



Research Article

Effect of molecular weight on the quality of poly(alpha-methylstyrene) mandrel

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Abstract

Hollow poly(alpha-methylstyrene) (PAMS) shows application in inertial confinement fusion experiments as the degradable mandrels of glow plasma polymer shells. However, the molecular weight of PAMS has great influence on the quality of mandrels. In this work, this influence was systematically studied using several PAMS samples with different molecular weights. For PAMS shells with 900 μm inner diameter and different wall thickness, when the molecular weight of PAMS is in the range of 300–500 kg·mol⁻¹, perfect sphericity and good wall thickness uniformity can be obtained. In contrast, when increasing molecular weight to 800 kg·mol⁻¹, the sphericity and the wall thickness uniformity become worse. Moreover, compared with the wall uniformity, the sphericity of PAMS shells was much less sensitive to the molecular weight. The results also showed that the stability of W/O compound droplets of PAMS shells were less affected by the molecular weight. It was revealed that the wall uniformity and the sphericity of the PAMS shells were associated with the diffusion rates of fluorobenzene (FB).

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1. Introduction

ICF (inertial confinement fusion), an efficient approach to explore new energy, has been rising as a new hot topic nowadays. As an important ignition capsules for ICF, the GDP and PI microspheres can be prepared from poly(alpha-methylstyrene) (PAMS) mandrels by the degradable mandrel technique [1–3]. In this process, these PAMS mandrels are coated with the plasma polymer, followed by a heat treatment to drive the PAMS to the polymer coating with monomer form, and leaving the desired hollow capsules as a result. This shows

that the quality of GDP microspheres is mainly affected by PAMS mandrel [4,5], and so the PAMS microspheres with excellent sphericity and surface finish play a crucial role in the preparation of GDP and PI microspheres to meet the requirement of ICF experiment.

Microencapsulation technology is a popular approach for preparing millimeter-size monodisperse PAMS capsules, which however, cannot fully meet the requirements of the ignition target in terms of sphericity and wall thickness uniformity. The most common process to prepare PAMS capsules with microencapsulation technology is as follows: (1) Preparing W/O composite microspheres, (2) obtaining solidified PAMS microspheres by oil phase removal, (3) drying to eliminate aqueous phase for a hollow PAMS microspheres. It is worthy noting that the quality of PAMS microsphere can be carefully controlled by the first two steps, while the sphericity is significantly affected by the matched density and the

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interfacial tension between the outer aqueous phase and the W1/O composite, which was confirmed by Nagai [6–8] and Takagi [8–10]. The viscosity of the oil phase is of importance to the stability of the emulsion [11,12], and the uniformity of the thickness and concentricity of the shell depend on the density, organic fluid viscosity and concentration of the oil phase [13–15], which are closely related to the molecular weight. Consequently, the morphology and properties of the microsphere are dominated by the molecular weight of PAMS, which has not been reported in detail so far.

To identify the optimum PAMS molecular weight in fabricating high quality capsules, we have employed four different PAMS samples whose molecular weights changing from $300 \text{ kg} \cdot \text{mol}^{-1}$ to $800 \text{ kg} \cdot \text{mol}^{-1}$, and conducted an investigation of the properties of PAMS mandrels, especially emulsion stability, sphericity, thickness uniformity and surface finish. Moreover, the relevant mechanisms of how the molecular weight affecting the wall uniformity and the sphericity of the PAMS shells by influencing the diffusion rates of fluorobenzene (FB) were discussed. The improvement in emulsion stability, sphericity, wall thickness uniformity and surface finish connection with PAMS molecular weight are newly discussed.

2. Experiment

2.1. Materials

The four PAMS samples, PAMS-1 ($M_w = 300 \text{ kg} \cdot \text{mol}^{-1}$), PAMS-2 ($M_w = 440 \text{ kg} \cdot \text{mol}^{-1}$), PAMS-3 ($M_w = 460 \text{ kg} \cdot \text{mol}^{-1}$) and PAMS-4 ($M_w = 800 \text{ kg} \cdot \text{mol}^{-1}$) were synthesized from laboratory, of which the molecular weights were measured by gel permeation chromatography. The other materials were: polyvinyl alcohol (PVA): $M_w = 13\text{--}23 \text{ kg} \cdot \text{mol}^{-1}$, 87%–89% mole hydrolyzed, Aldrich Company; CaCl_2 : Chengdu Kelong Chemical Reagent Factory; FB: Aldrich Company, purified by distillation. Distilled, deionized water was used in the preparation of all aqueous phases.

2.2. Fabrication of PAMS shells

To produce PAMS shells by microencapsulation, a triple orifice droplet generator was used to prepare water droplets (W1) encapsulated by a relatively thick layer of a non-aqueous polymer solution. The microencapsulated compound droplets (W1/O) were suspended in an aqueous bath [16] (shown in Fig. 1).

The PAMS-1/FB solution (O) with 10% PAMS mass fractions was fed to the second orifice of the droplet generator with the pure water added into the center of the oil phase through the first orifice to form a compound droplets with W1/O. The third orifice provided a gentle laminar flow of 2% PVA and 1.5% CaCl_2 solution (W2) around the second orifice (shown in Fig. 1(a)) [17]. Then the W1/O/W2 compound droplets were collected in an 800 mL cylindrical flask filled with 150 mL W2 solution. After about 150 W1/O compound droplets were collected in the cylindrical flask, the flask was gently rotated horizontally at 25 rpm in a heated water bath.

The temperature of the bath was kept at $55 \text{ }^\circ\text{C}$ for 48 h to remove the most FB solvent in the O phase, as shown Fig. 1(b) [18]. After the curing, the PAMS shells were transferred to another beaker and then washed several times with deionized water and ethanol to remove residual PVA and CaCl_2 on their outer surface. Finally, the thoroughly washed PAMS shells containing W1 in the cavity were dried at $45 \text{ }^\circ\text{C}$ at atmospheric pressure for at least 3 days to obtain hollow PAMS capsules, as shown in Fig. 1(c). To enhance the accuracy of characterization results, the above fabrication process was operated 3 more times respectively. The wall thickness of the as-prepared PAMS shell was mainly $10\text{--}20 \text{ }\mu\text{m}$.

In this work, the inner and the outer diameters of the W1/O/W2 compound droplets, which were controlled by the W1, O, and W2 flow rates, were respectively set as $(900 \pm 20) \text{ }\mu\text{m}$ and $(1250 \pm 20) \text{ }\mu\text{m}$ to eliminate the possible effects of shell size and wall thickness on different molecular weights. To obtain the target initial emulsion dimension, the 8 wt% PAMS-2/FB, 8 wt% PAMS-3/FB and 4 wt% PAMS-4/FB ($\eta = (36 \pm 5) \text{ mPa} \cdot \text{s}$) were prepared based on the viscosity value of 10 wt% PAMS-1/FB ($\eta = 36 \text{ mPa} \cdot \text{s}$) as a reference to keep the viscosity roughly at the same level. All the operations above were performed three times in parallel to enhance the accuracy of characterization results.

2.3. Characterization

2.3.1. Density and viscosity parameters test

The density of PAMS/FB solution with different PAMS mass fraction (4%, 8%, 10%, 12%, 15%, 18%, 20%) was measured precisely by a densitometer (Anton Paar®, DMA 5000) at $30 \text{ }^\circ\text{C}$. The densitometer accuracy is $10^{-6} \text{ g} \cdot \text{cm}^{-3}$ for the density measurement and $0.001 \text{ }^\circ\text{C}$ for the temperature measurement. The PAMS/FB solution viscosity was measured precisely by a microviscometer (Anton Paar®, Lovis 2000M) as the viscosity test. The microviscometer accuracy is $0.001 \text{ }^\circ\text{C}$ for temperature measurement.

2.3.2. Morphology and the stability of compound emulsion measurement

W1/O/W2 compound droplets are unstable in thermodynamics, as their stability is possibly affected by PAMS molecular weight. For each batch, 150 W1/O compound droplets were cursorily fabricated to calculate the rate of emulsion survival. In this paper, the percentage of W1/O/W2 double emulsions in the total particles number was used to evaluate the synthesis yield. The morphology and dimension of the W1/O compound droplets were characterized by a digital microscope.

2.3.3. Sphericity and wall thickness uniformity measurement

The diameter and wall thickness (t_w) of PAMS shells were characterized by a measuring microscope (VMR-3020, Nikon). For each batch, at least 50 PAMS shells were randomly picked up from about 150 PAMS shells for characterization. The sphericity of the shells was defined as out-of-round (OOR) and measured by the difference between the

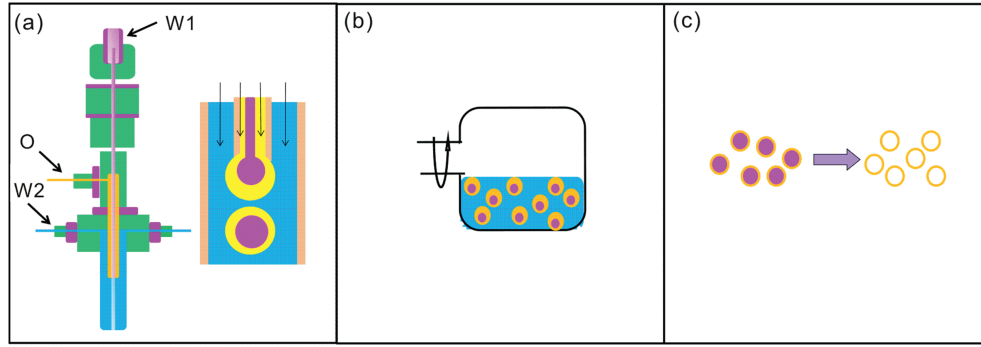


Fig. 1. Fabrication of PAMS shells: (a) generation of W1/O/W2 emulsions by a triple orifice droplet generator; (b) curing of W1/O compound droplets in W2 phase with a rotating cylindrical flask; (c) preparation of hollow poly(alpha-methylstyrene) shells.

maximum and minimum radius of PAMS shells in twelve directions. OOR is defined as:

$$\text{OOR} = \frac{D_{\max} - D_{\min}}{2} \quad (1)$$

where D_{\max} is the maximum value of the twelve measured outer diameters of PAMS shells, while D_{\min} is the minimum one.

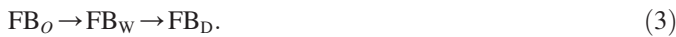
In this work, Δt_w is used to characterize the wall thickness uniformity of PAMS shells. Δt_w is defined as

$$\Delta t_w = t_{w_{\max}} - t_{w_{\min}} \quad (2)$$

where $t_{w_{\max}}$ is the maximum value of the twelve measured wall thicknesses of PAMS shells, while $t_{w_{\min}}$ is the minimum one [19–21].

2.3.4. Solidification rates of PAMS/FB droplets measurement

The O phase diffusion of FB solvent in curing process undergoes two stages. Firstly, the FB diffuses from the oil phase to the outer water phase by dissolving. Then, the FB evaporates from the outer water phase to the atmosphere. The process is illustrated as follows



Nagai [7] et al. pointed out that the evaporation rate of FB to the air was much larger than that of the FB diffuses to the outer water phase. Therefore, the curing process was governed by the FB dissolving rates from the O phase to the W2 phase.

Based on the diameter of PAMS/FB droplets, equation (4) is derived from the law of conservation of mass. The evolution of FB diffusion flux calculated from the formula is used to characterize the solidification rates of PAMS/FB droplets (FB mass per unit time through unit area). In order to simplify the calculation, the concentration process of PAMS solution with no volume effect is assumed, as the volume of FB diffusion is the reduced volume of the droplets.

$$\Omega_i = \frac{\Delta m}{s \times \Delta t} = \frac{2}{3} \cdot \frac{(r_{i+1}^3 - r_i^3) \cdot \rho_{\text{FB}}}{(t_{i+1} - t_i) \times (r_i^2 + r_{i+1}^2)}, \quad (4)$$

where Ω_i is the evolution of FB diffusion flux at the i th time, r is the radius of O/W2 drop during the curing process measured

by the digital microscope, ρ_{FB} is the density of the FB solvent, and t is the curing time.

3. Results and discussion

3.1. Effect of molecular weight on density and viscosity of PAMS/FB oil phase

It has been reported that controlling the density matching among the W1, O and W2 phases is one of the most effective methods to improve the wall thickness uniformity and the sphericity of polymer shells. Meanwhile, the higher initial concentration of the O phase, which leads to an increased viscosity, is important to the stability of the emulsion.

Generally, the density and viscosity of PAMS/FB solution are increased when the PAMS concentration is increased. Relations between the average density and viscosity profiles of a W1/O compound droplet and the different molecular weight of PAMS at 30 °C are shown in Fig. 2(a) and (b). It can be seen that for the different initial PAMS molecular weight of the O phase, the density increases linearly with its concentration, while the viscosity increases exponentially. Especially, the density values have slight distinctions among the four samples, but the viscosity values exhibit a dramatic difference. The higher molecular weight of PAMS obviously possesses tremendous viscosity values compared to the lower one, and the values tend to increase with large magnitude as well. Since in higher molecular weight polymer solution with long chain the bond–bond interaction among the molecules is stronger, the solution shows lower mobility. Hence, the molecular weight is an important factor attributed to the disparity of the viscosity among the four PAMS samples.

3.2. Effect of molecular weight on the stability of W1/O compound emulsion

Multiple emulsions are thermodynamically unstable due to the excess free energy associated with the surface of emulsion droplets. Marangoni instabilities driven by surface tension may result in outer surface deformations [7]. Marangoni convection cells may be the origin of the density matching level among the three phases. Similarly, the shear force and

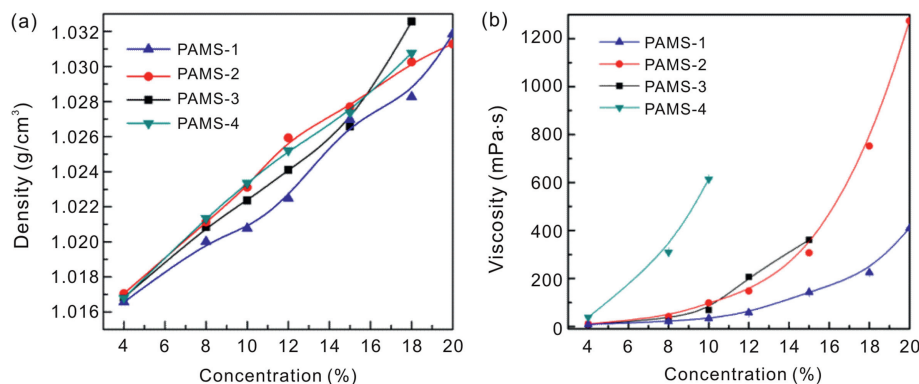


Fig. 2. (a) Density and (b) viscosity of PAMS-1/FB, PAMS-2/FB, PAMS-3/FB, PAMS-4/FB with different concentrations.

interfacial tension are nonlinearly connected with variable viscosity in O phase. Hence, it is essential to study the effect of molecular weight on the stability of compound droplets in favor of fabricating high quality final capsules. The statistic results of the experiment which was repeated 4 times are shown in Fig. 3. It can be seen that the molecular weight of O phase changing from $300 \text{ kg} \cdot \text{mol}^{-1}$ to $800 \text{ kg} \cdot \text{mol}^{-1}$ are not expected to exert an immense influence on the stability of W/O/W emulsions. The association of initial emulsion dimensions, W1 and W2 phases et al. with a series of control factors, which may lead to the changing quality of the final capsule, has also been considered to avoid the instability. As a consequence, the stability of W1/O compound emulsion is independent on the molecular weight.

3.3. Effect of molecular weight on the solidification rates of PAMS/FB emulsions

The uniformity of capsules is derived from the repeated deformations of the emulsions before the viscosity and concentration are increased. Delaying the curing rates is the key to ensure good sphere uniformity. Double emulsions have enough time to center and resist deformation before the emulsion loses its fluidity, which is attributed to the slowing down rates of FB diffusion. According to Eq. (4), the evolution of FB diffusion flux using three different PAMS molecular weights in the O phase with different W2 phase are shown in

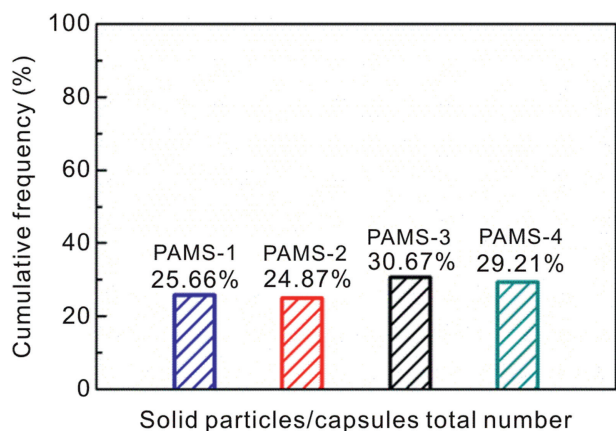


Fig. 3. Stability of W1/O emulsion with PAMS-1–4.

Fig. 4(a)–(b). The results present that the process of FB diffusion evolution flux achieved the maximum is prolonged with the decreasing molecular weight. Therefore, the curing time can be extended by reducing the molecular weight of PAMS and the diffusion rates of FB can be accordingly decreased. From the above results, it can be seen that the lower molecular weight of PAMS is favorable to improve the wall thickness uniformity and the sphericity of PAMS shells with the diffusion rates of FB prolonged.

To explain the effect of molecular weight on the diffusion rates of FB, a diffusion transport model was investigated for the emulsion system. According to Vrentas and Duda [22], an additional assumption must now be applied in order to derive an expression which relates the binary mutual diffusion coefficient D_m to the self-diffusion coefficients of the solvent and the polymer, D_1 and D_2 (small molecule is defined as component 1, while polymer is defined as component 2). The connection between D_m and the self-diffusion coefficients is made via the friction coefficients for the system ζ_{11} , ζ_{12} , and ζ_{22} [23]

$$D_1 = \frac{RT}{N_A^2(\rho_1\zeta_{11}/M_1 + \rho_2\zeta_{12}/M_2)}, \quad (5)$$

$$D_2 = \frac{RT}{N_A^2(\rho_2\zeta_{22}/M_2 + \rho_1\zeta_{12}/M_1)}, \quad (6)$$

$$D_m = \frac{M_2\rho_1V_2}{N_A^2\zeta_{12}} \left(\frac{\partial\mu_1}{\partial\rho_1} \right)_{T,p} = \frac{M_1\rho_2V_1}{N_A^2\zeta_{12}} \left(\frac{\partial\mu_2}{\partial\rho_2} \right)_{T,p}, \quad (7)$$

where N_A is Avogadro constant; ρ_i and M_i are the density and molecular weight of component i ; ζ_{ij} is the friction coefficient between i and j ; and μ_i is chemical potential of component i .

Eq. (5) illustrates the FB diffusion coefficient dependences of the PAMS molecular weight. Since M_2 varied from $300 \text{ kg} \cdot \text{mol}^{-1}$ to $800 \text{ kg} \cdot \text{mol}^{-1}$, there is no need to consider the variation of the effective entanglements in the system with temperature, density, and friction coefficients. Obviously, the decreasing value of M_2 leads to the reducing value of D_1 , which is also available for reducing diffusion rates of FB. Consequently, the lower molecular weight of PAMS contributes to delaying the solidification rates, prolonging the curing time and obtaining high sphericity and monodispersed-sized PAMS shells.

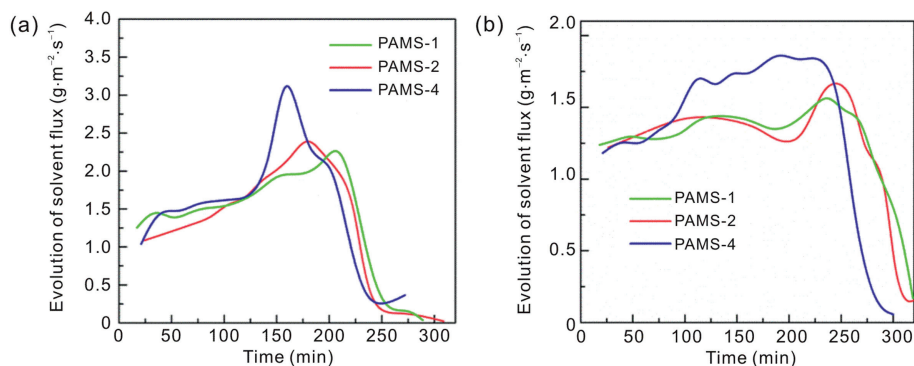


Fig. 4. Evolution of FB diffusion flux profile for PAMS-FB emulsion droplets with different W2 phase: (a) PVA solution; (b) PAA solution.

3.4. Effect of molecular weight on the sphericity of hollow PAMS microspheres

Under a given temperature of 55 °C, the aqueous solution with mass concentrations of 2.0% PVA and 1.5% CaCl₂ was chosen as the W2 phase. As a result, hollow PAMS microspheres with 850 μm inner diameter and different wall thickness were obtained. According to Eq. (1), the sphericity of the resulting PAMS shells produced using the four different PAMS molecular weights in the O phase are shown in Fig. 5(a). To further improve the accuracy of results, the experiment was repeated for 3 more times. As indicated in Fig. 5(b)–(d), it is clearly seen that the cumulative frequency profile of OOR differs significantly for samples with different weights. Within the scope of OOR values <10 μm, the cumulative frequency of PAMS-1–3 rapidly achieves to 100%

compared with that of PAMS-4. Moreover, the proportion from PAMS-4 increases obviously with regard to the OOR values >10 μm. It can be seen that using PAMS molecules with weights ranged from 300 kg·mol⁻¹ to 500 kg·mol⁻¹ can obtain more satisfactory sphericity of hollow PAMS microspheres than using 800 kg·mol⁻¹ one. Furthermore, the OOR values of the PAMS with molecular weight <500 kg·mol⁻¹ are close to each other, and the sphericity is more sensitive to the one with higher weight.

3.5. Effect of molecular weight on the wall thickness uniformity of PAMS shells

According to Eq. (2), to further characterize the quality of the PAMS capsules fabricated using the four PAMS samples with different molecular weight in the O phase, the cumulative

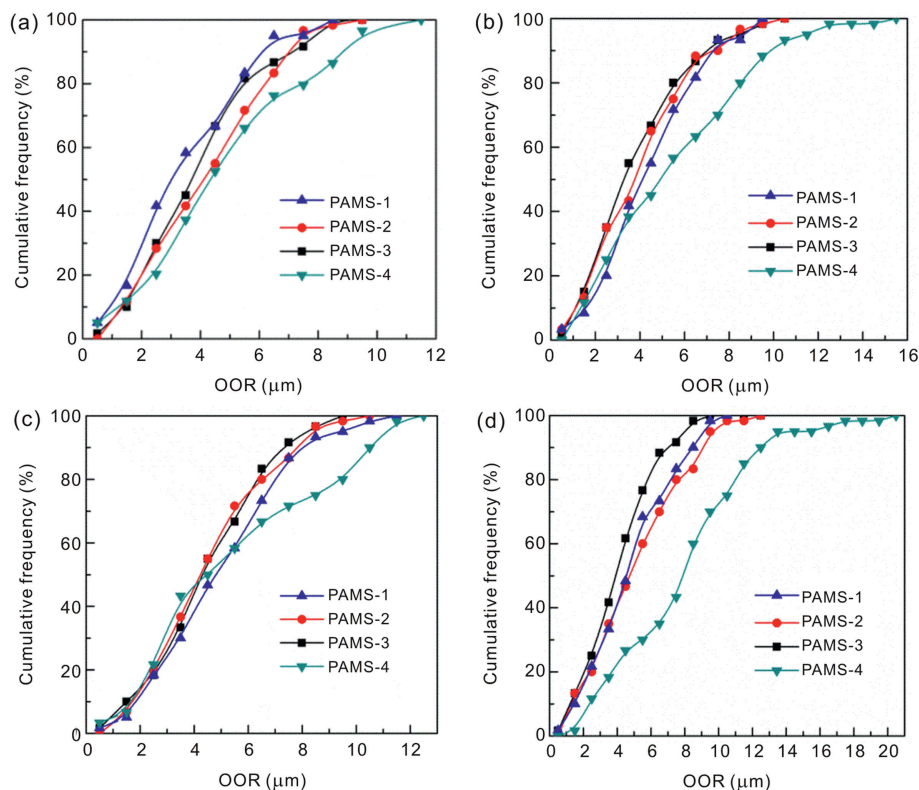


Fig. 5. (a)–(d): Repetitive measurements for 4 times of OOR v.s. cumulative frequency of PAMS-1–4 capsules.

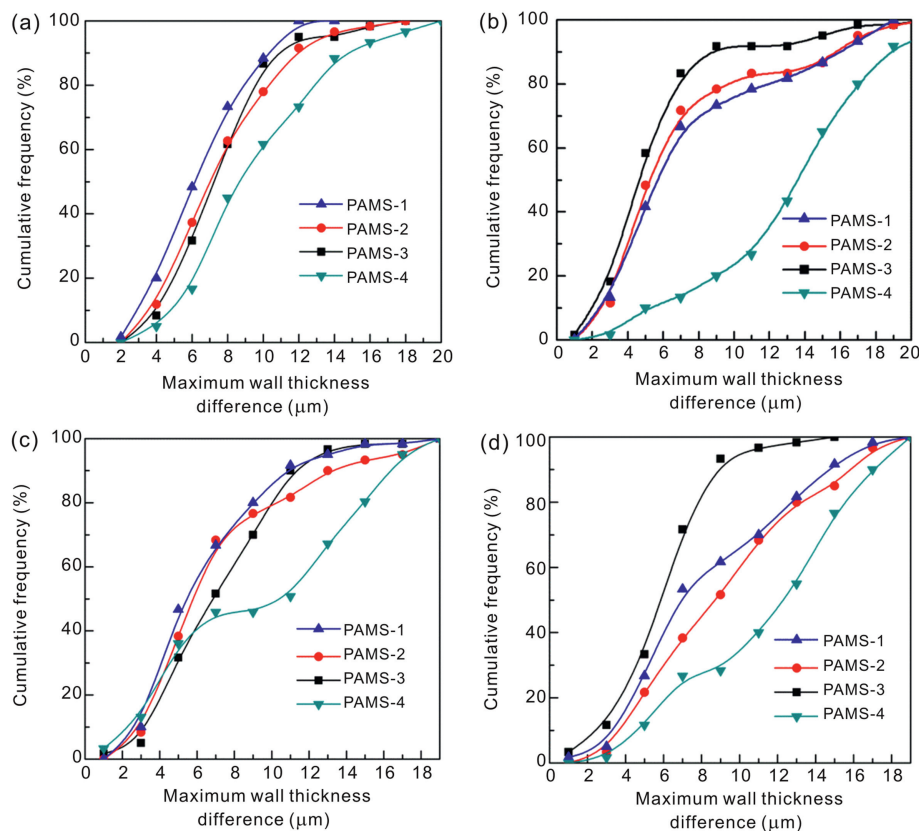


Fig. 6. (a) ~ (d): Repetitive measurements for 4 times of Δt_w v.s. cumulative frequency of PAMS-1—4 capsules.

frequency curves of the wall thickness uniformity (Δt_w) values for the outer spherical surface of the microcapsules are shown in Fig. 6(a). Similarly, the experiment were repeated for 3 more times and the results are shown in Fig. 6(b)–(d). As shown in Fig. 5, PAMS shells with the molecular weights ranging from $300 \text{ kg}\cdot\text{mol}^{-1}$ to $500 \text{ kg}\cdot\text{mol}^{-1}$ are provided with more satisfactory wall thickness uniformity. In contrast, the wall thickness uniformity of PAMS shells prepared from $800 \text{ kg}\cdot\text{mol}^{-1}$ material is substantially the worst. Moreover, seeing from Figs. 3 and 5, it can be further found that the wall thickness uniformity of PAMS shells is more sensitive to the variety of molecular weight than the sphericity of the capsules. Thus, PAMS with modest molecular weight ($300\text{--}500 \text{ kg}\cdot\text{mol}^{-1}$) is more suitable to offer a satisfactory final microsphere performance.

4. Conclusions

To investigate the effects of molecular weight on the preparation and quality of PAMS shells, thick-walled PAMS shells with $900 \mu\text{m}$ inner diameter and different wall thicknesses were fabricated by microencapsulation. The results show that the stability of W1/O compound emulsion is less sensitive to the PAMS molecular weight. However, the sphericity and wall thickness uniformity are significantly influenced by the molecular weight. Compared with the PAMS shells with $800 \text{ kg}\cdot\text{mol}^{-1}$ molecular weight, the PAMS shells with molecular weights in the range from $300 \text{ kg}\cdot\text{mol}^{-1}$ to

$500 \text{ kg}\cdot\text{mol}^{-1}$ show better sphericity and wall thickness uniformity, and their OOR values and Δt_w values are close to each other. Compared with the wall thickness uniformity, the sphericity of PAMS shells is less sensitive to the molecular weight. Moreover, the curing time can be extended by reducing the molecular weight of PAMS and the diffusion rates of FB can be accordingly decreased. Therefore, in fabricating high quality capsules with relative lower molecular weight ($300\text{--}500 \text{ kg}\cdot\text{mol}^{-1}$) should be applied to the PAMS/FB oil phase.

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