



Property study of structured self-crosslinking acrylic latex binder: Effect of molar mass and particle design



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ABSTRACT

In the present contribution, the optimization of coating properties of structured self-crosslinking latexes based on diacetone acrylamide and adipic acid dihydrazide crosslinking system by varying particle design and molar mass was investigated. Latexes were prepared by the semi-continuous non-seeded emulsion polymerization of methyl methacrylate, butyl acrylate, diacetone acrylamide and methacrylic acid as main monomers. Two series of latex particles of hard-core/soft-shell or soft-core/hard-shell design were synthesized with a predominant soft phase. In both series, molar mass of the soft phase was gradually reduced by isooctyl 3-mercaptopropionate included in the synthesis. The results show that desired coating properties can be tailored successfully by varying the molar mass and particle design. Better properties exhibited coatings drawdowned from latexes of soft-core/hard-shell particle design. A moderate reduction of molar mass of emulsion copolymers caused a significant improvement of adhesion and blocking resistance, obviously due to enhanced coalescence leading to favoured inter-particle diffusion of copolymer molecules of reduced molar mass.

1. Introduction

Currently, environmental and legislation pressure has driven coating industries to produce coatings with minimum content of volatile organic compounds (VOCs). Hence, waterborne latexes represent a promising alternative to organic solvent-based systems in coatings applications. Acrylic latexes are frequently used binders due to their low cost, and easy preparation. However, their performances such as relatively poor water resistance, surface tackiness and unsatisfactory film hardness limit their practical application in many fields. Therefore, conventional acrylic latex paint compositions are usually based on polymers with relatively high glass transition temperature (T_g) and coalescing agents that are VOCs. In order to reduce the level of VOCs and to improve the performance of latex coatings, the applications of structured core-shell latexes [1–4] and crosslinking chemistry in emulsion polymers [5–9] have attracted a considerable interest in recent years.

The use of structured core-shell latex particles in which two or more different polymers coexist allows an optimal combination of

properties from both components. This largely depends on the morphological features of the latex particles; hence, the investigation of latex particle morphology and factors controlling it has been the goal of many scientists [10–12]. Structured polymeric materials exhibit properties that are often superior to those of their component homopolymers due to their morphological structure. Core-shell particles are often designed with the low T_g polymer forming a shell around the high T_g core [13]. In this hard-core/soft-shell morphology, the low T_g shell facilitates film-formation while the high T_g core enhances the mechanical properties of the film. Li et al. [14] evaluated the effect of different glass transition temperatures of core and shell phases finding that latexes with a high T_g core and a low T_g shell can coalesce better than the representatives with a low T_g core and a high T_g shell. When the traditional morphology is inverted, the high T_g polymer forms a shell around the soft, low T_g core [15]. The soft-core/hard-shell morphology has received less attention due to the fact that the high T_g shell not only limits polymer inter-diffusion across particle boundaries, but also restricts the mobility of the soft polymer in the particle core [16]. However, Santos et al. found [17] that core-shell latex particles with

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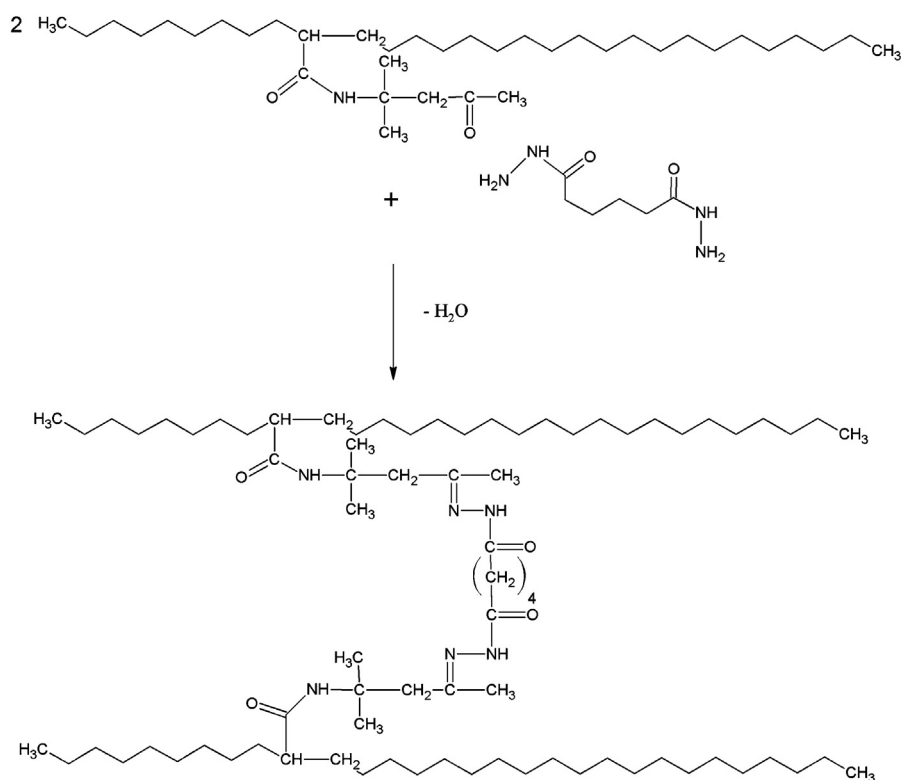


Fig. 1. Scheme showing the crosslinking reaction of ketone carbonyl groups with adipic acid dihydrazide.

poly(*n*-butyl acrylate) cores and poly(methyl methacrylate) shells in the content of 20 wt.% formed films with good mechanical strength at room temperature. This combination of coalescent free film formation and good mechanical properties was attributed to inclusions of soft cores in a continuous matrix of deformed tough shells.

Among different crosslinking reactions, the keto-hydrazone self-crosslinking has been studied extensively [18–22]. The latexes based on diacetone acrylamide (DAAM) and adipic acid dihydrazide (ADH) self-crosslinking system can get cured rapidly at room temperature and do not need any additional crosslinker to be added before use. The reaction between the carbonyl functionalities of DAAM and hydrazone groups of ADH proceeds rapidly at ambient temperature and is favoured by the loss of water and the simultaneous decrease in pH arising from the evaporation of ammonia or amines during the film forming process [23]. The network formation mechanism based on the keto-hydrazone crosslinking reaction is depicted schematically in Fig. 1. Nevertheless, by introducing the self-crosslinking chemistry in emulsion polymers, the physical and chemical integrity of latex films is enhanced, while the film formation process may be complicated [24]. In order to obtain increased mechanical properties, chemical resistance, and high cohesive and adhesive strength, an emulsion coating polymer requires a balance between chemical inter-particle crosslinking and polymer chain mobility resulting in sufficient inter-diffusion during the particle coalescence phase. The compromise between these two antagonist properties can be tailored by a proper design of latex particles. Hence, a precise control of particle design is particularly important in the case of self-crosslinking latexes [25–27].

In our recent work [28] we focused on the effect of molar mass of shell polymer on coating properties of keto-hydrazone self-crosslinking latexes based on core–shell microgel particles. We have found that the reduction of molar mass of shell polymer improved significantly coalescence of latex particles resulting in decreased minimum film forming temperature and enhanced water resistance. As the self-crosslinking latexes based on DAAM and ADH crosslinking system are usually composed of core–shell particles with DAAM repeat units localized in the shell layer, the influence of structured particle design and

morphology should be explored in particular. Nevertheless, no investigations on the influence of particle design concerning the self-crosslinking latexes have been published to the best of our knowledge so far and this lack of information has motivated the present work. This study assesses the combined effects of particle morphology, glass transition temperature and molar mass on coating properties of self-crosslinking latexes, confronting especially fundamental characteristics of emulsion copolymers with end-use properties of coating films. These properties included minimum film-forming temperature, hardness, adhesion, blocking resistance and water sensitivity. The attention is focused on the optimization possibility of desired coating properties of self-crosslinking latexes as well.

2. Materials and methods

2.1. Materials

Self-crosslinking latexes investigated in this research work were synthesized of methyl methacrylate (MMA), butyl acrylate (BA), methacrylic acid (MAA) and diacetone acrylamide (DAAM). All the monomers were purchased from Sigma–Aldrich (Czech Republic). Disponil FES 993 (BASF, Czech Republic) was used as a surfactant and ammonium persulfate (Lach-Ner Company, Czech Republic) was utilized as an initiator of the polymerization reaction. Isooctyl 3-mercaptopropionate (Sigma–Aldrich, Czech Republic) served as a chain transfer agent (CTA). Adipic acid dihydrazide (ADH) was utilized as the crosslinker and was purchased from Sigma–Aldrich, Czech Republic. Tetrahydrofuran (THF, Lach-Ner Company, Czech Republic) was used as the solvent for molar mass distribution measurements. All the chemicals were utilized as received without any further purification.

2.2. Latex preparation and characterization

Two series of hard-core/soft-shell and soft-core/hard-shell latex particles, respectively, with a predominant soft phase were synthesized comprising a variable content of acrylic monomers (see Table 1). The

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