

Polymers

Carbohydrate

www.elsevier.com/locate/carbpol

Carbohydrate Polymers 69 (2007) 445-454

# Microwave-assisted moulding using expandable extruded pellets from wheat flours and starch

Jiang Zhou a,1, Jim Song a,\*, Roger Parker b

<sup>a</sup> Mechanical Engineering, School of Engineering and Design, Brunel University, Uxbridge, Middlesex UB8 3PH, UK
<sup>b</sup> Institute of Food Research, Norwich Research Park, Colney, Norwich NR4 7UA, UK

Received 29 November 2005; received in revised form 20 December 2006; accepted 5 January 2007 Available online 17 January 2007

#### Abstract

Extruded pellets made from wheat flour and purified wheat starch were expanded by applying microwave heating within moulds, a method known as microwave-assisted moulding (MAM). Selection of adequate mould material, pre-treatment of pellets and control of the initial loading of pellets in the mould cavity were found to be the key issues to achieve a uniformly foamed block with good integrity. Polytetrafluoroethylene (PTFE) was found to be an appropriate mould material for the MAM process. The bonding between foamed pellets in a block can be significantly enhanced by soaking the pellets in a NaCl solution before microwave foaming. There exists an optimum initial loading of pellets in the mould for a given pellet formulation, which allows sufficient expansion to achieve an acceptable extent of interfacial bonding and mould filling. The work demonstrated the feasibility of moulding starch block foams and potential applications of the foams in packaging and lightweight composites.

© 2007 Elsevier Ltd. All rights reserved.

Keywords: Microwave heating; Moulding; Wheat; Starch; Foams; Blocks; Expandable; Extruded pellets; Fusion and bonding

#### 1. Introduction

Foams are manufactured on a large scale for industrial applications using their mechanical, thermal and other properties (Gibson & Ashby, 1997). Polymer foams, in particular, have found numerous applications in building, automotive and packaging industries for weight reduction, thermal/acoustic insulation or cushioning.

In the last two decades, there has been an increasing interest in the development of biodegradable foams and products from renewable resources to reduce environmental impact of polymer foams. As packaging waste has been identified as a main target in reduction of wastes to landfill (Linstead & Ekins, 2001), development of biodegradable packaging materials has been highlighted as a priority in

government strategic policies (e.g., DETR, 2000 & DEFRA, 2004).

Considerable effort has been made to develop extruded starch foams as alternative to expanded polystyrene (EPS) for loosefill packaging application (Bastioli, Bellotti, Del Giudice, Lombi, & Rallis, 1994, 1998a, 1998b; Bellotti, Bastioli, Rallis, & Del Tredici, 1995, 2000; Bhatnagar & Hanna, 1995a, 1995b; Cha, Chung, Seib, Flores, & Hanna, 2001; Fang & Hanna, 2001a, 2001b; Lacourse & Altieri, 1989, 1991; Willett & Shogren, 2002). Extruded starch loosefill foams have now been commercialized and captured a significant share of the EPS loosefills market (Tatarka & Cunningham, 1998).

Moulded foam trays have been developed based on baking technology (Shogren, Lawton, & Tiefenbacher, 2002) and are commercially available (e.g., Potatopak, 2005). Similar to the process for making ice cream cones, starch, additives and water are mixed to form a batter which is then foamed by using the water as a blow agent and solidified by drying within heated moulds. The technology is somewhat

<sup>\*</sup> Corresponding author. Tel.: + 44 1895 266 692; fax: +44 1895 256 392. *E-mail address:* jim.song@brunel.ac.uk (J. Song).

<sup>&</sup>lt;sup>1</sup> Present address: College of Biological and Agricultural Engineering, Jilin University, Changchun 130022, PR China.

limited by the low drying rate to remove the moisture in the batter, which in turn restricts the maximum wall thickness of the foams. Similar technology known as PaperFoam® (PaperFoam, 2005) has also been developed based on injection moulding of a viscous mixture of pulp, starch and water into hot moulds to produce thin-shelled mouldings for, e.g., internal packaging of electronic products.

Technologies for producing more bulk starch forms have also been developed. Corrugated foam planks (Lye, Lee, & Chew, 1998) made by extrusion foaming of modified cornstarch have been shown to have good cushion performance. The high foam density and cost of the materials, however, have somewhat restricted their widespread applications in packaging. Block foams have been made by combination of extrusion foaming and adhesion technology (Wang, Song, & Kang, 2001, 2002). The foams are of lower density and made from low-cost wheat flours. When combined with other materials to form lightweight sandwich composites, mechanical properties and resistance to water attack can be significantly enhanced (Song, 2005).

To date, however, there is still a lack of starch-based foam products as an alternative to moulded EPS cushion blocks. This is because it is difficult to prepare moulded foam blocks directly using the above-mentioned technologies. Therefore, it is desirable to explore and develop new technologies to manufacture moulded block foam products from starch raw materials for this potentially huge market.

Traditionally, synthetic EPS packaging cushion blocks are manufactured using a steam moulding technique. EPS beads, pre-expanded using pentane as a blowing agent, are "matured" to allow diffusion of air into the cells and further expanded by injection of steam into the mould, which heats the beads and causes additional expansion of the cells. The expanded beads fill the mould cavity and fuse together to form an integral foamed moulding (BPF, 2005) & Landrock, 1995). This procedure provides a possible route to produce a foam block from starch materials in a mould cavity if expandable starch pellets can be made and appropriate foaming method can be identified. Our previous work (Zhou, 2004; Zhou, Song, & Parker, 2005) has demonstrated that extruded wheat flour and starch pellets can be freely expanded (without mould) by using microwave heating. Boischot, Moraru, and Kokini (2003) also studied effect of microwave heating on expansion of amylopectin extrudates. Microwave heating has a distinctive advantage over conventional heating by convection, conduction or radiation: utilising the moisture contained within starch pellets as a blow agent, the rapid volumetric heating of the starch pellets in a mould cavity would cause expansion of them to fill the mould and fuse at the contacts so as to form a coherent structure. This novel method for producing starch based foam blocks is referred to as microwave-assisted moulding (MAM).

This work reports the feasibility study on the MAM method and the factors that influence the moulding technology. These include selection of mould materials and methods to achieve appropriate mould filling, good cohe-

sion between foamed pellets and uniform temperature profile within the mould.

### 2. Experimental details

#### 2.1. Raw materials and additives

Two types of wheat flour were used (referred hereafter as Temple and Superfine). These raw materials were supplied by Heygates Limited (Northampton, UK). The as-received flours contain about 11–14% moisture. The Temple flour contains 9–10% protein while the Superfine flour contains 7.5–8.5% protein. A purified wheat starch, Meritena 200, was obtained from Amylum Europe NV, Belgium.

Hydrated calcium chloride (CaCl<sub>2</sub>·2H<sub>2</sub>O > 99.0% purity) and sodium chloride (NaCl, >99.5% purity) from Sigma-Aldrich, UK were selected as additives in the extruded Temple flour. Addition of salts may modify behaviour of starch in a number of ways throughout the pellet preparation and microwave-assisted moulding. Salt has been shown to improve the solution quality of the water such that starch gelatinizes at room temperature (Evans & Haisman, 1982). This would increase the disruption of the native structure of the starch in the flour and reduce its melt viscosity and greater expansion has been observed in saltcontaining pellets (Zhou et al., 2005). In addition, salts act as microwave energy absorbent (Metaxas & Meredith, 1983) and during subsequent microwave heating, much rapid foaming has been observed for pellets containing salts (Zhou et al., 2005). Further more, salts as humectant has been shown to increase moisture absorption and plasticize microwave-expanded foams (Zhou et al., 2005). The concentration of sodium chloride was 5.5% w/w and that of the calcium chloride was 10.5% w/w based on the asreceived Temple flour.

## 2.2. Extrusion of pellets

The extruded pellets used in this study were prepared using a 5-barrel-section Betol BTS40 twin-screw co-rotating extruder with 40-mm diameter screws and a L/D ratio of 22 fitted with a 4-mm diameter die. Barrel sections 1–5 were maintained at 60, 80, 120, 80 and 70 °C during the extrusion at a screw speed of 100 rpm. Flour and water (or the salt solutions) were fed independently to give an overall feed rate of 145 g/min and water contents of around 30.3–31.1% w/w (wet weight basis). Once stable extruder condition was achieved, as judged from constant torque and product output, extrudates were collected. The extrudates were manually cut into cylindrical pellets about 4 mm long. These were further dried in the laboratory atmosphere (about 23 °C, 40–50% RH) to moisture contents of 10–13 wt%.

## 2.3. Mould materials and mould design

In comparison with microwave foaming of starch pellets in an open space (Zhou et al., 2005), the expansion of the pellets takes place within the mould positioned within a microwave oven. A suitable mould material needs to be selected for moulding the extruded pellets into a block by using microwave heating. The following aspects should be taken into account in selection of the mould materials:

- (a) It must be sufficiently "transparent" to microwave penetration, or low in microwave energy absorption. This requires a low effective loss factor (Metaxas & Meredith, 1983) so as to maximise the microwave absorption by the moisture-containing pellets.
- (b) It must also be heat resistant and maintain good mechanical properties up to the foaming temperature, which could reach around 200 °C (Zhou, 2004). Heat is transferred from the pellets to the mould and this would intensify with the increase in pellet temperature and contact area with the die wall when the pellets expand and fill the mould.
- (c) It should have low adhesion to the molten starch material to avoid difficulties in demoulding.
- (d) It should have good processibility for manufacturing the mould (e.g., by machining).

Several materials were considered and trials were carried out. These include moulds from silica glass, polyethylene, wood and a glass fibre reinforced nylon composite. Properties available in relation to the selection criteria are listed in Table 1. Glass is a material exceptionally low in the effective loss factors but it is brittle and difficult to process. The good adhesion with starch also gives rise to difficulties in demoulding. Wood, like paper products, often contains considerable moisture, which increases further with absorbed moisture from the foaming pellets. The absorbed moisture dissipates microwave energy (as shown from data for water in Table 1) and reduces the effectiveness of heating of the starch pellets. The relatively rough surface of wood moulds also readily adheres to the molten starch resulting in demoulding difficulties. Polyethylene (PE) is very low in the effective loss factor and resists adhesion to starch. However, the maximum service temperature of PE is low and it was found that heat from the pellets softened the mould and caused significant distortion under

the internal pressure from the foaming pellets. The glass fibre reinforced polyamide is relatively "opaque" to microwave and thus dissipates considerable microwave energy. Similar to PE, the service temperature is low and the mould was softened during moulding. It was found that polytetrafluoroethylene (PTFE) performed very well, exhibiting all the required properties. PTFE absorbs very little microwave energy, and can be effectively preheated by hot air to 180 °C without softening, a very important feature for achieving uniform foaming throughout a block (as will be discussed further). Furthermore, the non-stick characteristic to the starch materials allowed mouldings to be ejected with ease.

Fig. 1 shows the PTFE mould used in this work, which was machined from an extruded rod. The mould consists of three parts: the top and bottom caps of the mould can be screwed on to the cylindrical tube with 58 mm inner diameter and 13 mm wall thickness. 57 mm diameter discs with different thickness are used to adjust the height of cavity of the mould so that blocks with heights ranging from 22 to 48 mm can be moulded. A 2-mm diameter hole at the centre of the top cap allows steam to escape from the mould during foaming, which reduces the internal pressure in the mould to achieve better mould filling.

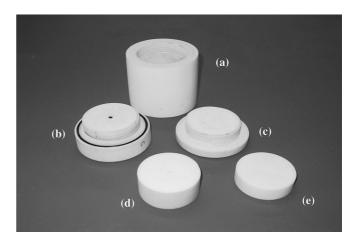


Fig. 1. The PTFE mould used in this work. (a) The central cylindrical tubular mould; (b) and (c) the end caps; (d) and (e) discs for height adjustment of blocks.

Table 1
Selected materials properties for moulds in MAM processing

Materials	Lose factor <sup>a</sup> ( $\varepsilon$ " <sub>eff</sub> )	Max. service temperature <sup>b</sup> (°C)	Tensile strength <sup>c</sup> (MPa)	Processibility	Adhesion to molten starch
Glass (96% SiO <sub>2</sub> )	0.00023	897–1397	45–155	Poor	High
Wood/paper	$0.22^{d}$	117–137	60-100	Medium	Medium/high
Polyethylene	0.0024	125–132	21–45	Good	Low
Polyamide	0.043	73–87	90-165	Good	Low
Glass reinforced polymers	_	140-220	138–241	Poor	Low/medium
PTFE	0.0003	250–271	20–30	Good	Very low
Water (distilled)	1.2	_	_	_	_
0.5 M NaCl	269.0	_	_	_	_

<sup>&</sup>lt;sup>a</sup> Measured at 25 °C and 10<sup>9</sup> Hz (Metaxas & Meredith, 1983).

<sup>&</sup>lt;sup>b</sup> CES (2005).

<sup>&</sup>lt;sup>c</sup> Measure at room temperature (CES, 2005).

<sup>&</sup>lt;sup>d</sup> Estimated from data in Metaxas and Meredith (1983) for paper.

### 2.4. Microwave foaming

Foaming tests were carried out in a combined microwave oven (Sharp R-8720M, 1000 W), which allows a combination of convection hot air and microwave heating. The closed mould loaded with pellets was positioned at the centre of the rotating dish in the microwave oven and heated by microwave at 100% power of 1 kW. About 35–60 s was required to foam the pellets depending on their compositions. Excessive microwave heating led to burning of the pellets.

## 2.5. Characterisations of the block foams

Block samples were conditioned in the laboratory atmosphere (about 23 °C, 40–50% RH) for two weeks before they were weighed for density calculations. Volume of the foams was obtained as follows: A foam block was placed in a graduated cylinder, then a known volume of glass beads (60-mesh, BDH Chemicals Ltd., Poole, England),  $V_{\rm gb}$ , sufficient to embed the foam was poured into the cylinder. The total volume of glass beads and the foam,  $V_{\rm t}$ , was recorded after tapping the cylinder for one minute. The volume of foam,  $V_{\rm f}$ , was calculated by subtracting  $V_{\rm gb}$  from  $V_{\rm t}$ . Care was taken to block any voids between foamed pellets to avoid the penetration of glass beads.

The foamed block with diameter of around 60 mm and height of 22.5–23.5 mm were conditioned at 20–22 °C and 50% relative humidity for two weeks before mechanical testing. A testing machine (H10KT, Hounsfield Test Equipment Ltd., Surrey, England) was used to compress the samples to 50% deformation at a rate of 10 mm/min. Compressive stress at 10% deformation and compressive elastic modulus of the samples were calculated based on British Standard BS ISO 844:2001 (E). Sectioned block samples coated with gold were then examined with SEM (250 MK2, Cambridge, England).

## 3. Results and discussion

#### 3.1. Adhesion of foamed pellets

When pellets are foamed and deformed at contacts with neighbouring pellets, the interfaces become multi-faceted as shown in Fig. 2. To form an integral block and have adequate strength, the foamed pellet block must have sufficient bonding strength at these contact interfaces. Such bonding may be achieved by forming intimate contact so as to enhance the molecular attraction across the contact surfaces and adhesion by interfacial diffusion across the contacts facilitated by sufficient molecular mobility (Kinloch, 1986). Both are promoted by maintaining water-plasticized interfaces and elevated temperature.

Preliminary moulding tests using pellets containing about 10% w/w water (Zhou, 2004) showed poor interface bonding strength, although intimate interfacial contacts had been established. The moulded blocks were weak and

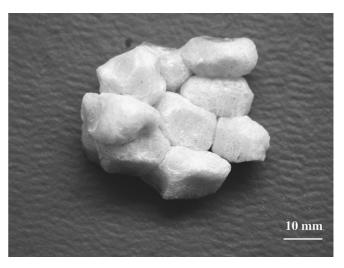


Fig. 2. Pellets showing faceted interfacial surfaces with good interfacial contacts in a microwave foamed block.

could be easily fragmented into foamed pellets, even during release from the mould. This clearly indicated a lack of sufficient adhesion at the contacts.

It has been shown that microwave foaming of extruded wheat flour and starch pellets was accompanied by vaporization of most of the water (75%–89%) contained within them (Zhou, 2004). Such loss of moisture during expansion of the pellets would have left low water content surfaces when they come to form flattened interfaces. Despite the softening of the pellets by heat, the lack of the plasticizing water was likely the cause of insufficient molecular adhesion across the interfaces. In addition, recognising that fusion is a kinetic process, the weak bonding may also be attributable, in part, to insufficient time during microwave foaming (<60 s) for molecular diffusion across the interfaces to occur at the low water content at the interfaces.

To enhance the bonding strength at the interfaces, maintaining a relatively high water content at the contact surface is crucial not only to plasticize the material so as to achieve more intimate contact but also to enhance the mobility of the starch molecules for interfacial diffusion.

It was found that the interfacial bonding between foamed pellets could be significantly enhanced by a prewetting treatment of the surface of pellets before the microwave foaming. The pellets were soaked in a 3.0 M NaCl solution for a predetermined time immediately before microwave foaming. Three minutes soaking was found to give adequate adhesion between the interfaces. Shorter soaking time was insufficient to produce the required cohesion, while prolonged soaking time gave rise to a decreased expansion of the pellets and led to hard and dense blocks. This is expected as foamability of pellets has been shown to decrease at higher water contents (Zhou et al., 2005). Maintaining plasticized interfaces is considered the key for the successful enhancement of interfacial bonding using the soaking pre-treatment. The layer should be sufficiently thick to avoid drying out of water during the microwave heating and thin enough not to sacrifice the foamability of the interior core material.

The reason to choose the NaCl solution instead of water for soaking is that NaCl addition can enhance microwave energy absorption of water (see Table 1) and to retain moisture content as a humectant in the starch materials (Zhou et al., 2005). These are beneficial for reducing melt viscosity in the surface layer and plasticizing the interface to bond more effectively. With other experimental conditions being the same, it was found that pellets soaked in NaCl solution produce better bonded blocks than those soaked in water.

## 3.2. Uniformity of heating and foaming within a moulding

To produce a uniformly foamed structure throughout a microwave-foamed block, it is critical to establish a uniform temperature distribution so that pellets throughout the moulding reach the foaming temperature at similar time. The consequence of a non-uniform temperature distribution in a microwave-foamed block can be dramatic – while the centre may have already burnt, the pellets near the mould surface remain insufficiently foamed as shown in Fig. 3.

Assuming a uniform radiation intensity within the mould cavity and all pellets absorb the same amount of radiation energy, the temperature rise throughout the moulding should be uniform, provided there is no heat loss at the mould surface. However, this assumption is unfortunately incorrect. As the mould absorbs little microwave energy, it would remain at a relatively lower temperature than the pellets. Therefore the pellets near the mould surface lose heat to the mould and their temperature would be lower than those pellets at the centre as schematically shown in Fig. 4a. If the pellets in a mould were heated by conduction from a heated mould without microwave radiation, then the temperature profile would be expected as shown schematically in Fig. 4b. A natural solution to the non-uniform temperature profile is to combine microwave

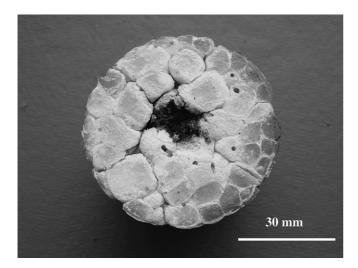


Fig. 3. A non-uniformly foamed block with burnt centre and insufficiently expanded surface.

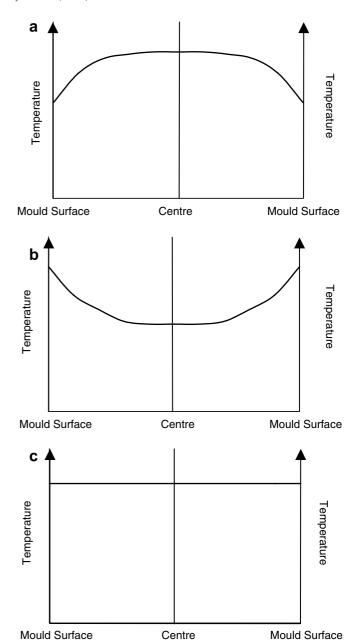


Fig. 4. Schematic diagrams showing likely temperature distribution within the pellets: (a) microwave heating only in a cold mould, (b) Conduction heating only in a heated mould, (c) Microwave heating in a preheated mould

radiation with preheating of the mould so as to create a uniform temperature profile as shown schematically in Fig. 4c.

It was found that when the mould was heated to about 160 °C (surface temperature measured using a infrared thermometer (Raytek®, ST6, Raynger, Santa Cruz, USA), and followed by microwave heating to complete the foaming, a uniformly foamed block as shown in Fig. 5 could be produced. Mould cooling during pellet loading was unavoidable and it was noted that the actual mould surface temperature dropped from 160 °C to about 120 °C while the pellets were loaded.

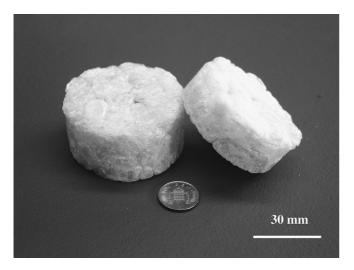


Fig. 5. Uniformly foamed starch-based blocks using microwave heating.

#### 3.3. Mould filling and fusion of foamed pellets

Since the extruded pellets were expanded in a confined space, the extent to which the mould is filled and the degree of interfacial contacts and bonding between foamed pellets depends on the initial amount of pellets loaded into the mould in addition to their expansion capability in an open space. Clearly, when the amount of loaded pellets is low, the pellets may expand freely without forming contacts with their neighbours. On the other hand, when an excessive amount of pellets are used, the expansion of pellets may be suppressed by the pressure from interfacial contacts with the neighbouring pellets, which results in insufficient expansion and a high-density block.

Therefore, there exist an optimum initial loading of pellets (dependent on the free expansion ratio of the pellets) for a given volume of a mould, which would allow the pellets:

- (1) to expand sufficiently so as to reduce the overall foam density and
- (2) to form intimate contact and sufficient bonding with the neighbouring pellets and fill the available space within the mould cavity.

To satisfy these conflicting requirements, a compromise must be reached. To obtain blocks with strong coherent structure, relatively low density and acceptable mould filling, it is necessary to experimentally determine such optimum initial loading for the different types of pellets.

Fig. 6 shows the effect of initial loading of pellets (measured by the ratio of solid pellet volume,  $V_{\rm p}$ , to the mould volume,  $V_{\rm m}$ ) on the final mould filling (measured by the ratio of the foamed block volume,  $V_{\rm b}$ , to the mould volume). When the initial loading is lower than 10% for pellets from the Superfine flour and purified wheat starch, 12% for pellets from the Temple flour with salts, and about 14% for pellets from the Temple flour alone, a coherent foam block

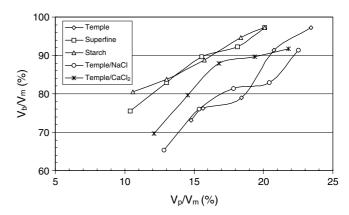


Fig. 6. Effect of initial loading of pellets  $(V_p/V_m)$  on mould filling by foamed pellets  $(V_b/V_m)$ .

could not be formed. In other words, these are the thresholds of minimum loading of pellets for achieving a coherent block.

As can be seen from Fig. 6, the extent of mould filling increases with  $V_p/V_m$ . Pellets made from the Superfine flour and purified wheat starch have better mould filling ability compared with those of the other compositions, i.e., with the same initial loading of pellets, a higher final mould filling can be achieved by using the pellets from the Superfine flour and purified wheat starch. Previous work showed that the addition of salts significantly increases the free expansion ratio of pellets and increase in heating rate (Zhou et al., 2005). Temple/NaCl and Temple/CaCl<sub>2</sub> pellets have larger free expansion ratio compared to the Superfine and pure starch pellets (Zhou et al., 2005) and were thus expected to have better mould filling properties. The results in Fig. 6 suggest that a high free expansion ratio of pellets does not always correspond to a higher mould filling. Mould filling is not only dependent on the free expansion ratio of the pellets, but also on the rheological behaviour of the pellets during foaming. Constrained within a mould and between the neighbouring pellets which are also expanding, better viscous flow of the pellets at contacts may be more important for achieving a high extent of mould filling.

Fig. 7 shows the changes of density of foamed block with the initial loading of pellets for various compositions. As expected for a fixed volume of the mould cavity, the density of the foamed block increases with the increase of the initial loading of pellets. The blocks made from Superfine flour and purified wheat starch pellets have lower density than the others. However, comparing with the density of free expanded pellets (Zhou et al., 2005), which ranged between 0.092 and 0.15 g/cm³, the density of the foamed block is much higher. The restriction to expansion of pellets from the mould wall and the neighbouring foaming pellets was clearly responsible for this inherent feature of low expansion or high density for the microwave-foamed blocks.

It was noted that the pellets near the top of the block expanded more than the pellets at bottom, but the foamed blocks could fill sharp corner of the mould better at the bottom (see Fig. 8). This may be explained by the observation

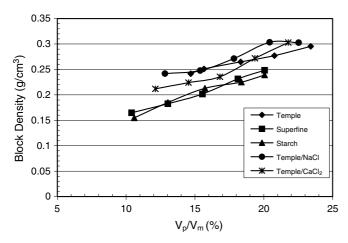
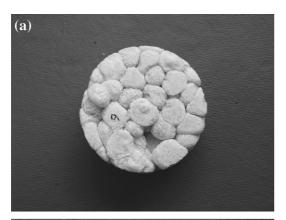


Fig. 7. Changes of foamed block density with initial loading of pellets  $(V_p/V_m)$  of various compositions.



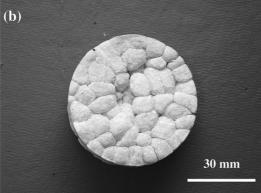


Fig. 8. The difference in appearance of the external surfaces at the top (a) and the bottom (b) for a block made from pellets of the Temple flour.

made in a glass mould that the pellets at the top started foaming first, probably due to the difference in the penetration of the microwave power. Since there was more free space in the mould in the earlier stages of foaming, the pellets near the top could expand more freely. These foamed pellets were then pushed up to occupy the available space near the top cap as the pellets beneath them continued to foam. With the reducing available space in the mould, and the contacts among the foaming pellets, the expansion of the pellets near the bottom of the mould was more restricted, and led to the

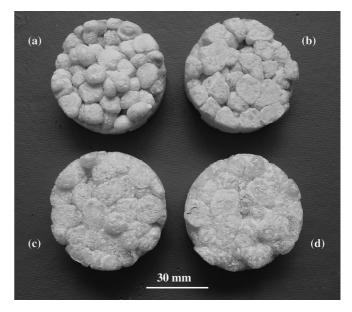


Fig. 9. Moulding surfaces showing difference in fusion for blocks made from (a) the Temple flour; (b) the Temple/CaCl $_2$  (10.5%); (c) the Superfine flour; (d) the purified wheat starch.

difference in the extent of expansion from top to bottom of a block. Furthermore, since the foamed pellets near the top were relative dry because of the moisture loss during foaming, it was more difficult for these foamed pellets to deform at the contacts and fill the mould as well as those at the bottom. This effect could be removed, if the exposure to microwave radiation is from variable angles (e. g., from both the top and the bottom of a block), which may be achieved by redesign of the microwave field.

It was noticed from the surface appearance of the foamed blocks that the fusion of the foamed pellets was different for those made from the three raw materials. On the surface of the blocks made from the Temple flour pellets, the individual foamed pellets were still clearly identifiable (Fig. 9a). The addition of salts did not significantly improve the fusion of pellets from the Temple flour during the microwave foaming (Fig. 9b). The extent of fusion for pellets from the Superfine flour was much improved (Fig. 9c) and for the blocks made from the purified wheat starch pellets, the interfaces almost disappeared (Fig. 9d). These observations seem to support the arguments made earlier in Section 3.3 on mould filing. The pellets from the Superfine and purified starch are more capable to form intimate contact by viscous flow at the contact areas to fill voids between pellets as well as the available mould cavity. This may be attributable to the fact that these raw materials contain much less impurities (e.g., bran), which increase the melt viscosity of starch (Macosko, 1994).

#### 3.4. Properties of the foamed blocks

#### 3.4.1. Cell structures

Fig. 10 shows SEM micrographs of sections cut along the vertical axis of the cylindrical foamed blocks made from

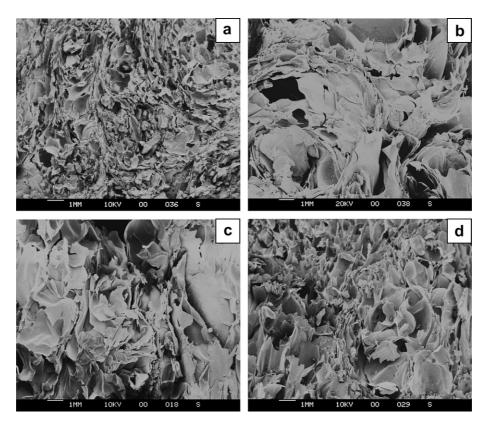


Fig. 10. SEM micrographs of the sections of microwave foamed blocks made from extruded pellets with different compositions: (a) the Temple flour (0.254 g/cm<sup>3</sup>); (b) the Superfine flour (0.215 g/cm<sup>3</sup>); (c) the purified wheat starch (0.217 g/cm<sup>3</sup>); (d) the Temple/CaCl<sub>2</sub> (0.221 g/cm<sup>3</sup>).

pellets with different compositions. Although there may exist fusion lines between foamed pellets on the external surface of foamed blocks, most of pellets have been fully fused together in the interior of the blocks. Some interfaces between foamed pellets may be identifiable (see Figs. 10a and d), but it is difficult to identify any voids between pellets. No obvious difference in cell structure was observed through the height of blocks, even when the expansion of pellets differs notably at the bottom and top of blocks as described in Section 3.3.

Comparing Fig. 10 with the SEM micrographs of cross section of freely expanded pellets (Zhou et al., 2005), it was found that the cell structures in the foamed block were not only smaller in average cell size, the cell shape was much more elongated compared with the more spherical cells in the freely expanded foams. The distortion of the cells in foamed blocks clearly suggests that the expansion of pellets was restricted from the contact interfaces or the mould wall during foaming. Since such restriction to expansion is dependent on the free space around the periphery of pellets and the area of contacts with the neighbouring pellets, the bubbles in the pellets have an elongated shape with the longer axis parallel to the contact interfaces.

## 3.4.2. Mechanical properties of the foamed blocks

Fig. 11 shows the typical compressive stress-strain curves of the microwave-foamed blocks measured at 20 °C and 50% relative humidity. The initial low gradient of the

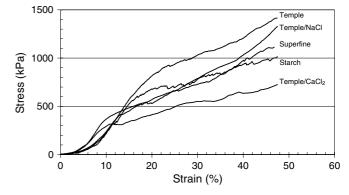


Fig. 11. Typical compressive stress-strain curves for the foamed blocks made from pellets with different compositions.

stress-strain curve was attributable to the compression of the surface irregularities of the block samples. This was followed a linear stress-strain relationship reflecting the elastic deformation once good contacts between the compression platens and the foam block sample were established. At higher stress, the foam enters a region of yielding where cell structures start to buckle or collapse. This continues progressively throughout the foam and hardening can be observed as the cell structure densifies until the tests stopped at about 50% strain. The foams display typical behaviour for compression of flexible foams (Gao & Song, 2005; Gibson & Ashby, 1997). Unlike the

Table 2
Mechanical properties of microwave foamed blocks from extruded pellets with various compositions

Composition	Density (g/cm <sup>3</sup> )	Compressive modulus (kPa)	Stress at 10% strain (kPa)	Deformation energy at 40% strain (J/cm <sup>3</sup> )
Temple flour	0.254 (0.079)*	$61 \pm 7 \ (55 \pm 22)$	$750 \pm 80 \ (314 \pm 47)$	$380 \pm 20 \ (129 \pm 22)$
Superfine flour	0.215 (0.063)	$65 \pm 12 \ (24 \pm 6)$	$570 \pm 70 \ (180 \pm 36)$	$290 \pm 20 \ (69 \pm 10)$
Starch	0.217 (0.063)	$57 \pm 13 \ (22 \pm 4)$	$480 \pm 130 \ (161 \pm 27)$	$260 \pm 30 \ (68 \pm 6)$
Temple (+5.5% w/w NaCl)	0.226 (0.069)	$61 \pm 8 \ (36 \pm 16)$	$600 \pm 120 \ (250 \pm 97)$	$290 \pm 30 \ (88 \pm 25)$
Temple (+10.5% w/w CaCl <sub>2</sub> )	0.221(0.060)	$48 \pm 14 \ (31 \pm 10)$	$350 \pm 70 \ (174 \pm 42)$	$180 \pm 20 \ (63 \pm 11)$
Extruded foam	0.013	$2.5 \pm 0.2$	$15 \pm 1.2$	$5.4 \pm 0.4$

<sup>\*</sup> Data in brackets are those from freely expanded pellets (Zhou et al., 2005).

compressive stress-strain curves for the freely foamed pellets where yield can be clearly identified as a stress peak (Zhou et al., 2005), yield of the foamed blocks is more progressive. This is understandable, as the cell buckling of foamed pellets within a block would take place at different time due to the random orientation of elongated cells. To compare the compression behaviour of these blocks with that of freely foamed pellets and extruded starch (Greenfill, Green Light Products Ltd., UK) foams, three parameters have been selected: compressive modulus of elasticity, the compressive stress at 10% strain and deformation energy at 40% strain, which are listed in Table 2. The higher values of compressive modulus and deformation energy, compared with that of the freely expanded pellets and the extruded foams, indicates that these blocks are much more rigid and resistant to compressive deformation, which makes them potentially more suitable for cushion packaging at high stress levels or as a more rigid core for composites such as sandwich panels.

It is noticeable in Fig. 11 that the slat-containing foams are apparently more compressible. The addition of salts has been show to enhance microwave heating rate (Zhou et al., 2005) and degree of foaming, which result in lower density and more compressible foams (Table 2). In addition, salts also increase absorbed moisture in the foams (Zhou et al., 2005) and contribute to plasticization. Therefore salt addition can be used as a method to control the compressibility of the MAM foams.

### 4. Conclusion

Foam blocks using extruded pellets as feedstock can be produced by the microwave-assisted moulding (MAM) method. Selection of mould materials, combining microwave heating with preheating of mould and pre-treatment of pellets are the key requirements for fabricating a uniformly foamed block with sufficient bonding and fusion among the foamed pellets. The mould filling and block density are influenced by the initial loading of pellets in the mould cavity and the expansion ratio of the pellets, but the former is the more dominant factor. Mould filling and interface fusion by pellets made from the purified wheat starch or the Superfine wheat flour are better than those by the Temple with or without addition of salts. Due to the constraint to pellet expansion from the neighbouring pellets

and the mould wall, the pellet formulations have less effect on the cell structure and hence the mechanical properties of the formed blocks compared with the freely expanded pellets. The density of the MAM blocks are higher compared with starch foams by free microwave expansion and extrusion foaming and thus are more rigid and resistant to compression. Yet they are able to absorb energy at high stress levels and hence potentially applicable for moulded cushion packaging for heavy goods or foam core for more rigid lightweight composite panels.

#### Acknowledgements

The financial support from EPSRC (Grant No. GR/N00272/01) is acknowledged. The authors wish to thank Dr. Karnik Tarverdi of Wolfson Centre for Materials Processing at Brunel University for his help during the extrusion operations and Heygates Limited, Northampton, UK for supply of flours and starches. Dr Roger Parker acknowledges receipt of the Core Strategic Grant from the BBSRC (biotechnology and Biological Sciences Research Council).

#### References

Bastioli, C., Bellotti, V., Del Giudice, L., Lombi, R., & Rallis, A. (1994).
Expanded articles of biodegradable plastic materials. US Patent 5 360 830.

Bastioli, C., Bellotti, V., Del Tredici, G., Montino, A., & Ponti, R. (1998a). Biodegradable foamed plastic materials. US Patent 5 736 586.

Bastioli, C., Bellotti, V., Del Tredici, G., & Rallis, A. (1998b). Biodegradable foamed articles and process for preparation thereof. US Patent 5 801 207

Bellotti, V., Bastioli, C., Rallis, A. & Del Tredici, G. (1995). Expanded articles of biodegradable plastic material and a process for the preparation thereof. Europe Patent, EP0667369.

Bellotti, V., Bastioli, C., Rallis, A. & Del Tredici, G. (2000). Expanded articles of biodegradable plastic material and a process for the preparation thereof. Europe Patent, EP0989158.

Bhatnagar, S., & Hanna, M. A. (1995a). Properties of extruded starch-based plastic foam. *Industrial Crops and Products*, 4, 71–77.

Bhatnagar, S., & Hanna, M. A. (1995b). Physical, mechanical, and thermal properties of starch-based plastic foams. *Transactions of the ASAE, 38*, 567–571.

Boischot, C., Moraru, C. I., & Kokini, J. L. (2003). Factors that influence the microwave expansion of glassy amylopectin extrudates. *Cereal Chemistry*, 80, 56–61.

CES (2005). Cambridge Engineering Selector, Edupack 2005, Granta Design Limited, UK.

- BPF (2005). http://www.bpf.co.uk/bpfindustry/process\_plastics\_mould-ing\_expanded\_polystyrene.cfm.
- Cha, J. Y., Chung, D. S., Seib, P. A., Flores, R. A., & Hanna, M. A. (2001). Physical properties of starch-based foams as affected by extrusion temperature and moisture content. *Industrial Crops and Products*, 14, 23–30.
- DETR, (2000). Waste Strategy: A Waste Strategy for England and Wales.

  Department for the Environment, Transport and the Regions, HMSO,
  London
- DEFRA, (2004). A Strategy for non-food crops and uses-creating value from renewable materials (PB10188), October, 2004, Department of food, environment and rural affairs, DEFRA Publications, Admail 6000, London (www.defra.gov.uk).
- Evans, I. D., & Haisman, D. R. (1982). The effect of solutes on the gelatinisation temperature range of potato starch. *Starch/Stäke*, 34, 224–231.
- Fang, Q., & Hanna, M. A. (2001a). Characteristics of biodegradable Mater-Bi<sup>®</sup>-starch based foams as affected by ingredient formulations. *Industrial Crops and Products*, 13, 219–227.
- Fang, Q., & Hanna, M. A. (2001b). Preparation and characterisation of biodegradable copolyester-starch based foams. *Bioresource Technol*ogy, 78, 115–122.
- Gao, Y. X. & Song J. H. (2005). Rheological behaviour and cellular structure of a biodegradable foam, *Proceedings of the 4th Pacific rim conference on rheology*, August 7–11, Shanghai, China.
- Gibson, L. J., & Ashby, M. F. (1997). Cellular Solids (2nd ed.). Cambridge, MA: Cambridge University Press.
- Kinloch, A. J. (1986). Adhesion and adhesives: science and technology. London: Chapman and Hall.
- Lacourse, N. L. & Altieri, P. A. (1989). Biodegradable packaging material and the method of preparation thereof. US Patent, 4,863,655.
- Lacourse, N. L. & Altieri, P. A. (1991). Biodegradable shaped products and the method of preparation thereof. US Patent, 5,043,196.
- Landrock, A. H. (1995). *Handbook of plastic foams: types, properties, manufacture, and applications*. New Jersey: Noyes Publications.

- Linstead, C., & Ekins, P. (2001). Mass balance UK, mapping UK resource and material flows. *Royal Society for Natural Conservation*, 12.
- Lye, S. W., Lee, S. G., & Chew, B. H. (1998). Characterisation of biodegradable materials for protective packaging. *Plastics Rubber and Com*posites Processing and Applications, 27, 336–383.
- Macosko, C. W. (1994). Rheology principles measurements and applications. Wiley–VCH p. 456.
- Metaxas, A. C., & Meredith, R. J. (1983). *Industrial microwave heating*. London: Peter Peregrinus.
- PaperFoam (2005). http://www.trhubnet.com/packaging/news.nsf. Potatopak (2005). http://www.potatopak.org/home.html.
- Shogren, R. L., Lawton, J. W., & Tiefenbacher, K. F. (2002). Baked starch foams, starch modifications and additives improve process parameters, structure and properties. *Industrial Crops and Products*, 16, 69–79.
- Song J. H. (2005). Lightweight sandwich panels based on starch foam. In proceedings of CIMNFC LINK seminar, Warwick University, 19th April, Warwick.
- Tatarka, P. D., & Cunningham, R. L. (1998). Properties of protective loose-fill foams. *Journal of Applied Polymer Science*, 67, 1157–1176.
- Wang, B., Song, J. H., & Kang, Y. G. (2001). Modelling of the mechanical behaviour of biodegradable foams-from physical fundamentals to applications. In L. C. Zhang (Ed.), *Engineering plasticity and impact* dynamics (pp. 117–134). Singapore: World Scientific Chapter 7.
- Wang, B., Song, J. H., & Kang, Y. (2002). On modelling of biodegradable foams for packaging applications. Key Engineering Materials, 227, 253–260
- Willett, J. L., & Shogren, R. L. (2002). Processing and properties of extruded starch/polymer foams. *Polymer*, 43, 5935–5947.
- Zhou, J. (2004). Ph. D. theses: Microwave assisted moulding of starch-based foams, Brunel University, UK.
- Zhou, J., Song, J., & Parker, R. (2005). Structure and properties of starch-based foams prepared by microwave heating from extruded pellets. *Carbohydrate Polymers*, 63, 466–475.