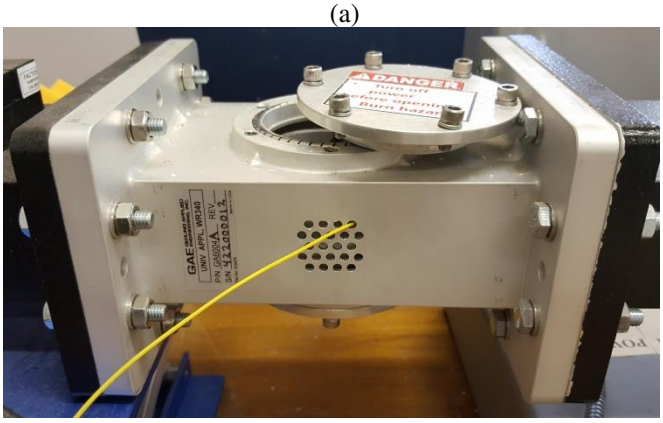


Fig 1 Waveguide heating experimental setup



(a)



(b)

Fig 2 Waveguide applicator (a) Top view with Acrylic sample holder & HLAS sample (b) Side view- 3mm perforations for fibre optic probes

Table 1 : Dielectric properties of LAS samples

% equilibrium Relative Humidity	Dielectric constant	Loss factor
18.4 % eRH (~1.8 % water)	2.03	0.0021
33 % eRH (~3.7 % water)	2.21	0.045
64% eRH (~8 % Water)	2.45	0.11

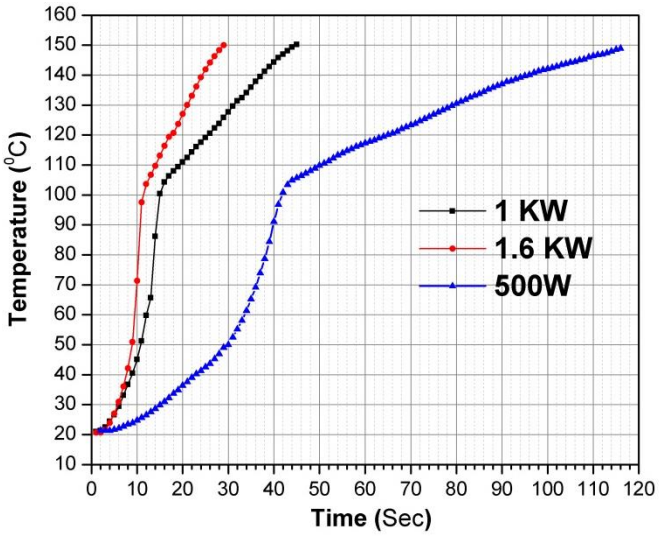


Fig 03 Temperature vs time graph for 3 different input power levels

Table-2 Bulk density & residence time for samples exposed to different input powers

Variable	Sample 1	Sample 2	Sample3
Sample Mass	18 g	18 g	18 g
Power input	500 W	1000 W	1600 W
Residence time	116 s	44 s	29 s
Max temp	150 ^{0}C	150 ^{0}C	150 ^{0}C
Bulk density before	959 g/l	959 g/l	959 g/l
Bulk density after	902 g/l	854 g/l	823 g/l

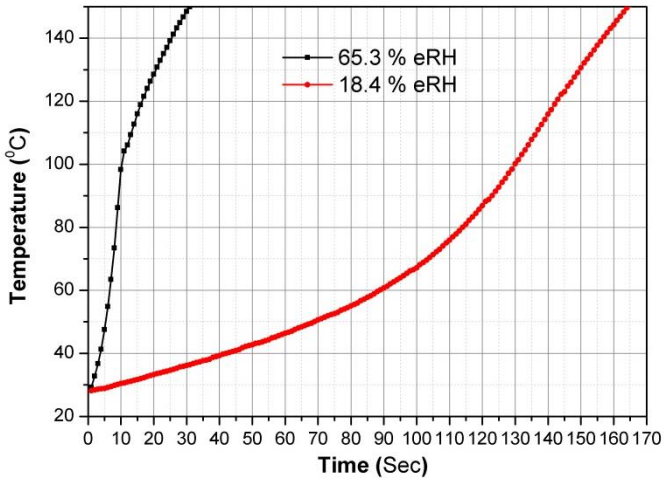


Fig 04 Temperature vs time graph for 2 different initial moisture content samples heated with 1KW input power

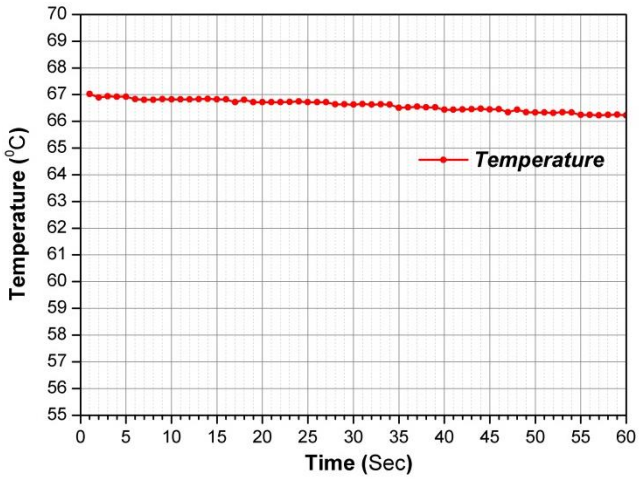


Fig 5 Temperature vs time graph for pre-heated (dry) sample at 500 W

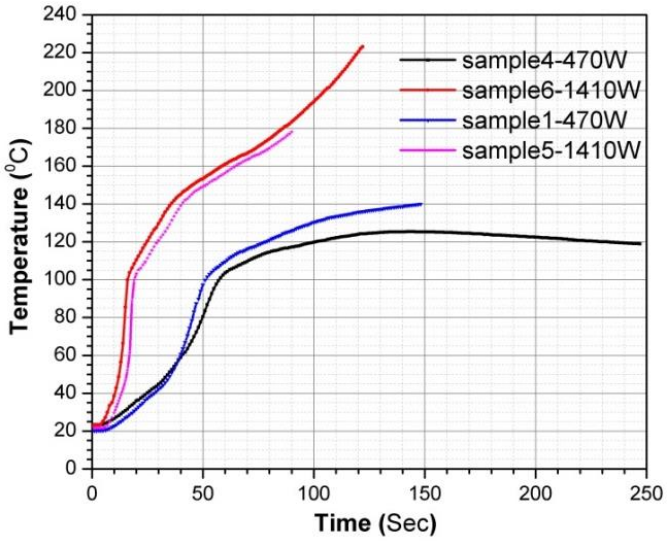
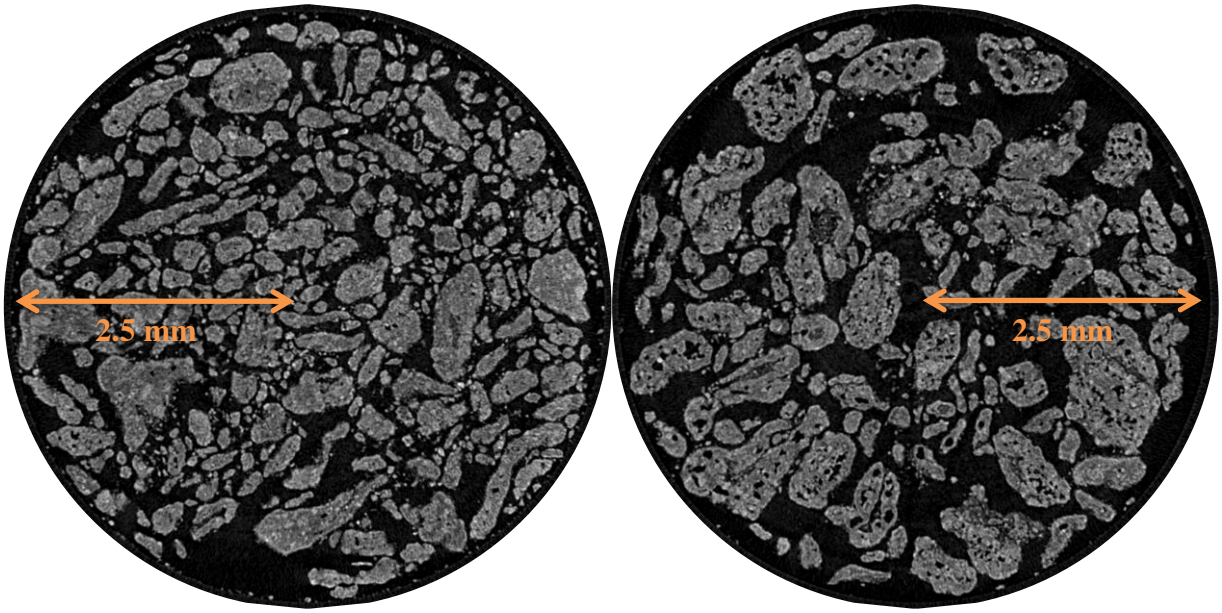


Fig 6 Temperature vs time graph for different residence time

Table-3 Bulk density & residence time for sample 1, 4, 5 & 6

Variable	**Sample1**	**Sample4**	**Sample5**	**Sample6**
Weight	10g	10g	10g	10g
Bulk density before	812 g/l	812 g/l	812 g/l	812 g/l
Power incident	470 W	470 W	1410 W	1410 W
Max temperature	139 C	125 C	167 C	220 C
Residence time	**150 sec**	**240 sec**	**90 sec**	**120 sec**
Bulk density after	710 g/l	663 g/l	634 g/l	592 g/l



(a) (b)

Fig 7 Micro_CT images of (a) untreated and (b) Microwave puffed LAS sample

Table-4 3D volume and porosity of untreated and microwave puffed LAS sample.

Measurements	**Untreated/ Unpuffed sample**	**Microwaved sample**
	0W	**1.6kW**
Object volume	1.110E+10um^3	1.056E+10 um^3
Percent object volume	97.73%	93.70%
Volume of closed pores	1.037E+08 um^3	2.485E+08 um^3
Closed porosity	0.93%	2.30%
Volume of open pore space	1.536E+08 um^3	4.621E+08 um^3
Open porosity	1.35%	4.10%
Total volume of pore space	2.572E+08 um^3	7.106E+08 um^3
Total porosity	2.27%	6.30%

Porosity Differences	4.04%