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Investigation of particle size effect on film formation of polystyrene latexes using fluorescence technique

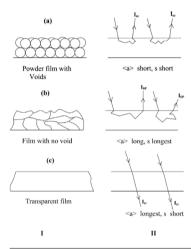
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HIGHLIGHTS

GRAPHICAL ABSTRACT

- Fluorescence technique was used to investigate particle size effect.
- Film formation occurs at lower temperatures with less energy with decreasing size.
- Results obtained from optical data were verified with AFM images.



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ABSTRACT

This study reports the particle size effects on the film formation behavior of pyrene labeled polystyrene (PS) latexes by a combination of steady state fluorescence (SSF), UV–vis (UVV) and atomic force microscopy (AFM) techniques. For this purpose, PS latex dispersions of same molecular weight but different particle size were used. Four different latex films were prepared from dispersions of 203, 320, 400 and 900 nm PS particles by drop casting at room temperature and annealed at elevated temperatures in the range of 100–250 °C for 10 min time intervals above the glass transition (T_g) temperature of PS. Scattered light intensity (I_{sc}) and fluorescence intensity (I_P) from P were measured after each annealing step to monitor the stages of film formation. Evolution of transparency of latex films were monitored using photon transmission intensity, I_{tr} . Atomic force microscopy (AFM) was used to detect variation in the physical structure of annealed films. Onset temperatures for minimum film formation (T_0), void closure (T_v), and healing (T_h) processes were determined. The film formation stages (void closure (ΔH) shifted to lower temperatures as PS particle size decreases. In addition, void closure (ΔH) and interdiffusion (ΔE) energy values also decreased with decreasing the particle size. These

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results showed that the PS particles undergo complete film formation at much lower temperatures and requiring less energy as the particle size decreases. AFM images were also verified these results produced from optical data.

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1. Introduction

Polymer latexes have been important to industry for many decades due to their performance and in a number of applications in a wide variety of products in daily life. They are often used as film-forming agents in paints, adhesives, varnishes, paper coatings, binders, etc. They have many applications including functioning as a film former or a binder in paints and coatings. For example, in a typical paint formulation, a binder is the main factor that determines the physical, mechanical, and chemical properties of the coating film [1]. Obviously, film formation is a critical aspect of all latex applications that involve coating a surface or forming a sheet. For many years, researchers in industry as well as in academia have been exploring various strategies [2] to improve latex film properties. In order to obtain good film properties, excellent film formation is required. To achieve this, a certain amount of coalescent agent is often used, which ultimately evaporates from the coating. In recent times, the environmental and health concerns due to emission of large amounts of volatile organic compound (VOC) have forced the paint and coating industry to move away from the traditional organic based formulations towards water based dispersions. Hence the water-based latexes have become an attractive alternative to the traditional formulations [3–5]. Latex film formation is a complicated, multistage phenomenon and depends strongly on the characteristics of latex particles. Film formation from soft (low-Tg) and hard (high-Tg) latex dispersions can occur in several stages [3]. In both cases, the first stage corresponds to the wet initial stage. Evaporation of water leads to second stage in which the particles form a close packed array, here if the particles are soft they are deformed to polyhedrons. Hard latex however stays undeformed at this stage. Annealing of soft particles causes diffusion across particle-particle boundaries which lead to a homogeneous continuous material. In the annealing of hard latex system, however, deformation of particles first leads to void closure [6,7] and then after the voids disappear diffusion across particle-particle boundaries starts, i.e. the mechanical properties of hard latex films evolve during annealing; after all water has evaporated and all voids have disappeared.

An exhaustive review of the literature [1,2,8–25] showed that the latex film formation is influenced by several factors including the polymer glass transition temperature (Tg) [1], particle size and size distribution [8–10], particle morphology [2,11], drying conditions[10], film forming and curing temperatures [12,13], molecular weight [14,15], surfactant level [16,17], plasticizers and pigments [18], and so on. In recent years, more attention was paid to the particle size effect on film formation from latexes [19–25]. The latex particle size and distribution are of primary importance because of their strong influence on the chemical [19,20], physical [21,23,24] and mechanical properties [22,26] of the polymer colloids. It is well known that films made from nanosize latexes in general have superior gloss, solvent resistance, and adhesion and produce smoother films. It is apparent that the particle dimensions and polydispersity affect the number of defects during the packing step of the film formation [24]. These defects act as fracture initiators when the coating is submitted to mechanical strength [22]. A good packing of polymer particles is a condition to obtain high performance films from latexes [27]. Goudy et al. [28] observed

that smaller particle size resulted in a faster rate of interparticle fusion for the polystyrene (PS) latex in the size range of 0.24–1.05 µm. Mazur et al. [29] confirmed the existence of a maximum particle size and minimum packing fraction in the sintering of acrylic copolymer lattices excluding any contribution from viscous flow. Eckersley and Rudin [30] have demonstrated the decrease in the degree of interparticle fusion with increasing particle size in MMA/BA/AA films using scanning electron microscopy. Boczar et al. [31] studied the polymer interdiffusion between latex particles in BMA lattices prepared with particle sizes of 100 and 300 nm using fluorescence technique of nonradiative energy transfer (NET). They have found that the mixing rate is about 3 times greater for 100 nm particles. Do Amaral et al. [23] also studied the latex particle size and distribution on the rheological and adhesive properties of waterborne acrylic pressure-sensitive adhesives (PSA) films. Both the rheological and adhesive properties were found to be very dependent on the initial latex particle size distribution.

Transmission electron microscopy (TEM) has been the most common technique used to investigate the structure of the dried films [32]. Pattern of hexagons, consistent with face centered cubic packing, are usually observed in highly ordered films. When these films are annealed, complete disappearance of structure is sometimes observed, which is consistent with extensive polymer interdiffusion. Freeze fracture TEM (FFTEM) has been used to study the structure of dried latex films [33]. Small angle neutron scattering (SANS) has been used to study latex film formation at molecular level. Extensive studies using SANS have been performed by Sperling and co-workers [34] on compression-molded polystyrene film. Winnik and coworkers introduced the direct-nonradiative energy transfer (DET) method, which is based upon fluorescence decay measurements, to study polymer diffusion rates [35]. A typical approach includes the synthesis of two virtually identical samples of latex particles, one of which was labeled with a fluorescent donor dye (D), while the other was labeled with an acceptor dye (A). Latex films were cast from a mixture of the two dispersions. D and A are initially separated from each other in a newly formed film. DET does not occur at this stage and no fluorescence decay can be observed. As polymer chains diffuse, D and A are brought together. DET take places and leads to a strong decrease in the measured fluorescence decay. Using this technique, a number of factors that affect the rate of polymer diffusion in latex films were explored, which include temperature, composition of the latex, and the presence of various additives such as coalescing agents and common surfactants [36]. The steady state fluorescence technique combined with DET has been used to examine healing and interdiffusion processes in the dye labeled poly(methyl methacrylate) (PMMA) latex systems [37,38]. Recently UV-visible technique was used to study film formation from PMMA and polystyrene (PS) particles [39–44] where the transmitted light intensity was monitored during film formation process.

In the present study, we investigated the film formation behavior of the PS latex particles depending on particle size. For this purpose, four polystyrene latexes with the same molecular weight but with different particle size were synthesized by emulsion polymerization technique.

The film formation stages and morphology of PS latexes during the annealing process were investigated by steady state fluores-



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