Effects of the Properties of Blowing Agents on the Processing and Performance of Extruded Starch Acetate

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ABSTRACT: The extrusion of polysaccharide-based polymers, such as starch acetate, is quite different from that of ordinary synthetic polymers. To understand how the physiochemical properties of blowing agents affect plasticization and expansion processes, starch acetate was extruded with water, ethanol, and ethyl acetate. The studied properties and factors were the evaporation rate, surface tension, boiling point, solubility index, latent heat of vaporization of blowing agents, extrusion temperature, and nucleating- and blowing-agent concentrations. The properties of the blowing agents and operating conditions affected the solubility of the matrix polymer, the nucleation process, and cell growth, which affected the foam density and specific volume. A high temperature increased the cell density and specific volume when water and ethanol were used because a high temperature increased the solubility of starch acetate in water and ethanol and promoted nucleation. Ethyl acetate already had high solvency to starch acetate and a high evaporation rate. A high temperature reduced the melting strength, thereby reducing the cell density and specific volume. Water evaporation was greater, despite a high latent heat of evaporation (h_r) and boiling point, than the average volumes of ethanol and ethyl acetate that evaporated. The blowing-agent efficiency was a function of the solvency, blowing-agent evaporation rate, and operating conditions. © 2005 Wiley Periodicals, Inc. J Appl Polym Sci 97: 1880–1890, 2005

Key words: biopolymers; blowing agents; foam extrusion

INTRODUCTION

Starch acetate is a biodegradable polymer made from natural renewable resources. Its preparation has been studied for many years, but its application as a biomaterial has not made remarkable progress until recently. In the early stage of study, efforts were concentrated on low-substitution-degree starch acetates for food industry applications, whereas high-substitution-degree polysaccharides were dominated by cellulose acetates. With the advent of high amylose starch, more efforts were concentrated on high-substitution-degree starch acetate to produce biodegradable foams as substitutes for polystyrene.[1–5] However, starch acetate foam is inferior to polystyrene foam. One reason is the difference in the intrinsic properties of the two polymers, and another is the difference in the processing requirements. Further improvement of the intrinsic properties of starch acetate is limited by the cost and raw material. On the other hand, the investigation of the starch acetate foaming process is still in its early stages. There is great potential for further improvement. Shogren[6] studied the extrusion of starch acetate with water and proved that it was an effective plasticizer and a suitable blowing agent for starch acetate. Miladinov and Hanna[7,8] studied the extrusion of starch acetate with ethanol. Guan and Hanna[9] found that steaming re-expanded starch acetate fiber based foams. However, no comprehensive reports are available on the relationship between the physicochemical properties of the blowing agent, the operating conditions, and the expansion of starch acetate.

The extrusion foaming of polysaccharide-based polymers is quite different from that of ordinary synthetic polymers. Solvency or plasticizing effects of blowing agents or other additives are important to the extrusion process. Therefore, the blowing agent must play a double role of plasticization and expansion in extrusion foaming. Understanding how the physicochemical properties of blowing agents affect both plasticization and expansion is crucial to the extrusion of starch acetate foam.

The objective of this research was to study the effects of the physical chemistry properties of blowing agents such as the boiling point, evaporation rate, surface tension, and solubility index, together with operating conditions such as the temperature and nucleating- and blowing-agent concentrations, on two

primary functionalities of foam, the cell density and specific volume.

EXPERIMENTAL

Starch acetate

High amylose starch was dried at 50°C for 48 h. To begin the acetylation process, 110 kg of acetic anhydride was placed in a steam-jacketed reactor with a rotating, self-wiping paddle. Then, 45.5 kg of starch was added to the reactor with 5 min of continuous mixing. Finally, 5 kg of a 50% NaOH solution was added with mixing. The temperature of the reactor jacket was maintained at 123°C. After 3 h, the reaction was stopped by the quick addition of 200 L of cold water to the reactor. The pH value was adjusted to 5.0 via washing with tap water before drying at 50°C to a moisture content of 4% wet basis (w.b.). The dry starch was ground in a standard model 3 Wiley mill (Arthur H. Thomas Co., Philadelphia, PA) to pass through a 5-mm-opening sieve.

Degree of substitution (DS)

DS indicates the average number of substitutions per anhydroglucose unit.[7] There are three free hydroxyl groups available for modification. Therefore, the highest possible DS is 3. DS of esterified starch was determined by the hydrolysis of substituted groups with 1 N NaOH and then titration with 0.5 N HCl to the original pH before the addition of NaOH.[7] A 5-g sample was placed in a 500-mL conical flask, and 50 mL of distilled water was added. The mixture was conditioned at 30°C for 1 h in a Tecator (Hoganas, Sweden) 1024 shaking water bath after the measurement of the pH. To each flask, 10 mL of 1 N NaOH was added. Each sample was conditioned for 48 h at 50°C to hydrolyze the fatty acid substitutes. Then, the sample was titrated with 0.5 N HCl to the original pH. DS was calculated as follows:

$$\text{DS} = \frac{M_{\text{FA}} \times \text{MW}_{\text{AN}}}{W - M_{\text{FA}} \times (\text{MW}_{\text{FA}} - \text{MW}_{\text{H}_2\text{O}})}$$

where W is the weight of the sample (g), M_{FA} is the moles of titrated fatty acid, MW_{FA} is the molecular weight of the fatty acid, $\text{MW}_{\text{H}_2\text{O}}$ is the molecular weight of water (18), and MW_{AN} is the molecular weight of an anhydroglucose unit (162). DS of the sample used in this research was 2.4.

Extrusion sample preparation

The acetylated starch was dried in a precision mechanical convection oven (GCA Corp., Chicago, IL) at 105°C for 8 h and then cooled in a desiccator for 4 h to ensure that it was moisture-free before being used in sample preparation. Talc was added to all samples at a 2% level (dry starch base). Different amounts of blowing agents were added to the starch and mixed in a model C-100 mixer (Hobart Corp., Troy, OH) for 5 min. The samples were then allowed to equilibrate for 24 h in air-tight jars. The ranges of the blowing agent and temperature for the extrusion of DS 2.4 starch acetate were chosen according to preliminary tests.

Extrusion

Starch acetate samples were extruded with twin corotating screws with a length-to-diameter ratio of 12:1 at 120 rpm. The temperature profile was 50°C–150°C–150°C–150°C for samples extruded at 150°C, 50°C–160°C–160°C–160°C for samples extruded at 160°C, and 50°C–170°C–170°C–170°C for samples extruded at 170°C. The nozzle diameter was 4 mm, and the length was 23.6 mm. To control shrinkage of the foam, upon cooling, specially designed postextrusion equipment was employed.

Foam cell density

Foamed samples for scanning electron microscopy were sectioned with a razor blade and then mounted on aluminum stubs with graphite-filled tape, sputter-coated with gold/palladium, and examined with a Hitachi scanning electron microscope (Tokyo, Japan). The foam density was reported as the number of cells per square centimeter of projected cross-sectional area.

Specific volume

The specific volume of each sample was determined with a modified glass bead displacement method originally developed for determining the volume of cookies.[1] Glass beads 0.1 mm in diameter were used as the displacement medium. The specific volume was reported as the ratio of the volume to the unit weight (mL/g).

RESULTS AND DISCUSSION

Cell density

The cell size and uniformity contribute significantly to the mechanical properties of foams of a given specific volume and matrix polymer. The cell densities of foams extruded at various water contents and temperatures, in the absence of talc, are shown in Figure 1. The cell density increased with temperature and decreased with water. The low cell density of the foam extruded at 150°C resulted from low compatibilities of water with starch acetate. Starch acetate (DS 2.4) had
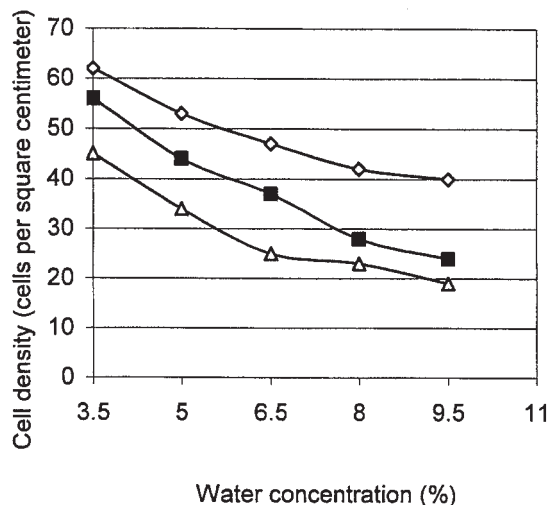
Figure 1 Effects of the water concentration and temperature on the cell density in the absence of talc.

about 20% of the hydroxyl groups left. In the extrusion of native starch, the highest specific volume was achieved with 14–15% water. Water concentrations lower than 14% were impractical for conventional extrusion because of the high viscosity and mechanical degradation of starch. According to this, the amount of water needed for starch acetate should be 2.8–3% (w/w) on the basis of the hydroxyl groups available. However, starch acetate could not be extruded with so little water, especially at a low temperature, because water could only plasticize some of the starch acetate molecules with hydroxyl groups. Therefore, water in excess of the requirement of the hydroxyl groups of starch acetate was needed for extrusion. On the other hand, only water within the affinity region of starch acetate could be mixed with starch acetate easily and evenly. Water above that level worked as a lubricant and was difficult to mix evenly with starch acetate. It was not easy to determine how much water could be bound closely to starch acetate because of the affinity between water and starch acetate. Experimental results showed that foams extruded at 150°C foamed with a lot of big cells (sometimes called big holes) together with small cells at a 3.5% water concentration in the presence of talc. The number of big cells increased with the water content. At higher temperatures, the affinity of water to starch acetate increased with temperature, so that big cells did not appear until the water content reached 5% at 160°C in the presence of talc. At 170°C, big cells appeared when the water content exceeded 6.5% in the presence of talc. For extrusion at 150°C with high water concentrations, only big cells existed even in the presence of talc. Water above the affinity level of the hydroxyl groups of starch acetate was less bound and more readily nucleated and migrated from inner cells to outer cells.

These cells grew faster to form holes inside the extrudate. The nucleation and growth of smaller cells may have been completely suppressed at higher water concentrations, and this resulted in only big cells being formed in the final products.

Figure 2 shows the cell densities of foams extruded with ethanol in the absence of talc and at preselected ethanol concentrations and temperatures. The cell densities of the foams extruded with ethanol increased with the temperature and ethanol concentration in the low ethanol concentration region and then decreased. The cell densities of the foams extruded with ethanol were higher than those of the foams extruded with water, especially at low temperatures. No big holes were observed in the foams extruded with ethanol, even at low temperatures and high ethanol concentrations. Figure 3 shows the cell densities of the foams extruded with ethyl acetate at various ethyl acetate concentrations and temperatures. The cell densities of the foams extruded with ethyl acetate were the highest among all the blowing agents. However, the cell density of the foam extruded with ethyl acetate dropped with the temperature instead of increasing with the temperature as in the cases of water and ethanol.

According to classical nucleation theory,[10] the nucleation rate of blowing agents (N_{nucl}) is

$$N_{nucl} = f_0 C_0 \exp(-\Delta G_{nucl}/kT) \qquad (1)$$

where ΔG_{nucl} is equal to $16\pi\gamma^3_{bp}/3\Delta P^2$, γ is the surface tension of the foam, ΔP is the pressure drop at the die, k is the constant, T is the absolute temperature of the extrudate, and C_0 is the concentration of the blowing agent.

The surface tension and other physicochemical properties of preselected blowing agents are shown in
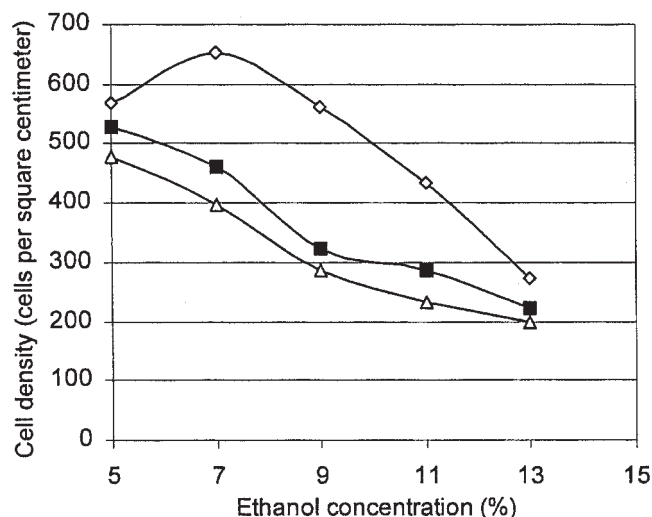

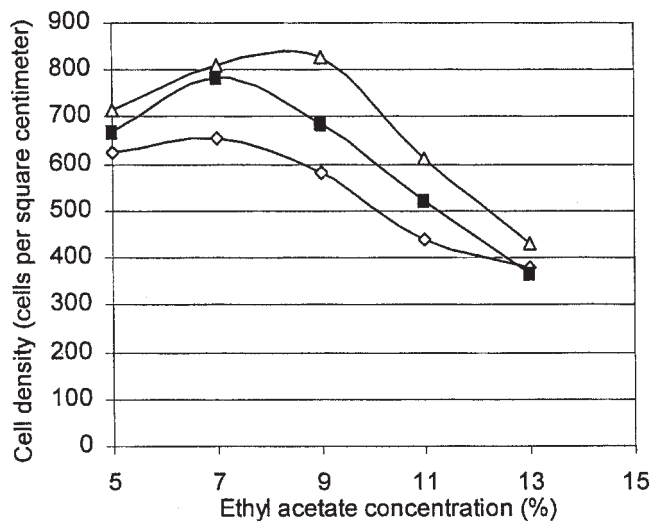**Figure 2** Effects of the ethanol concentration and temperature on the cell density in the absence of talc.

Figure 3 Effects of the ethyl acetate concentration and temperature on the cell density in the absence of talc.

Table I, where dd, dp, dh, and dt is the dispersion component, polarity component, hydrogen bonding component, and total Hildebrand parameter, respectively; D is the distance between solvent and center of solubility sphere, R is the radius of interaction, and Tb is the boiling temperature. According to eq. (1), the nucleation rates of ethanol, ethyl acetate, and water are in the order of ethanol > ethyl acetate > water. However, the final cell densities were in the order of ethyl acetate > ethanol > water. One reason may be that the surface tensions of the blowing agents at high temperatures were different from those at room temperature. The other may be that the cell density depended not only on how many nuclei were formed but also on how many cells broke or merged and how much they grew after nucleation.

Cell breakage depends on the melting strength and radius of a cell. Cell merging is a function of the evenness of the cell size and melting strength, whereas cell growth depends on the amount of gas evaporated. It is very clear that melting strength plays an important role in the whole cell growth process. A simple mathematical model was proposed to describe the effects of the temperature (T), pressure (P), and blow-ing-agent concentration (C) on the extensional viscosity (η_e; also called the melting strength) of a polybutylene succinate/CO_2 system:[11]

$$\eta_e = m_e \varepsilon^{t_e - 1} \qquad (2)$$

where

$$m_e = A_e \exp[\alpha_e/(T - T_{re}) + \beta_e P + \Psi_e C] \qquad (3)$$

where A_e, T_{re}, t_e, α_e, β_e, and ψ_e are material constants.

The relationship between the extensional viscosity, temperature, blowing-agent concentration, and pressure of a starch acetate/blowing-agent system may follow the same trend. After the die is exited, the effect of pressure should be negligible. Then, the concentration of the blowing agent and the temperature are the two main factors in extensional viscosity. The extensional viscosity decreased exponentially with the concentration of the blowing agents. The role of the temperature in extensional viscosity is a little complicated. As water and ethanol are not good solvents for starch acetate, a high temperature increased the solvency of the blowing agents to starch acetate and compensated for part or all of the negative effect of the temperature on the extensional viscosity. This can be seen clearly in Figures 1 and 2. The cell densities of the foams extruded with water and ethanol increased with the temperature. However, part of this increase resulted from the role of the temperature in the nucleation rate. Ethyl acetate already had high solvency to starch acetate, as a high temperature led to a drop in the extensional viscosity. A high nucleation rate, resulting from a high temperature, could not compensate for the drop in the extensional viscosity, so the cell density of the foam extruded with ethyl acetate dropped with the temperature.

The evaporation rate of the blowing agents correlated with the cell density better than the surface tension. Nucleation is a process in which a liquid blowing agent evaporates into gas. The evaporation rate should have a more significant effect on the nucleation rate than the surface tension. Ethyl acetate had both a higher evaporation rate and better sol-

TABLE I
Physical Properties of the Blowing Agents

Blowing agent	δ_{d_3} (J/cm$^{1/2}$)	δ_{p_3} (J/cm$^{1/2}$)	δ_{h_3} (J/cm$^{1/2}$)	δ_{t_3} (J/cm$^{1/2}$)	D	Latent Heat (J/g) at 20°C	Surface tension (Dyne/cm) at 20°C	Tb (1 atm)	Evaporation rate of butyl acetate (100)
Starch acetate	18.6	12.7	11.0	25.1	7.6 (R)	N/A	N/A	N/A	N/A
Water	15.5	16.0	42.4	47.9	32.2	2260	72.75	100	5
Ethanol	15.8	8.8	19.4	26.6	12.4	838	22.75	78.3	230
Ethyl acetate	15.8	5.3	7.2	18.2	10.0	366	23.75	77	675

N/A = denotes not applicable.

Figure 4 Effects of the water concentration and temperature on the cell density in the presence of talc.
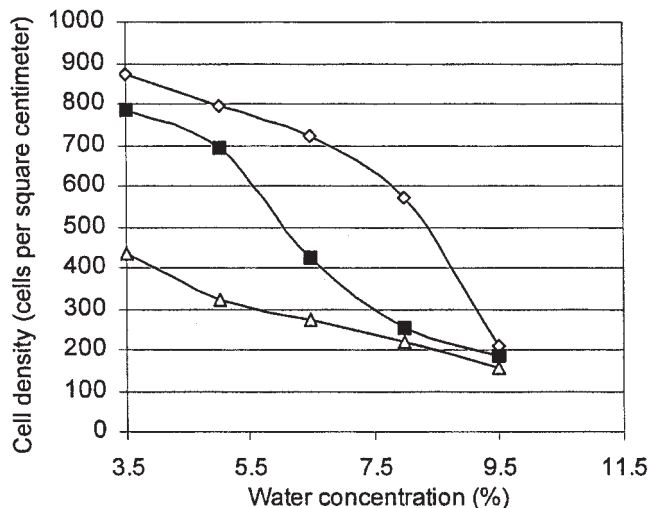

Figure 5 Effects of the ethanol concentration and temperature on the cell density in the presence of talc.

vency than ethanol and water and hence produced foam with a higher cell density than ethanol and water.

Cell merging or coalescence is another important factor in the final cell density. Cell merging results mainly from the unevenness of cells or nuclei. The unevenness of cells may be caused by several factors. The first is the uneven mixing of the blowing agent and matrix polymer. This one should be common to blowing-agent/matrix-polymer systems of low compatibility. The second is a low nucleation rate. This one is more common in extrusion. The cells that form earlier grow larger than the cell that forms later. The third is cell breakage. Therefore, all factors that influence the nucleation and solvency of the blowing agent to the matrix polymer affect cell merging.

To understand how nucleation and solvency contribute to the final cell density, talc was added to the matrix polymer. The results are shown in Figures 4 through 6. Talc acted as an inorganic nucleating agent. It reduced the viscosity of the polymer paste and die pressure significantly. The mechanism of talc in increasing the cell density was increasing the nucleation rate and hence the evenness of the cells. The result was less cell merging. As talc is a very effective nucleating agent, the difference in the nucleation rates of the blowing agents should have been negated by talc. The remaining difference in the cell density should have come from solvency. This agrees with the experimental results.

The concentration of the blowing agent is an important factor in cell density. The nuclei density increased with the blowing-agent concentration according to eq. (1).[10,12] However, the final cell density generally decreased with the blowing-agent concentration. There are three possible explanations for this phenomenon:
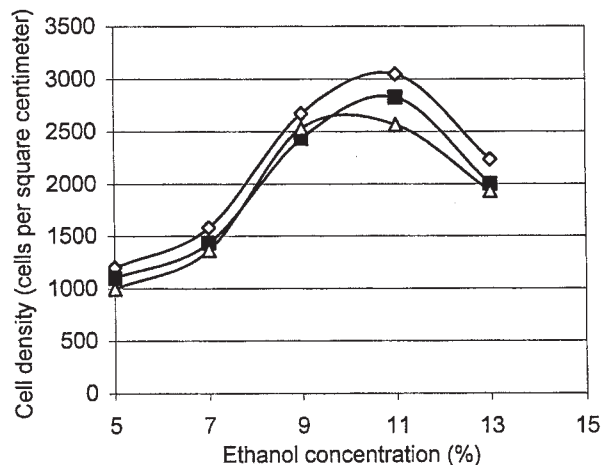
(1) cells grew more when there was more blowing agent, (2) cells merged and broke because of low melting strength, and (3) explanations 1 and 2 both occurred. The first explanation could not be very prevalent because the specific volume generally decreased with the blowing-agent concentration. Explanation 2 is reasonable, but explanation 3 is more complete. At high blowing-agent concentrations, the matrix polymer was not strong enough to separate individual cells or nuclei. Cell merging and cell overgrowth contributed to the low cell density of final products. That was consistent with the results of Park and coworkers.[13,14]

Specific volume

Figures 7–9 show the specific volumes of foams extruded with water, ethanol, and ethyl acetate in the
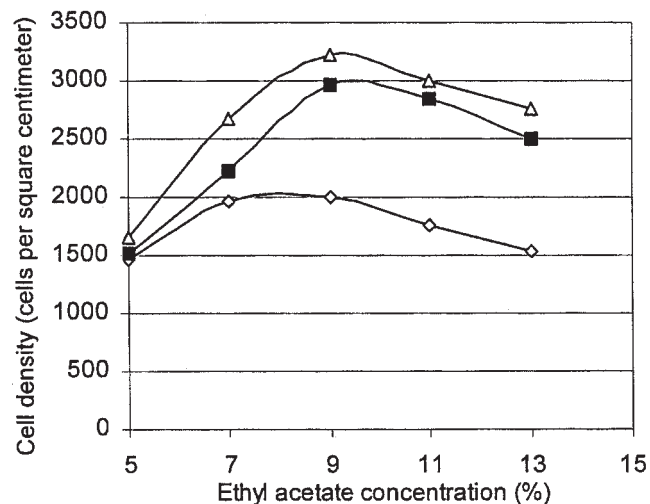

Figure 6 Effects of the ethyl acetate concentration and temperature on the cell density in the presence of talc.
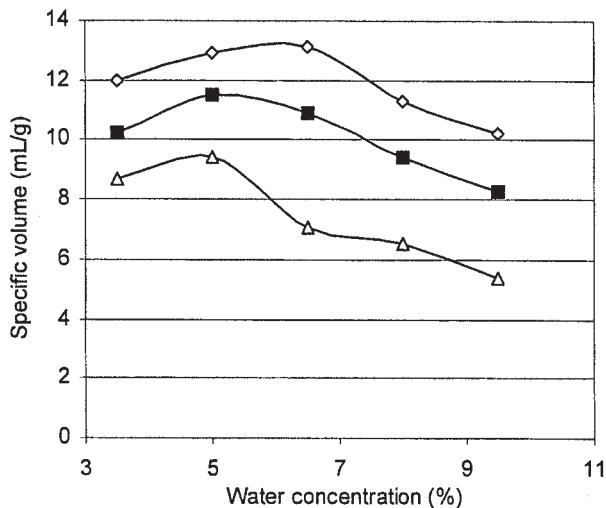
Figure 7 Effects of the water concentration and temperature on the specific volume in the absence of talc.
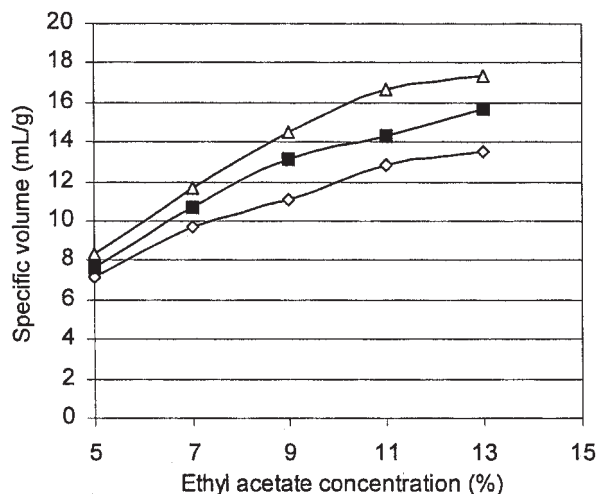

Figure 9 Effects of the ethyl acetate concentration and temperature on the specific volume in the absence of talc.

absence of talc, respectively. The specific volume increased with the blowing-agent concentrations and then decreased. Ethyl acetate and ethanol produced foams with higher specific volumes than water at low temperatures. The temperature played a positive role in the specific volume of foam extruded with water and ethanol but a negative role in those extruded with ethyl acetate. Figures 10–12 show the specific volumes of foams extruded with water, ethanol, and ethyl acetate in the presence of talc, respectively. The specific volumes of all foams extruded with talc were higher than those of their counterparts without talc. Just as in the case of cell density, talc increased the specific volume of foams extruded with water to a higher degree than that of foams extruded with ethanol and to even a higher degree than that of foams extruded with ethyl acetate. The role of temperature in the

specific volume of foams extruded with talc was similar to that of foams extruded without talc. However, its effect on the specific volumes of foams extruded with selected blowing agents in the presence of talc was less significant than that without talc.

If there is no leakage of gas out of an extrudate, the ideal specific volume should be the specific volume of the polymer plus the volume of gas evaporated from a unit of weight of the extrudate. However, that is never the case. On the one hand, the matrix polymer expands only when the inner pressure is greater than the resistant force. The resistant force can be so high that no significant expansion occurs, even if a significant amount of the blowing agent can be evaporated. This is discussed in greater detail later. On the other hand, gas can leak both from diffusion at the surface of the
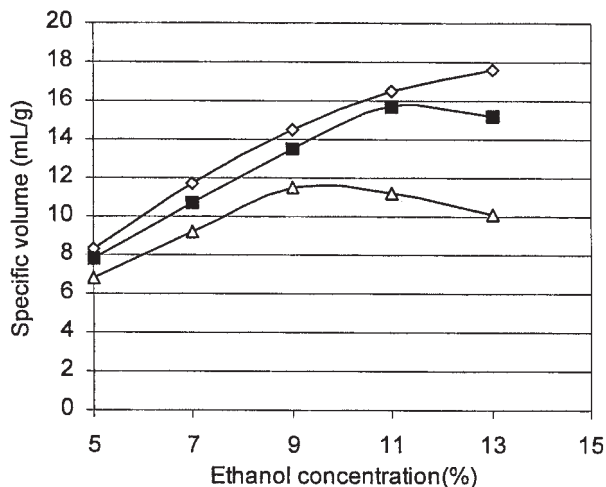

Figure 8 Effects of the ethanol concentration and temperature on the specific volume in the absence of talc.
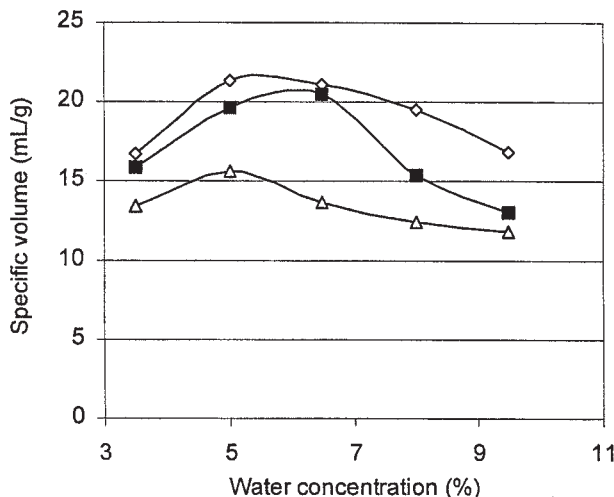

Figure 10 Effects of the water concentration and temperature on the specific volume in the presence of talc.
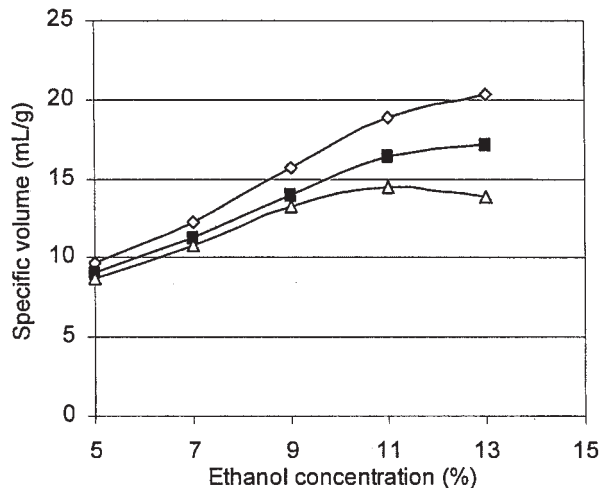
Figure 11 Effects of the ethanol concentration and temperature on the specific volume in the presence of talc.
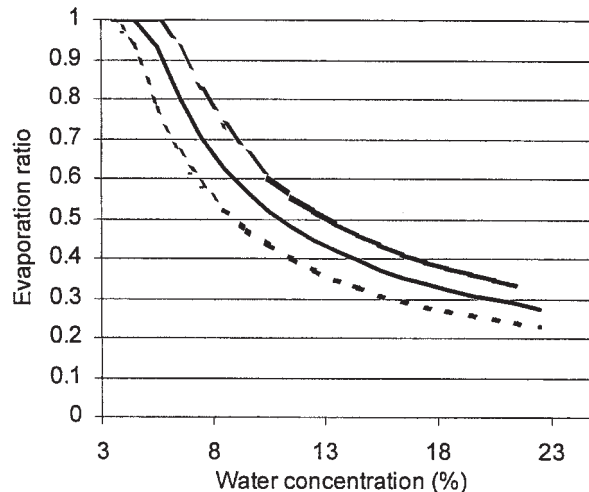

Figure 13 Effects of the water concentration and temperature on the evaporation ratio.

extrudate and from the breakage of cells on or close to the surface. Surface evaporation competes with foaming expansion inside the extrudate. Almost all blowing agents leak from the surface eventually if there is no foaming inside. Surface leakage drops to a significantly low level if an extrudate foams quickly and effectively.

The amount of the blowing agent that can be evaporated depends on the degree of overheating and latent heat of evaporation of the blowing agent. If the amount of heat lost to the environment and the change in the heat capacity of the blowing agent and polymer during expansion can be neglected and only the gas flashed off the matrix polymer, by overheating, contributes to the expansion of the foam, then the amount of the blowing agent that can be evaporated (B_g) can be calculated roughly as follows:

$$BA_g = [C_bC + 1.8(1 - C)](T_{die} - T)/H \qquad (4)$$

where C_b is the heat capacity of the blowing agent (J/g); C is the weight ratio of the blowing agent; T_{die} and T are the temperature of the die and the boiling temperature of blowing agent, respectively; and H is the latent heat of evaporation of the blowing agent (J/g). The heat capacities of starch acetate, water, ethanol, and ethyl acetate are 1.8, 4.18, 2.44, and 1.94 J g^{-1} K^{-1}, respectively.

The amounts of the blowing agents that can be evaporated are drawn on the basis of eq. (4) in Figures 13–15.

Water had a very high latent heat of vaporization, and hence the amount of water that could be evaporated was affected less by the die temperature and water concentration. The evaporation ratios of ethanol
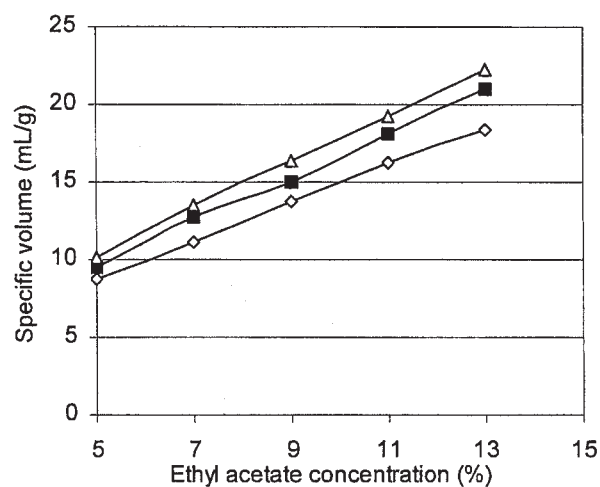

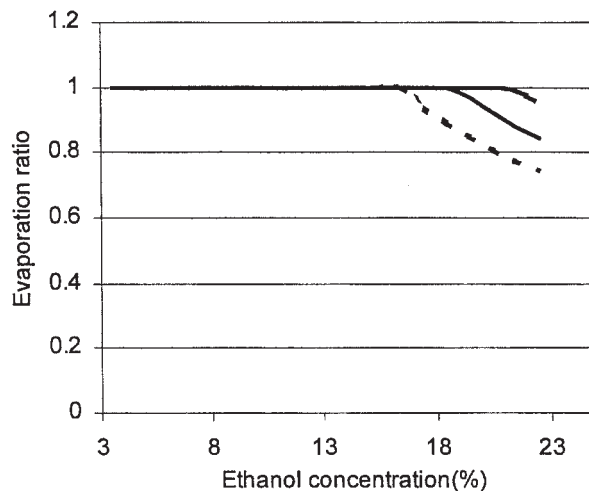**Figure 12** Effects of the ethyl acetate concentration and temperature on the specific volume in the presence of talc.

Figure 14 Effects of the ethanol concentration and temperature on the evaporation ratio.
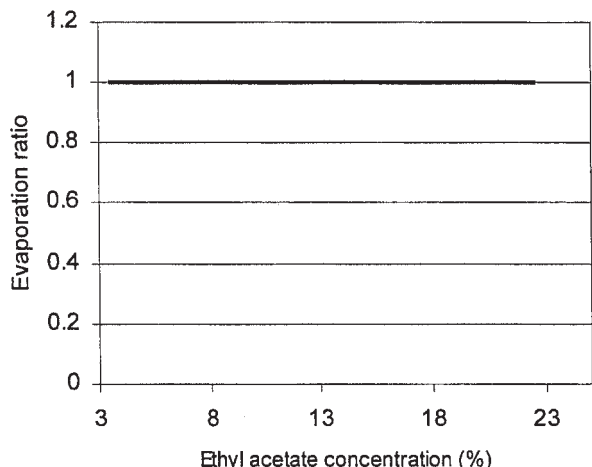
Figure 15 Effects of the ethyl acetate concentration and temperature on the evaporation ratio.
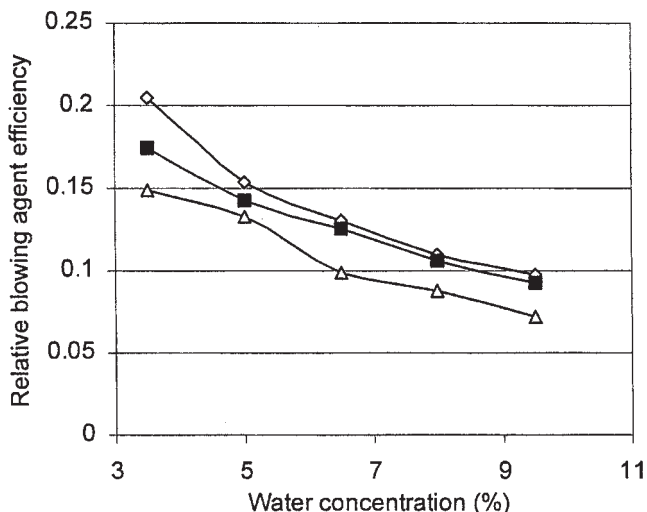

Figure 16 Effects of the water concentration and temperature on the relative blowing-agent efficiency in the absence of talc.

and ethyl acetate were much higher than that of water under the same conditions. To understand the function of blowing agents in extrusion, the concept of blowing-agent efficiencies was introduced. The ratio of the volume of gas in a unit of weight of foam to the volume of gas that was evaporated in a unit of mass of extrudate was reported as the relative blowing-agent efficiency, whereas that of the volume of gas in a unit of mass of foam to the volume of all blowing agent in the gaseous state in a unit of mass of extrudate was reported as the absolute blowing-agent efficiency. The evaporation ratio was recorded as the amount of the blowing agent that evaporated during expansion to the amount of the blowing agent added to the polymer. The evaporation ratio should be the ratio of the relative blowing-agent efficiency to the absolute blowing-agent efficiency. Under the assumption that the foam stopped expanding and shrinking when the temperature of the extrudate reached the boiling point of the blowing agent, the effects of the temperature and blowing-agent concentration of water, ethanol, and ethyl acetate, in the absence of talc, on the relative blowing-agent efficiency are depicted in Figures 16–18. Similarly, those in the presence of talc are depicted in Figures 19–21. Under the conditions employed in this experiment, all ethanol and ethyl acetate could be evaporated. Therefore, the relative blowing-agent efficiencies and absolute efficiencies of ethanol and ethyl acetate were the same. The absolute blowing-agent efficiencies of water, with and without talc, are shown in Figures 22 and 23, respectively.

The results of the blowing-agent efficiency reveal that it dropped with the blowing-agent concentration in both the presence and absence of talc for all blowing agents. Higher temperatures increased the blowing-agent efficiencies of water and ethanol but decreased the blowing-agent efficiency of ethyl acetate.

The relative efficiency of the blowing agent depended on how much evaporated gas could be held by starch acetate. The capability of starch acetate paste to hold gas depended on two important factors, the cell wall strength and cell density, as shown by the classical pressure vessel strength equation, which simulates a closed spherical cell configuration:

$$P = E\sigma_\mu t / r_i \qquad (5)$$

where P is the net inner pressure; E is a constant; σ_μ is the ultimate strength of the material, which can be taken as the combination of the melting strength and elasticity of starch acetate paste; t is the thickness of the cell wall; and r_i is the inner radius.
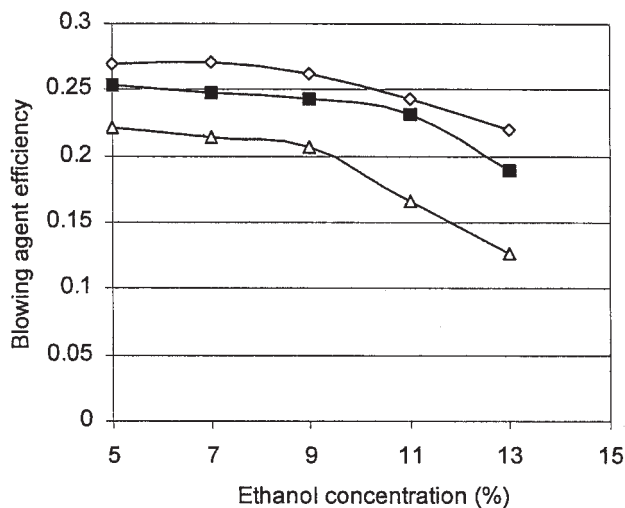

Figure 17 Effects of the ethanol concentration and temperature on the blowing-agent efficiency in the absence of talc.
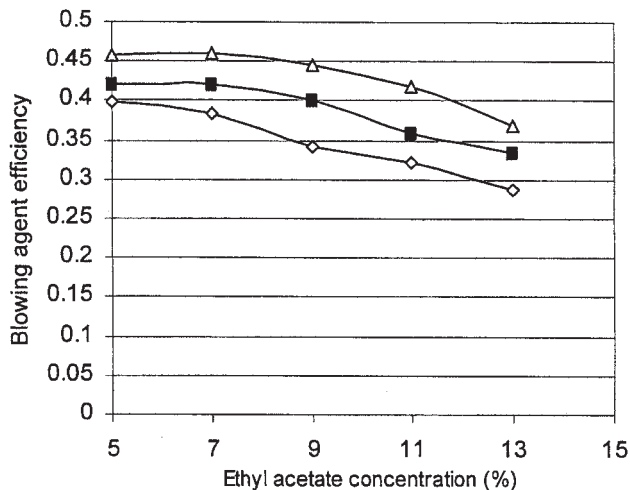
Figure 18 Effects of the ethyl acetate concentration and temperature on the blowing-agent efficiency in the absence of talc.

Figure 20 Effects of the ethanol concentration and temperature on the blowing-agent efficiency in the presence of talc.

According to eq. (5), cells of small radii and of the same thickness can withstand higher pressure. However, the relationship between the pressure that a foam can withstand and the radii of a foam cell is not so simple. Suppose a big cell is broken into n smaller separated cells of the same total volume and weight. The relationship between the thickness (t) and radius (r_i) of individual cell is

$$t = Br_i \tag{6}$$

where B is a constant.

When eqs. (5) and (6) are combined, we find that the cell density or cell radius has no effect on how much pressure it can withstand. Nevertheless, in the extru-
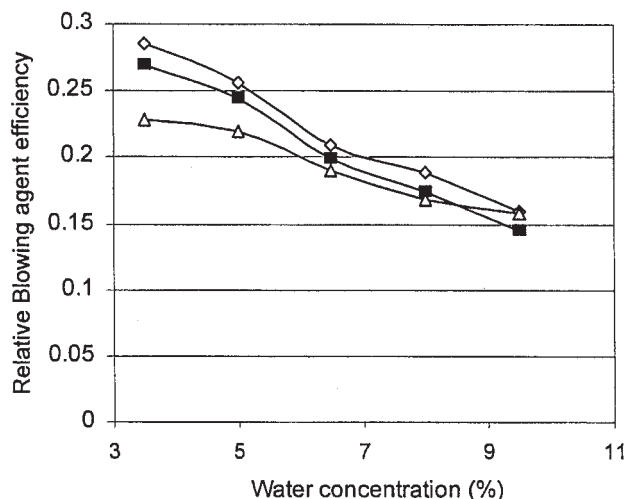
sion of starch acetate, all cells were conjoined. The sharing of some surfaces by various foam cells means the thickness of the cell wall does not drop in proportion to the radius of the cell but much less slowly:

$$t = Br_i f(r_i) \tag{7}$$

where $f(r_i)$ is a thickness adjustment factor related to the radius and geometry of cells. Its value lies between 1 and 2. $f(r_i)$ is 1 when there is only one big cell. It is 2 when all surfaces of a cell are shared with other cells.

For a given amount of the matrix polymer, the smaller the radius is, the more area is shared by neighboring cells and the slower the thickness drop is with the decrease in the cell radius. When eqs. (5) and (7) are combined, it is easy to see that the pressure that a

Figure 19 Effects of the water concentration and temperature on the relative blowing-agent efficiency in the presence of talc.
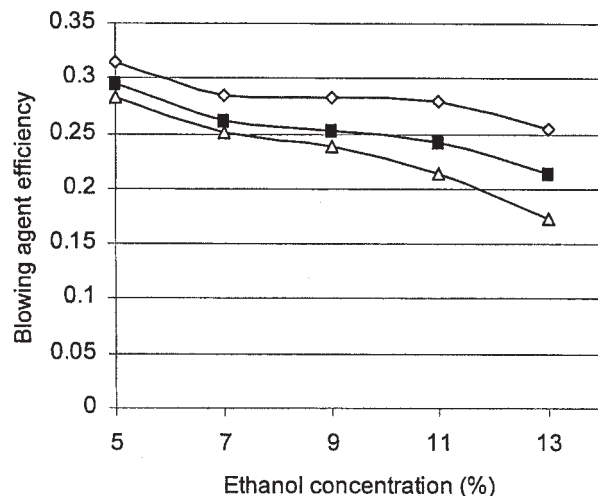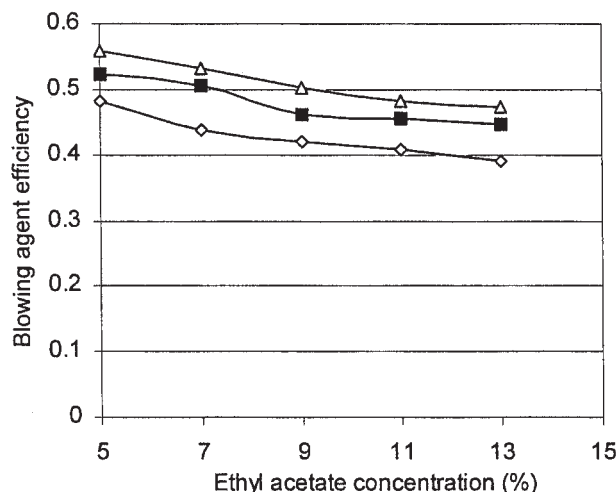

Figure 21 Effects of the ethyl acetate concentration and temperature on the blowing-agent efficiency in the presence of talc.

foam can withstand increases with the cell density. Another important factor in expansion is surface tension, which can be written as follows:

$$P_\sigma = 2\sigma / r_i \qquad (8)$$

Surface tension counteracts cell expansion. It reduces the net inner pressure exerted on the cell wall. The combined action of the radius, in reducing the net positive pressure and in increasing resistance to positive pressure, helps foamed cells hold the evaporated blowing agent. Now, the effect of talc in increasing the specific volume should be obvious. Talc increased the evenness of the cells and reduced the radius of the cells. Therefore, fewer cells merged and less gas leaked out, and this resulted in more expansion.

However, the same force, which may break a foam cell, is also the driving force for expansion. If the melting strength of the matrix polymer is too high, and the blowing-agent concentration is too low, cells may not grow because of a lack of net expansion force. A low specific volume of foam extruded at low blowing-agent concentrations may result from both a high resistance force and a low volume of evaporated gas.

CONCLUSIONS

The cell density and specific volume were two very important functional properties of the studied foams. The final cell density depended on how many nuclei were initiated and how many broke, merged, and grew up. The nucleation process was largely controlled by the physical properties of the blowing agent, that is, the evaporation rate of the blowing agent. The physical properties of the matrix polymer also had some important roles in the nucleation process. The initial nuclei density increased with the con-
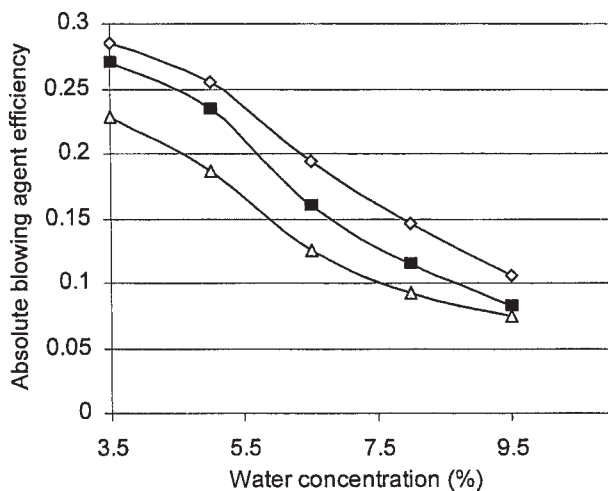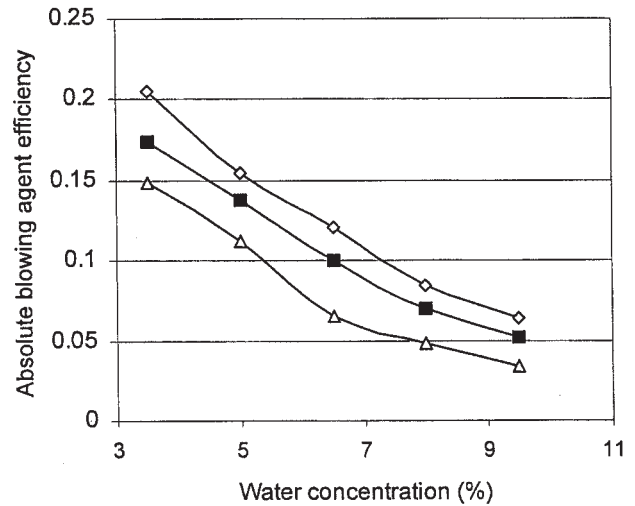

Figure 23 Effects of the water concentration and temperature on the blowing-agent efficiency in the absence of talc.

centration, evaporation rate of the blowing agent, operation temperature, and pressure drop at the extruder die. The physicochemical properties of the matrix polymer and blowing agent and the amount of the blowing agent played major roles in the cell growth process. After nucleation, the final cell density depended on how much the cells grew and how many cells merged. The growth of a cell depended on the supply of evaporated gas. Cell merging and breaking depended on the melting strength of the matrix polymer and the number and evenness of cells. The unevenness of cells led to the overgrowth of cells and the shrinkage or disappearance of small cells, especially in the early stage of cell growth when the strength of the cell wall was still low because of the high residual blowing agent and relatively high temperature. A high melting strength of the matrix polymer was crucial for keeping the final cell density close to the nuclei density.

The specific volume depended on how much of the blowing agent evaporated and how much of the gas the matrix polymer held. The amount of the blowing agent that could be evaporated depended on the latent heat of vaporization, boiling point, and heat capacity of the blowing agent and the temperature of the starch acetate paste when the die was exited. The latent heat of vaporization played an important role in the amount of the blowing agent that could be evaporated by overheating. Water had a much higher latent heat than other blowing agents. The amount of gas that evaporated did not increase much with the amount of water. The heat capacity of the blowing agent also played some role in the amount of gas that evaporated, but its contribution was not great because the blowing agent accounted only for a small amount of the starch acetate paste. Generally, the amount of gas

Figure 22 Effects of the water concentration and temperature on the blowing-agent efficiency in the presence of talc.

that evaporated increased with the concentration of the blowing agents. However, the final specific volume did not always increase with the amount of gas that evaporated. The same blowing agent, which was the driving force of expansion when evaporated, also reduced the capacity of the polymer paste to hold the evaporated blowing agents. The highest specific volumes were achieved when the two counteracting effects were well balanced.

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