



Synthesis and properties of SiO₂/P(MMA-BA) core-shell structural latex with siloxanes



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ARTICLE INFO

Article history:

Received 15 October 2015

Received in revised form 18 March 2016

Accepted 28 March 2016

Available online 8 April 2016

Keywords:

Core-shell latex

Nanocomposite coatings

Weathering durability

Siloxane-acrylic resin latex

ABSTRACT

In this study, we have successfully introduced siloxane monomer, 2,4,6,8-Tetravinyl-2,4,6,8-tetramethylcyclotetrasiloxane (V₄) into the shell of silica/poly(methyl methacrylate-butyl acrylate) (SiO₂/P(MMA-BA)) core-shell nanocomposite colloids *via in-situ* emulsion copolymerization. The obtained nanocomposite films with 50% silica content all show excellent transparency (nearly 100% transmittance in visible range) and film-forming property (T_g under 30 °C). More importantly, the addition of V₄ can significantly increase the weathering durability of the film by improving the water repellency, cross-linking degree and UV-blocking property of the film.

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1. Introduction

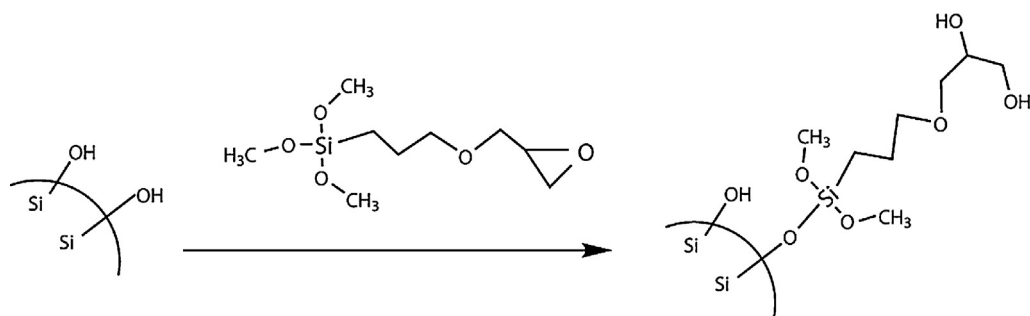
Acrylic latexes have been widely used in coatings, adhesives for papers, buildings, equipments, ships, automobiles and so on, not only because of their low price and excellent properties, but also their low even zero volatile organic compounds compared to traditional solvent-based coatings. However, there are still some drawbacks that limit their more applications [1], e.g., poor water/solvent resistances, low hardness, and not good enough weatherability. In contrast, polysiloxanes have demonstrated many satisfactory properties, such as high flexibility and hydrophobicity, low surface energies, low glass transition temperatures (T_g), excellent thermal stability, outstanding weathering resistance [2]. Accordingly, a lot of efforts have been made to fabricate siloxane-modified acrylic latexes with better integrated coating properties in the past decades, by using block polymers [3], graft polymers [4,5], interpenetrating polymer networks [6,7], core/shell structure (using either polysiloxane [8,9] or polyacrylate as seeds [10,11]). The obtained colloids with various morphologies show superior properties which can cater to the needs of specific applications. For example, Bai et al. prepared polysilsesquioxane/polyacrylate/polydimethylsiloxane (PDMS) hybrid latex particles with PDMS rich on the surface of the hybrid latex film [12], the

results showed that introducing PDMS into the system significantly increased the hydrophobicity of the film. Park et al. synthesized a superweatherable silicone-acrylic resins from the copolymerization of *n*-butyl acrylate (BA), methyl methacrylate (MMA) and 3-methacryloxypropyltrimethoxysilane (MPTS) [13]. Zielecka et al. found that the silicone-acrylic-polyolefin polymer could be applied as antisoiling coatings for protection of porous building materials, by controlling the cross-linking process [14]. Hu and his coworkers did a series of works on epoxy coatings modified with silane monomers, either by simple physical blending or by chemically grafting, the obtained films demonstrated better adhesion, less water uptake and swelling, and dramatically enhanced corrosion resistance [15–18]. Zou et al. used gamma ray to induce BA and styrene in the presence of vinyl-containing polysiloxane latex *via* seeded emulsion polymerization [9], and the obtained composite latex films would not yellow after accelerated weathering test for 573 h when the seed content of polysiloxane reached above 10%.

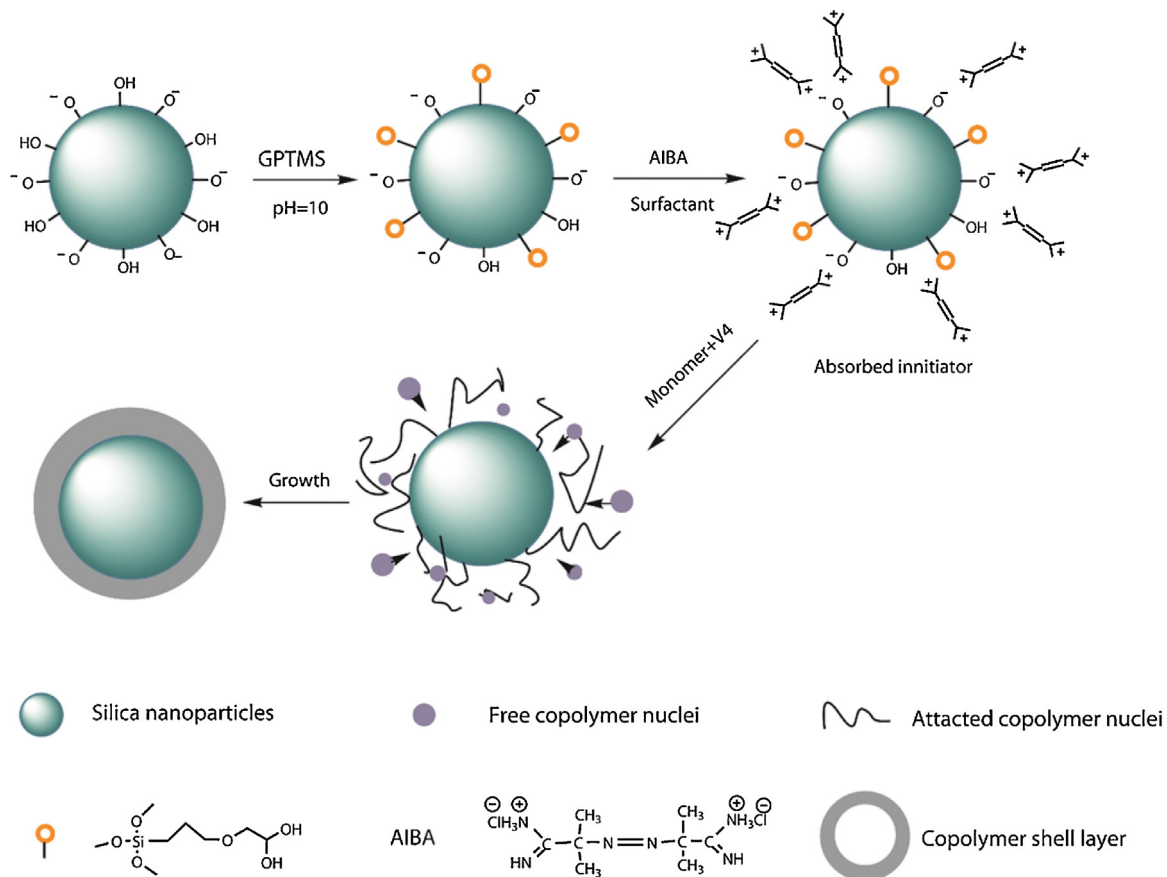
In our previous work [19], we have successfully introduced as high as 60 wt.% of inorganic silica into acrylic latex by synthesizing SiO₂/P(MMA-BA) core-shell structure *via in situ* emulsion copolymerization. The nanocomposite film presented excellent transparency and mechanical property compared with its pure polymer film. More importantly, the incorporation of silica nanoparticles can still increase the UV-blocking property which would cause enhanced weatherability. In order to further improve the aging resistance of the hybrid film, in this study, we introduced siloxanes, using 1,3,5,7-tetravinyl-1,3,5,7-tetramethylcyclotetrasiloxane (V₄) as the model compound, *via*

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Scheme 1. Schematic diagram of silica sol modification with GPTMS.



Scheme 2. Schematic illustration of the formation of $\text{SiO}_2/\text{P}(\text{MMA-BA})$ nanocomposite particles.

Table 1
Formulations of the nanocomposite latex.

Runs	GPTMS- SiO_2 (g)	Monomers (g)			V_4 (g)	Diameter ^a (nm)	PDI	Conversion ^b (%)
		n-BA	MMA	CHMA				
1	0	10	10	0	0	91.5	0.180	99.3
2	50	10	10	0	1	91.5	0.168	98.1
3	50	10	10	0	2	95.9	0.157	96.8
4	50	10	10	0	3	106.2	0.162	94.7
5	50	10	5	5	0	93.5	0.167	99.0
6	50	10	5	5	1	104.1	0.180	98.6
7	50	10	5	5	2	88.9	0.180	97.2
8	50	10	5	5	3	97.2	0.157	97.3
13	0	20	20	0	0	87.7	0.082	99.0

^a by Dynamic light scattering.

^b by gravimetry.

in situ emulsion polymerization, into the shell of this $\text{SiO}_2/\text{P}(\text{MMA-BA})$ core-shell latex with high loading SiO_2 and high solid content.

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