

## Structuring and calorie control of bakery products by templating batter with ultra melt-resistant food-grade hydrogel beads

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We report the use of a temperature insensitive, food-grade hydrogel to reduce the caloric density of pancakes that were prepared at temperatures much higher than the boiling point of water. This cheap, facile method utilises a mixed agar-methylcellulose hydrogel, which was blended to produce slurry of hydrogel microbeads. The pancake batter was mixed with a controlled volume percentage of slurry of hydrogel beads and cooked. From bomb calorimetry experiments, the composites were found to have a reduced caloric density that reflects the volume percentage of hydrogel beads mixed with the batter. Using this procedure, we were able to reduce the caloric density of pancakes by up to  $23 \pm 3\%$  when the volume percentage of hydrogel beads initially used was 25%. The method is not limited to pancakes and could potentially be applied to various other food products. The structure and morphology of the freeze-dried pancakes and pancake-hydrogel composites were investigated and pores of a similar size to the hydrogel beads were found, confirming the gel beads maintained their structure during the cooking process. There is scope for further development of this method by the encapsulation of nutritionally beneficial or flavour enhancing ingredients within the hydrogel beads.

### Introduction

The rapid incline in cases of obesity is cause for major concern due to the health issues that are attributed to it. Cardiovascular diseases, type 2 diabetes, hypertension and certain cancers are some of the numerous comorbidities associated with this global epidemic.<sup>1</sup> The impact of obesity is recognised by health organisations, the food industry and consumers alike. A main culprit of excessive weight gain is the over consumption of food with low nutritional benefits and high fat and refined sugar content. This has led to major investment into the development of new, healthier food products.<sup>2</sup> A challenge in producing these reformulated, nutritionally beneficial foods is preserving their physical and sensorial properties whilst maintaining a flavour that will appeal to consumers.

Composite foods, foods comprising of two or more different materials, are widely available. Emulsions, dispersions and emulsion-filled gels are all forms of composite foods. Hydrocolloids are frequently used in their formulation as thickeners, emulsifiers and gelling agents.<sup>3</sup> Xanthan gum is used as a fat replacement in the formulation of low-fat mayonnaise where its role is to increase the viscosity of the aqueous phase,

increasing shelf life.<sup>4</sup> The techniques in which hydrocolloids can be employed in food formulations is constantly expanding, with a recent study showing that chia mucilage gel can partially replace vegetable fat in bread and margarine in cakes to reduce fat content and caloric density.<sup>5</sup>

Another way hydrocolloids have been utilised is in the preparation of hollow salt particles. This involves formulation of salt-hydrogel liquid marbles by producing an aerosol of a hot liquid gelling agent and passing it through a cold column, which yields hydrogel beads, and then rolling these gel beads in salt microcrystals. After drying the salt-hydrogel composites, hollow salt particles could be obtained. These salt particles exhibited an increased dissolution rate when compared to table salt, showing they would have an increased perception of saltiness at the same concentration which could lead to a decreased salt intake.<sup>6,7</sup>

The incorporation of hydrogel beads into food products has previously been employed in the formulation of chocolate-hydrogel composites, through emulsification. When incorporating water droplets alone into chocolate, the mechanical properties will suffer when their volume fraction is too high. This led to gelling agents being added to the aqueous phase. One group utilised gelatine to gel the water droplets, where the hydrogel network within the droplets contributes to the overall strength of the composite.<sup>8</sup> A drawback of gelatine, however, is that it is derived from animals, making it unsuitable to a large consumer base. Agar, an abundant biopolymer derived from red seaweed, has been used as an alternative to gelatine to produce suspensions of up to 80% v/v aqueous microgel agar particles dispersed within chocolate.<sup>9</sup>

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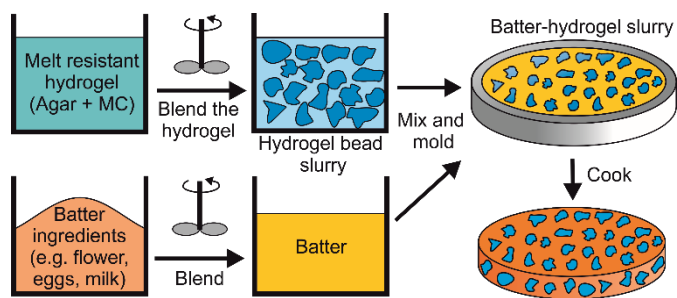


Fig. 1. Schematics of the process of templating batter with hydrogel slurries of highly melt-resistant hydrogel.

Whilst using agar in chocolate formulations is an excellent method to reduce fat content, an issue when using it for the design of other food formulations however, is that it melts at around 85 °C.<sup>10</sup>

We developed a two-component hydrogel comprising of agar and methylcellulose (MC) that maintains its structural integrity well above the melting point of agar. This combination of food-grade hydrocolloids maintains the advantage of being derived from nature, does not require salts for gelation and, once gelation has occurred, will not melt at temperatures above 100 °C. This resistance to melting at elevated temperatures allows the incorporation of hydrogel beads of this composition into food products that require high temperatures during preparation. In this paper, we demonstrate the structuring and calorie control by the use of melt-resistant hydrogel beads of agar-MC (Agar-MC) hydrogel blended in a pancake batter that were cooked at temperatures above 100 °C. The scheme for preparation of this type of batter-hydrogel slurry composite is presented in Fig. 1. It can be easily applied for a range of bakery products. We have measured the caloric density of a control sample of pancakes and pancake-hydrogel bead composites. The reduction in caloric density of the pancake composites closely reflects the initial volume percentage of hydrogel beads that were incorporated within them. We have studied the microstructure of the pancakes and pancake-hydrogel composites *via* SEM and have seen pores in the freeze-dried composites consistent with the size of the hydrogel beads, confirming that these elements of the composite maintained their structure during the cooking process.

## Experimental

### Materials

Agar (food grade) was purchased from Special Ingredients, MC (SG A7C, food grade) was kindly supplied by The Dow Chemical Company®. Benzoic acid (1 g pellets) were purchased from Parr Instruments company. Eggs, milk, flour and butter were purchased from a local shop. Deionised water (MilliQ purification unit, Millipore) was used in all experiments.

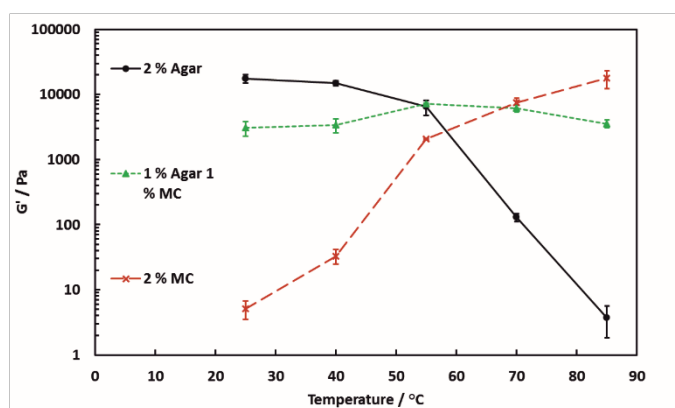


Fig. 2. The storage modulus of 2.0% w/v agar hydrogel, 2.0% w/v MC hydrogel and 1% w/v : 1% w/v Agar:MC hydrogel as a function of the temperature.  $G'$  is measured at 100 Pa and averaged over three samples. Error bars are the standard deviation.

## Methods

### Rheology measurements

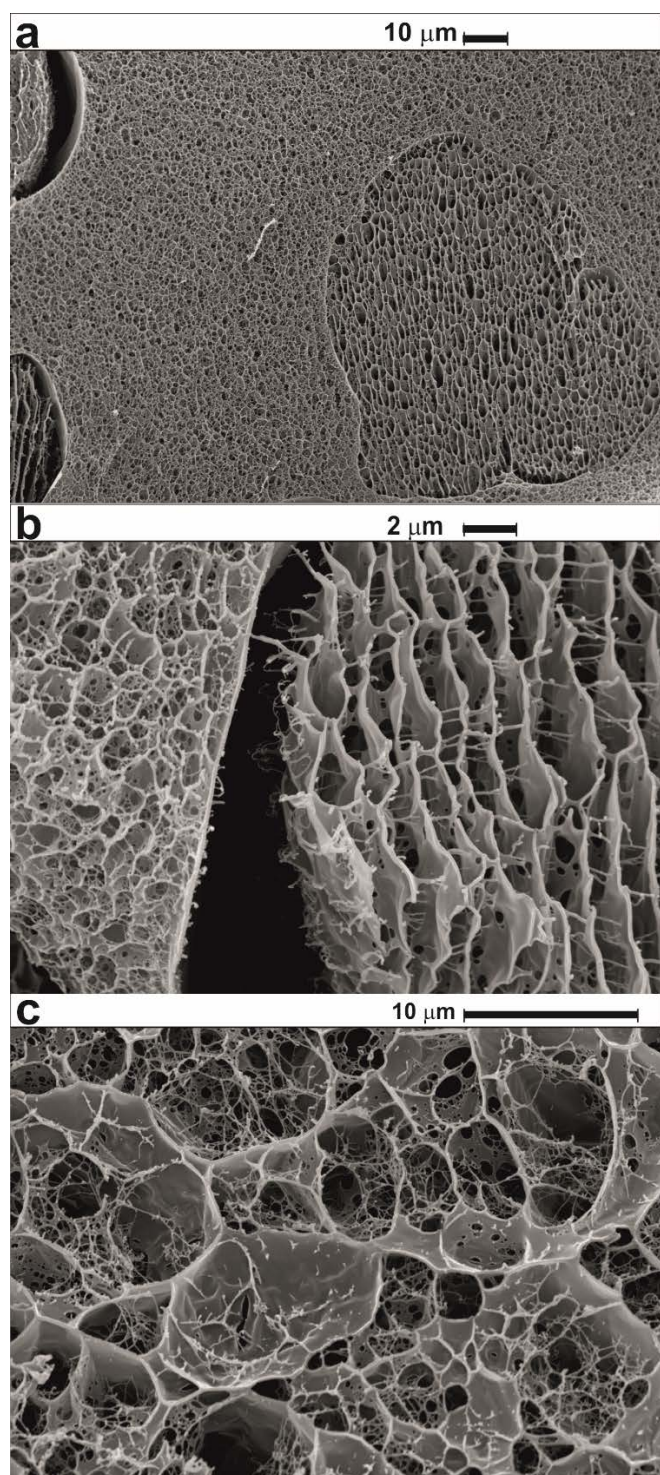
Rheological measurements were performed on Bohlin CVO 120 rheometer (Malvern Instruments) with a 20 mm diameter parallel plate geometry, a gap size of 2200  $\mu\text{m}$  and temperature control by the use of a Peltier plate. Oscillatory tests were performed and the storage modulus at 25, 40, 55, 70 and 85 °C was measured at increasing shear stress from 1 - 100 Pa. Prior measurement, the sample was equilibrated for 30 seconds at each temperature. Evaporation of water from the agar and Agar-MC hydrogels during testing was minimized by adding a thin layer of low viscosity silicone oil (40 cP) to their surface.

### Preparation of hydrogel beads

To produce the hydrogel, 500 cm<sup>3</sup> of water was heated to 97 °C using a water bath. Agar powder (5.0 g, 1.0% w/v) and MC powder (5.0 g, 1.0% w/v) were added and the solution was homogenised using an Ultra-Turrax homogeniser for 15 minutes. The temperature of the water bath was then decreased to 50 °C whilst the solution was continually agitated. Once the desired temperature was reached, the solution was removed from the water bath and placed in a fridge (4 °C) overnight for the agar to set and the MC to fully dissolve. The resulting hydrogel was then transferred to a Tefal food processor minipro food blender (a 500 W blender with 3 stacked blades) and blended at full power to produce a slurry of hydrogel beads.

### Preparation of pancake-hydrogel beads composites

Pancake batter was prepared using two UK standard medium sized eggs, 110 g of plain flour and 300 cm<sup>3</sup> of semi-skimmed milk. The ingredients were combined using a handheld blender (Goodman's Cuisine Blender). The batter was mixed with various controlled volume percentages of slurry of hydrogel beads to an overall volume of 40 cm<sup>3</sup>.



**Fig. 3.** SEM images of a freeze-dried samples of Agar-MC hydrogel (1% w/v – 1% w/v) after it was flash-frozen in slush nitrogen, fractured and then the water was sublimated at  $-60\text{ }^{\circ}\text{C}$  for 20 minutes. (a) and (b) SEM images show co-existing regions of a MC-like structure surrounded by an agar-like structure. (c) Higher magnification image of the agar-like structure of the melt-resistant hydrogel.

A pan was heated on a hot plate until it reached  $170\text{ }^{\circ}\text{C}$  (temperature was monitored using an infrared thermometer with laser target) and then butter (0.3 g) was melted onto the pan. The pancake batter containing hydrogel beads was then

poured onto the pan and heated for 4 minutes on each side. Three different volume percentages of slurry of hydrogel beads were used (10%, 17.5% and 25%) as well as a control sample containing only pancake batter.

#### Calorimetry

Samples of approximately equal mass and shape were cut from each composite and their mass recorded. They were then placed in open polypropylene tubes and frozen in liquid nitrogen for 15 minutes. Subsequently, the tube was covered with a tissue, transferred to a freeze drier and dried for 72 hours ( $-84\text{ }^{\circ}\text{C}$ , 1.6 Pa). The mass of the dried samples was recorded. They were ground to a fine powder and pellets of roughly 1 g mass were pressed. A 1341 Plain Jacket Bomb Calorimeter (Parr Instruments Company) was used for all measurements. Benzoic acid was used as a standard to calculate the calorimeter constant as its heat of combustion is well documented.<sup>11</sup> A pellet was placed into the crucible. Fuse wire (Ni-Cr) was cut, weighed, then wrapped around the bomb terminals and put into contact with the sample. Water ( $1.00\text{ cm}^3$ ) was added to the bomb which was then assembled and pressurised with oxygen (20 atm). The chamber was filled with water ( $2000\text{ cm}^3$ ) and the bomb was placed inside. Two electrodes were connected to the bomb, then a lid with a stirrer and digital thermometer of resolution of  $0.001\text{ }^{\circ}\text{C}$  (model 6775, Parr Instruments Company) connected was placed on top of the calorimeter and the temperature was measured as a function of time. After equilibration, the samples were ignited. The temperature was continually recorded until it stabilised. The bomb was then removed from the chamber, depressurised and washed for next use. Any remaining fuse wire was weighed. This method was applied with the pancake and pancake-hydrogel samples.

#### Microscopy analysis

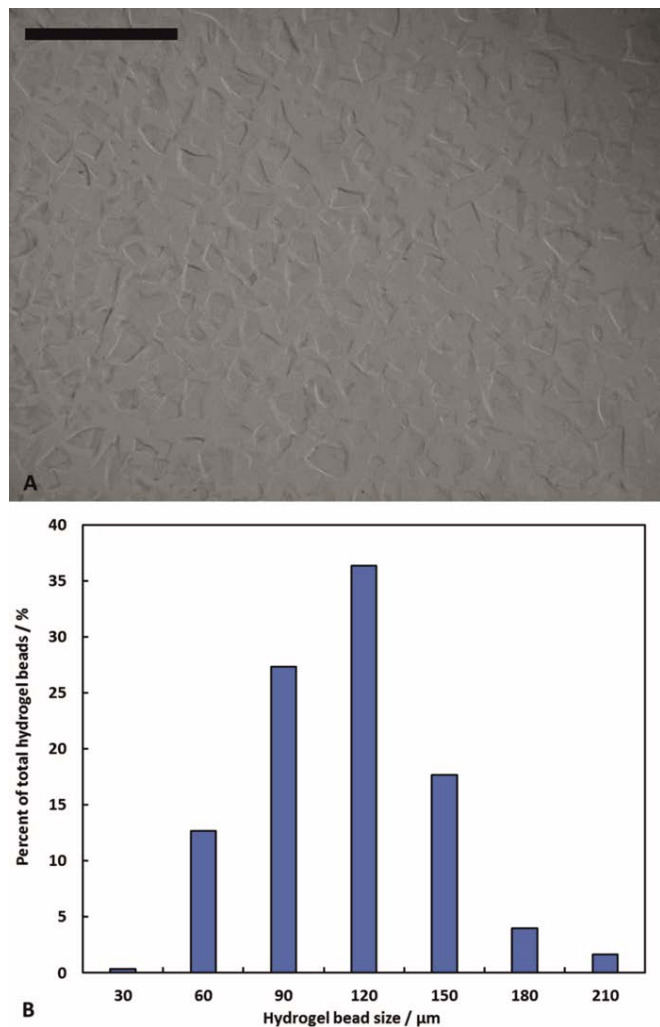
Hydrogel beads were dispersed in water and images taken in bright field light using an Olympus BX-51 optical microscope fitted with DP70 CCD camera. A sample size of 300 hydrogel beads were measured manually and the size analysis was done using Image J software. Freeze-dried samples of pancake and pancake-hydrogel composites were gold coated and viewed using a Hitachi TM-1000 scanning electron microscope. To visualise the Agar-MC hydrogel, a sample was mounted onto a copper stub, flash-frozen in slush nitrogen ( $\sim -210\text{ }^{\circ}\text{C}$ ), transferred to a preparation stage at  $-140\text{ }^{\circ}\text{C}$  and fractured with a blade. After sublimating at  $-70\text{ }^{\circ}\text{C}$  and 2–4 Pa for 7 minutes, it was coated with platinum with an integrated sputter coater. Finally, they were transferred to the SEM chamber of a Zeiss Evo 60 Scanning Electron Microscope at  $-140\text{ }^{\circ}\text{C}$  and visualised with an accelerating voltage of 15 kV.

## Results and discussion

#### Rheology and microstructure of Agar-MA hydrogel

We measured the rheological behaviour of the Agar-MC hydrogel as a function of temperature. The two-component

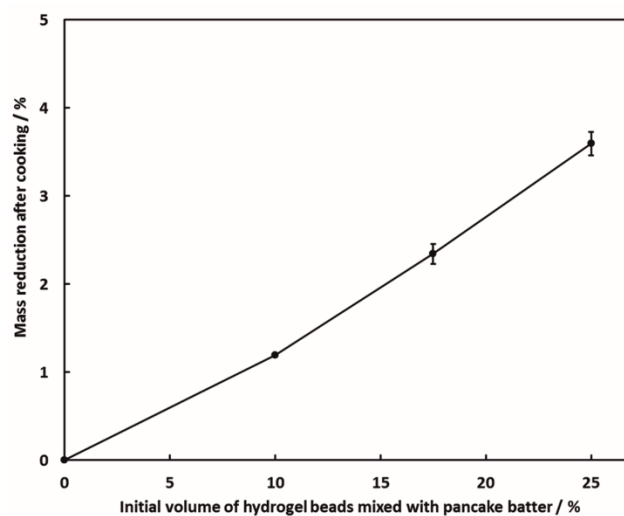
1%:1% Agar-MC hydrogel showed characteristics of both agar and MC, depending on the temperature range. Figure 2 shows that the Agar-MC hydrogel has a fairly constant storage modulus upon increasing the temperature from 25 °C up to nearly 90 °C with a maximum around 55 °C which is above to the gelling point of MC. Note that the Agar-MC hydrogels are significantly stronger than agar hydrogel alone at 85 °C.



**Fig. 4.** (A) An optical microscopy image of Agar-MC hydrogel beads dispersed in water. They were blended at full power with a Tefal food processor Minipro for 10 minutes. The scale bar is 500  $\mu\text{m}$  and the magnification was 4x. (B) The size distribution diagram of these beads. A sample size of 300 beads within several fields of view was used and their sizes measured using Image J software. The histogram represents the number percentage of the hydrogel beads of various sizes.

At low temperatures, its rheological behaviour is similar to that of agar hydrogel while at higher temperatures, MC is the polymer dominating the hydrogel rheology. Our experiments on thermal stability of Agar-MC hydrogels (not presented here) show that the hydrogel does not melt even after submerging in water and autoclaving at 121 °C or after heating in an oven at 150 °C for 45 min. Figure 3 shows SEM images of freeze-dried samples of the melt-resistant hydrogel after flash freezing and freeze-drying under vacuum. The microstructure of this

hydrogel involves two phase regions with agar-like structure (continuous phase) surrounding domains of MC-like structure (Figs. 3A, 3B). Both phases contain network features that look more interlinked than the microstructure of agar or MC hydrogels alone at the same concentration.



**Fig. 5.** Mass reduction of the pancake-hydrogel composites after the cooking process as a function of the initial volume percentage of hydrogel beads in the composite.

#### Hydrogel bead size analysis

In order for the hydrogel beads in the pancake-hydrogel composites to be undetectable during consumption, their size must be on the micron scale.<sup>12</sup> Therefore it was important to achieve control over the size of the beads produced. Hydrogel bead size was measured as a function of blending time and their size distribution obtained by analysing microscope images of the gel beads dispersed in water. Due to the irregular shape of the beads, they were measured horizontally through their widest section. After blending for 10 minutes, the average diameter of the hydrogel beads was  $100 \pm 30 \mu\text{m}$  where the standard deviation represents their polydispersity. Their morphology and size distribution can be seen in Figs. 4A and 4B. It was seen that further increasing the blending time showed insignificant reduction in the size distribution of the beads. In future studies, we will concentrate on reducing the average diameter and polydispersity of the hydrogel beads in an effort to produce composites with a smoother mouthfeel.

#### Formulation of pancake-hydrogel composites

The mass reduction of the composites after preparation,  $R_{m(\text{prep})}$ , was calculated using the following formula:

$$R_{m(\text{prep})} = \left(1 - \frac{m_c}{m_0}\right) \times 100 \%. \quad (1)$$

Here  $R_{m(\text{prep})}$  is the mass reduction after preparation (%),  $m_c$  is the mass of the pancake-hydrogel composite and  $m_0$  is the mass of pancake control sample containing no hydrogel beads.

It is shown in Fig. 5 that there are some small mass reductions of the composites and this increases with an increase in the volume fraction of hydrogel beads initially present in the pancake batter. The probable causes for this are two-fold.



**Fig. 6.** An optical photograph of a pancake (A) and pancake-hydrogel composites initially containing (B) 10%, (C) 17.5% and (D) 25% by volume slurry of A-MC hydrogel beads. They were prepared by mixing a controlled volume percentage of slurry of hydrogel beads with pancake batter, then heated at 170 °C for 4 minutes on each side.

Firstly, hydrogel beads on the surface of the pancake batter are able to lose mass due to the evaporation of water. Furthermore, hydrogel syneresis and loss of water from the hydrogel beads into the pancake batter could occur due to an osmotic pressure gradient.

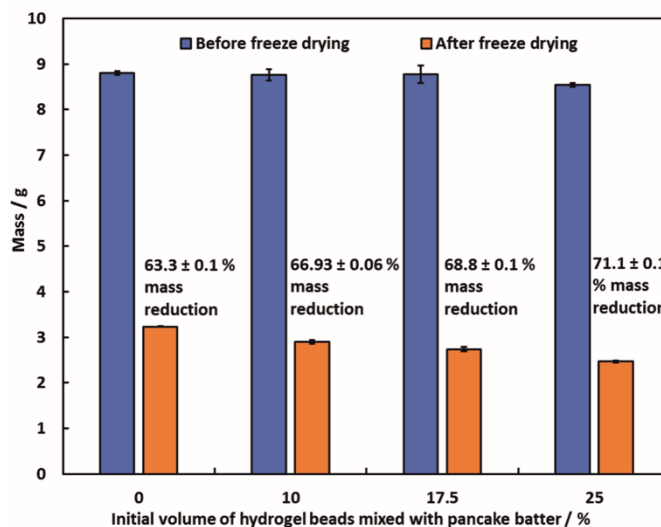
The MC present in the hydrogel beads gels upon heating which maintains their structure, whereas if the gel beads were to melt, mass transport through the batter could occur which would lead to further evaporation of water. In Fig. 6, the appearance of the pancake-hydrogel composites are shown to have a very close likeness to the pancake control sample so they will be as aesthetically appealing to the consumer as regular pancakes that contain no hydrogel beads.

Before the caloric density of the pancakes and pancake-hydrogel composites could be measured, they needed to be freeze dried to remove all moisture and allow easy ignition of the sample inside the bomb. The samples were weighed before and after freeze drying and their reduction in mass ( $R_m$ ) calculated:

$$R_m = \left(1 - \frac{m_{dry}}{m_{wet}}\right) \times 100 \% \quad (2)$$

Here  $R_m$  is the reduction in mass (%),  $m_{dry}$  is the mass after freeze drying measured in grams and  $m_{wet}$  is the mass of the sample prior to freeze drying measured in grams. The masses of the samples before and after freeze drying, along with overall

mass reduction, are shown in Fig. 7. It was found that with an increase in initial volume fraction of hydrogel beads present in the composite that the overall mass reduction was larger.



**Fig. 7.** The mass of the samples of pancake and pancake-hydrogel composites before and after freeze-drying for 72 hours. The mass reductions were calculated by equation 2. They were measured in triplicate and the values are an average with the error bars showing their standard deviation.

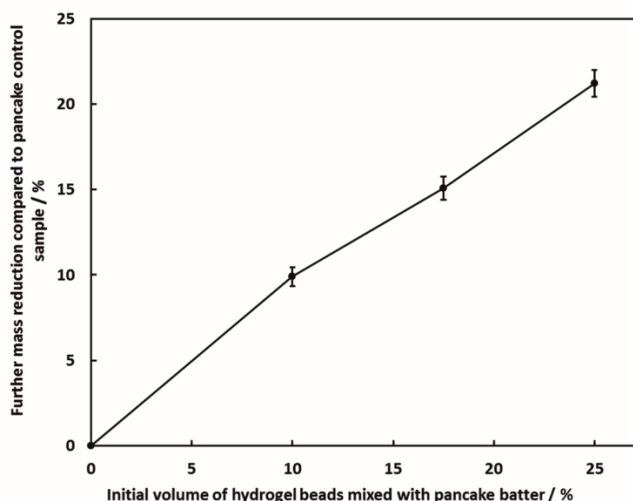
Further to this, it was possible to calculate the extra mass lost in the composites compared to the control sample due to the presence of the gel beads. This was done using the following equation:

$$R_{m(gb)} = \frac{R_{m(c)} - R_{m(std)}}{100 - R_{m(std)}} \times 100 \% \quad (3)$$

Here  $R_{m(gb)}$  is the extra mass reduction due to the presence of gel beads initially in the composite (%),  $R_{m(c)}$  is the mass reduction of the composite (%) and  $R_{m(std)}$  is the mass reduction of the standard pancake containing no hydrogel beads (%).

Figure 8 shows the percentage mass decrease due to the presence of the hydrogel beads. It can be seen that it closely reflects the initial volume of hydrogel beads present in the composite, being offset by 0.1% when the initial volume of hydrogel beads was 10%, 2.5% when the initial volume was 17.5% and approximately 4% when the initial volume was 25%. This can be attributed to evaporation of water from hydrogel beads at the surface and loss of water from the hydrogel beads due to osmotic effects.

The freeze dried samples were visualised using a scanning electron microscope. The microstructure of both the control sample and the pancake-hydrogel composites were studied and their images are shown in Fig. 9A-9C. Figure 9A shows the pancake matrix alone, whereas Figs. 9B and 9C show the pancake matrix with pores of a similar size to the hydrogel beads that were incorporated within the pancake batter.



**Fig. 8.** This shows the further mass reduction of the pancake-hydrogel composites compared to the pancake control sample due to the presence of hydrogel beads. Each data point is the average of three measurements with the error bars showing the standard deviation.

#### Caloric density of pancake-hydrogel composites

Calibration of the calorimeter was done using benzoic acid which has an energy of combustion of  $(-26433 \pm 3) \text{ J g}^{-1}$ . This was measured three times to obtain a constant that corresponds to the calorimeter when 2000 g of water was used. The calibration results gave a calorimeter constant of  $10500 \pm 200 \text{ J K}^{-1}$  or  $2.51 \pm 0.05 \text{ kcal K}^{-1}$ .

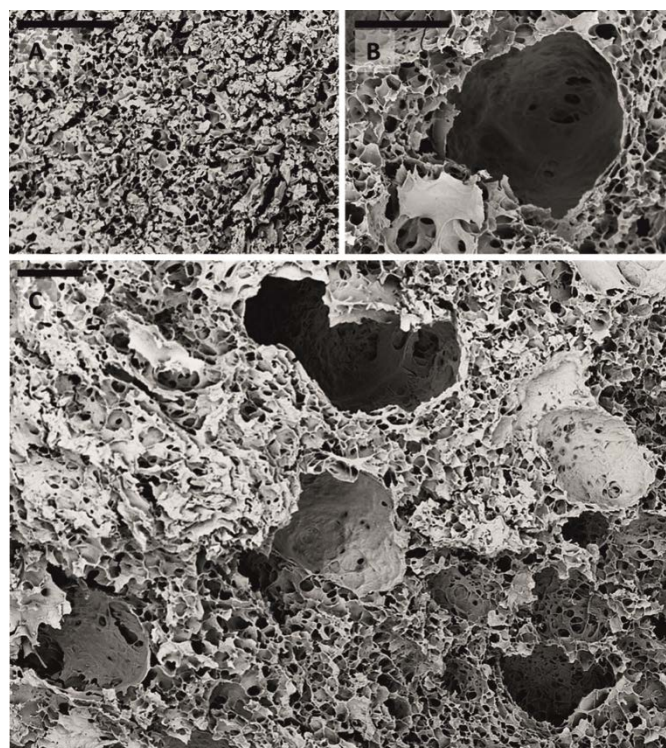
The freeze-dried samples were ground into a fine powder and pellets of approximately 1 g were pressed. The standard pancake as well as the pancake-hydrogel composites were measured three times each and the caloric density was calculated:

$$q = \frac{c_v \times \Delta T - m_{\text{wire}} \times \Delta H_{c(\text{wire})}}{m_{\text{sample}}} \quad (4)$$

Here  $q$  is the caloric density ( $\text{kcal g}^{-1}$ ),  $c_v$  is the calorimeter constant ( $\text{kcal K}^{-1}$ ),  $\Delta T$  is the change in temperature (K),  $m_{\text{wire}}$  is the mass of fuse wire used (g),  $\Delta H_{c(\text{wire})}$  is the heat of combustion of the fuse wire ( $\text{kcal g}^{-1}$ ) and  $m_{\text{sample}}$  is the mass of the sample (g) which is the wet mass that is represented by the mass of the dry pellet.

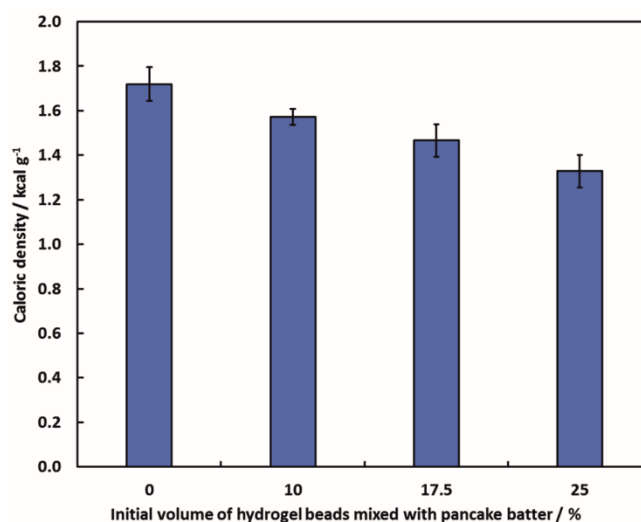
Figure 10 shows that the caloric density decreases with increasing volume fraction of hydrogel beads initially in the composite. This is due to the fact that the beads are made from a hydrogel containing 98% water which contains zero calories. The small amount of biopolymer present is mostly indigestible; it is a soluble dietary fibre known to be a satiety aid.<sup>13,14</sup> The reduction in caloric density was then calculated using the following equation:

$$q_{\text{red}} = \left(1 - \frac{q_c}{q_{\text{std}}}\right) \times 100\% \quad (5)$$

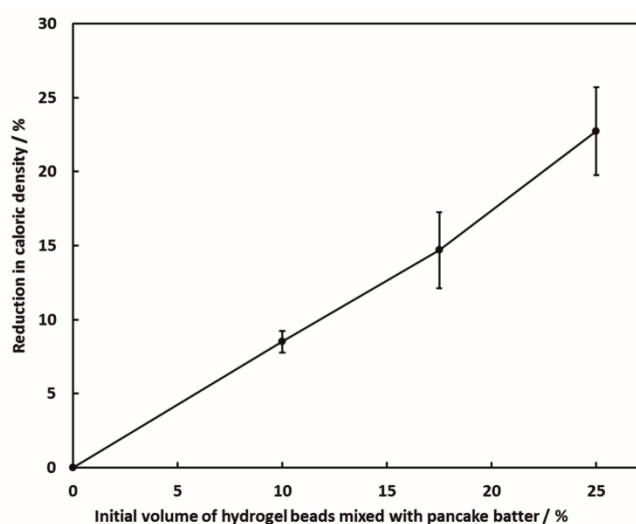


**Fig. 9.** SEM images of freeze-dried pancake (A) and pancake-hydrogel composites (B) and (C). The composites were produced with an initial volume percentage of 17.5% of Agar-MC hydrogel beads (1% w/v – 1% w/v). (C) corresponds to a larger scale of view than (B). All scale bars are 100  $\mu\text{m}$ .

Here  $q_{\text{red}}$  is the caloric density reduction (%),  $q_c$  is the caloric density of the pancake-hydrogel composites and  $q_{\text{std}}$  is the caloric density of the standard pancake containing no hydrogel beads.



**Fig. 10.** The caloric density of the pancake control sample and the pancake-hydrogel composites containing increasing volume percentages of A-MC hydrogel beads (1% w/v – 1% w/v). Each sample was measured three times to obtain an average caloric density, with the error bars showing the standard deviation.



**Fig. 11.** The reduction in caloric density of pancake-hydrogel composites compared to the pancake control sample as a function of the initial volume percentage of hydrogel beads mixed with the pancake batter. Each value is an average of three experiments, with the error bars being the standard deviation.

It can be seen in Fig. 11 that the caloric density reduction in the pancake-hydrogel composites closely follows the initial volume fraction of hydrogel beads in the composite. The incorporation of water into food decreases the caloric density by increasing the mass of the food whilst not adding any calories. Furthermore, addition of water to food has been shown to significantly increase fullness and suppress appetite in subsequent meals.<sup>15</sup> Here, we incorporate water into food in the form of hydrogel beads and we expect that it will have the same beneficial effects. An additional advantage gained from the use of hydrogel beads is the ability to encapsulate flavour or taste enhancing compounds, thereby allowing for a range of flavoured products to be formulated.

## Conclusions

We designed a new method for the reduction of caloric density in bakery products and have demonstrated it on pancakes. We utilised a highly melt resistant hydrogel consisting of agar and methylcellulose. Upon blending of this mixed hydrogel, we obtained a slurry of hydrogel microbeads. Controlled volume percentages of slurry of hydrogel beads were mixed with pancake batter and cooked to produce pancake-hydrogel composites. The caloric density of the composites was measured and when compared to a pancake control sample, it exhibited a reduction in caloric density comparable to the volume percentage of hydrogel beads incorporated within the composite. This method allowed controllable reduction in the caloric density up to  $23 \pm 3\%$ , when 25% by volume of slurry of hydrogel beads was mixed with the pancake batter prior to cooking. We have analysed the microstructure of freeze-dried pancakes and pancake-hydrogel composites and the composites showed pores on a similar length scale to the

hydrogel beads used, confirming that the hydrogel beads maintained their structure during the high temperature cooking process. We expect that this strategy is not limited to pancakes, and could find applications in a variety of food products, like waffles, crumpets, muffins, biscuits, cereal bars and many others. This method could be developed further by encapsulation of nutritionally beneficial or flavour enhancing ingredients within the hydrogel beads.

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