



Effect of N,N-dimethylacrylamide (DMA) on the comprehensive properties of acrylic latex pressure sensitive adhesives

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ABSTRACT

Acrylic pressure sensitive adhesive (PSA) latexes were synthesized via a monomer-starved seeded semi-continuous emulsion polymerization process with butyl acrylate (BA), methyl methacrylate (MMA), N,N-dimethylacrylamide (DMA), acrylic acid (AA) and 2-hydroxyethyl acrylate (HEA) as monomers. Impacts of DMA on the resultant latex and PSA properties were comprehensively investigated. Results indicated that latex particle size was independent of the amount of DMA in the pre-emulsion feed with excessive and constant surfactant concentration. Latex viscosity increased with DMA concentration. It was also found that water resistance of acrylic latex PSA became worse by the presence of DMA, confirmed by water contact angle measurements. Besides, DSC results showed that as the amount of DMA increased, glass transition temperatures (T_g) of the polymers were elevated significantly. TGA results showed that thermal stability of PSA was improved with DMA as a co-monomer. Furthermore, as DMA amount increased, gel content slightly increased, while sol molecular weight (M_w , M_n) of the polymer decreased. Finally, with respect to the adhesive properties of the PSA, it was observed that loop tack initially increased and then decreased with the addition of DMA from 0 to 4 wt%, and the maximum value appeared at 1 wt%. Peel strength reduced, while shear strength improved with increased DMA concentration.

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1. Introduction

Pressure sensitive adhesives (PSAs) are a class of materials that allow an instantaneous adhesion to a variety of surfaces within short contact time and low contact pressure without any phase transition or chemical reaction due to its specific viscoelastic properties [1]. PSAs can generally be classified into five categories based upon: natural rubber, thermoplastic elastomer, silicon, polyurethane (PU) and acrylic materials. Acrylic PSAs have a wide application field such as tapes, labels and protective films [2] due to their excellent adhesion properties, resistance to light, water and aging performance [3]. Although acrylic PSAs can be obtained by different polymerization processes (i.e., emulsion, solution, hot-melt, or radiation curing), much attention has recently been devoted to the use of more environmentally friendly processes, such as emulsion polymerization [4–6].

In traditional emulsion polymerization, a functional monomer is such that, when used in small proportions (generally in a concentration not greater than 10% of the total monomer mixture), it provides the base polymer with a chemical group which is capable

of modifying the physicochemical properties of the resulting latex (e.g., viscosity, stability), and/or of the polymer which is recovered from that latex (e.g., adhesion, softening point). Extensive research has been done on the synthesis of emulsion copolymers of well-known monomers (e.g., styrene, methyl methacrylate, butyl acrylate, etc.) with such functional monomers as carboxylic acids [7–13], and acrylamide derivatives [14–16]. Li et al. [17] studied the morphology of P(BA-St-DAAM) latex particles prepared by seeded-emulsion polymerization. They found that the more acrylamide monomer that was used, the more distinct the phase separation was. Volfova, et al. [15] prepared PS/PBA polymer dispersions with core/shell particles functionalized by N-methylol acrylamide (N-MA) through two-steps emulsion polymerization. They discovered that, in the presence of N-MA, the crosslinking reaction occurred during the polymerization. Films from functionalized dispersions exhibit improved tensile strength and higher resistance against organic solvents. These researchers dealt mainly with such problems as the effect of the functional monomers in the polymerization mechanism and kinetics [7,8,13–15], the location of functional groups in the systems (whether they were located in the aqueous phase, at the particle/water interface or within the particles) [7–10,12,14,15], and the effect of the functional monomers on the morphology of the particles [11,18].

Recently, polymers based on N,N-dimethylacrylamide (DMA)

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have attracted increasing interest because of their suitability for several practical applications such as drug-delivery hydrogels, medical diagnostics, polymeric supports for polypeptide synthesis, flocculation agents and catalytic reactions. Unlike polyacrylamide, poly(DMA) is soluble in water as well as in various organic solvents because it is less polar than polyacrylamide. Owing to this diverse solubility, DMA can be polymerized in various media, allowing a better understanding of the effect of the medium on the kinetic parameters and on the polymer properties. Valdebenito et al. [19] investigated the effect of solvent on the free radical polymerization of N,N-dimethylacrylamide. The results suggest that the main factor that controls the polymerization rate is a kinetic effect due to the hydrogen bonding between the amide carbonyl group and water molecules. However no study to date has examined the application of N,N-dimethylacrylamide (DMA) monomer in both emulsion polymerization and acrylic PSAs. The aim of the present work was to investigate systematically the effect of functional monomer DMA on the performance properties of the acrylic latex PSA.

In the present study, acrylic PSA latexes were synthesized by a monomer-starved seeded semi-continuous emulsion polymerization process. The influence of the acrylamide derivative monomer DMA on the viscosity and particle size of the latexes and the gel content, sol molecular weight (M_w , M_n), water resistance and adhesive properties (loop tack, peel strength and shear strength) of the PSA copolymers were investigated. In addition, infrared spectroscopy (IR), differential scanning calorimetry (DSC) and thermogravimetric analysis (TGA) were used to characterize the latex films.

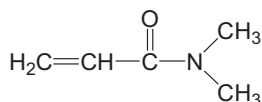
2. Experimental

2.1. Materials

Butyl acrylate (BA; technical grade, Shanghai Huayi Acrylic, China), methyl methacrylate (MMA; reagent grade, Shanghai Lingfeng Chemical, China), acrylic acid (AA; reagent grade, Shanghai Lingfeng Chemical, China), 2-hydroxy ethyl acrylate (HEA; technical grade, Shanghai Huayi Acrylic, China), N,N-dimethylacrylamide (DMA; technical grade, Energy Chemical, China), the initiator ammonium persulfate (APS; technical grade, Shanghai Aijian Modern Reagent Factory, China) and the buffer sodium bicarbonate (NaHCO_3 ; reagent grade, Shanghai Lingfeng Chemical, China). All these materials were used without further purification. The conventional ammonium nonyl phenol ethoxylate sulfate surfactant (Rhodapex CO-436), which contains 4 poly (ethylene oxide) (PEO) groups, was supplied by Shanghai Honesty Fine Chemical (China) and used as received. Distilled deionized water (DI- H_2O) was used throughout the study. Ammonia (25 wt% in H_2O) was obtained from Nanjing Chemical Reagent Co. The solvent used in the polymer characterization, tetrahydrofuran (THF, HPLC grade, Shanghai Lingfeng Chemical) was also used as supplied by the manufacturer. The molecular structure of DMA is shown in Scheme 1.

2.2. Emulsion polymerization

The recipes for water-based PSA latexes synthesized with



Scheme 1. The molecular structure of DMA.

Table 1
Emulsion polymerization pressure sensitive adhesive recipe.

Component	Amount (g)	
Monomers^a	BA	92–88
	MMA	5
	DMA	0–4
	AA	1
	HEA	2
CO-436	2.1	
APS	0.5	
NaHCO_3	0.2	
Deionized water	105	

^a The total monomer weight was set 100 g.

different contents of DMA are described in Table 1. 25 g deionized water and 2.0 g CO-436 were added to a 500 mL four-neck round-bottom flask and were stirred rapidly to aid dissolution of the emulsifier. The monomer mixtures were then slowly added into the water-emulsifier mixture through a constant pressure funnel over a period of 20 min. After that, the pre-emulsion was stirred for a further 30 min.

The polymerization was carried out by a monomer seeded semi-batch emulsion polymerization process in a 500 mL four-neck round-bottom flask equipped with an electromotive stirrer, thermometer, two separated addition funnels, and a reflux condenser. The stirring speed was maintained at 270 rpm throughout the experiments. First, a homogeneous aqueous solution containing 50 g DI- H_2O , 0.1 g CO-436, and 0.2 g NaHCO_3 was charged into the reactor. When the temperature reached 78 °C, a monomer mixture containing 5.69 g BA and 0.31 g MMA and an initiator solution containing 0.15 g APS and 5 g water were charged to the flask to form the seed latex. The temperature was then raised to 83 ± 2 °C and the seed polymerization was continued for an additional 30 min. Subsequently, the pre-emulsion and initiator solutions containing 0.35 g APS and 25 g water were added slowly to the reactor using two separate constant pressure funnels. The feeding times for the pre-emulsion and the initiator solutions were 3.5 and 4.0 h, respectively. After the feed was completed, the reaction was allowed to proceed for an additional 1 h to increase monomer conversion. The latex was then cooled to room temperature and poured into a glass bottle to be used for further characterization.

2.3. Characterization

2.3.1. 2IR spectrographic analysis

The latex was dried in a vacuum oven at 105 °C until it reached a constant weight. Infrared (IR) spectra of the dried latex films were recorded with a Bruker VERTEX80 IR spectrometer (Germany) in the range 4000–400 cm^{-1} .

2.3.2. Particle size and viscosity

Latex particle sizes were measured using a Dynamic Light Scattering (DLS) instrument (Malvern NanoS Zetasizer). The analyses were carried out at 25 °C, and every result was an average of three parallel measurements. The reported diameter is an intensity-weighted average particle size. Latex viscosity was measured using a NDJ-8S rotational viscometer (Shanghai Pingxuan, China) at 25 °C with 1 # spindle and 30 rpm speed.

2.3.3. Differential scanning calorimetry (DSC)

A differential scanning calorimeter model 204 F1 from NETZSCH Instruments was used to determine glass transition temperatures (T_g). 5–15 mg of dry polymer was weighed into a standard DSC hermetic alumina crucible. High-purity nitrogen was

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