

Water-Soluble/Dispersible Cationic Pressure-Sensitive Adhesives. II. Adhesives from Emulsion Polymerization

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ABSTRACT: In our previous work, we reported that cationic water-soluble pressure-sensitive adhesives (PSAs) could be synthesized in ethanol or methanol. These cationic water-soluble adhesives would not cause a stickies problem during paper recycling and can be easily removed from the papermaking system by adsorbing on wood fibers. In this study we report the synthesis and application of water-based cationic PSAs using miniemulsion polymerization. A redox initiator system of cumene hydroperoxide/tetraethylethylenepentamine was used to force interfacial polymerization. The end-use properties of the PSAs were evaluated, and the repulpability of the PSAs in paper recycling was studied. It was found that the cationic PSA from miniemulsion poly-

merization itself was insoluble and nondispersible in water during the paper recycling process. However, if this water-insoluble cationic PSA from miniemulsion was formulated with a water-soluble cationic PSA made from ethanol, the solubility or dispersability of the former PSA in water was improved. The molecular weight and degree of crosslinking of the PSA polymer have significant effects on the properties and dispersability of PSA. © 2003 Wiley Periodicals, Inc. *J Appl Polym Sci* 91: 347–353, 2004

Key words: addition polymerization; copolymerization; emulsion polymerization; adhesives; recycling

INTRODUCTION

Because of environmental concerns, an increasing amount of waste paper is recycled to supplement the use of virgin fiber in papermaking. The increased use of recycled fiber has been accompanied by an increasing level of contaminants, such as stickies, which are hydrophobic, tacky, polymeric materials that are introduced into the papermaking system from a mixture of recycled-fiber sources containing pressure-sensitive adhesives (PSAs). Stickies can break down and then reaggregate to foul various parts of the paper mill. When subjected to a number of factors including changes in pH, temperature, concentration, charge, and shear forces, stickies can accumulate on paper machine felt, wire, dryer, and other places. Stickies deposits can lead to decreased runability, decreased productivity, and increased machine downtime.²

Stickies are mainly generated from pressure-sensitive adhesive products such as postage stamps, self-sealing envelopes, and labels, for example. In recent years, there has been growing interest in developing paper recycling-friendly PSAs. In our previous

study,¹ we reported a new class of solvent-based cationic PSAs. There are several benefits from the use of cationic PSAs. First, they can be dissolved or dispersed in water by controlling the cationic charge density in the backbone of PSA; thus they will not deposit as stickies during paper recycling and papermaking processes. Second, they can be easily removed from the system by adsorbing onto fiber and fine surfaces because they are cationically charged. Third, the adsorbed colloidal or dissolved PSAs have little effect on paper properties. However, solvent-based PSA is not preferred because of economic and environmental concerns. It is therefore desirable to develop recycling-friendly water-based cationic PSAs.

Emulsion polymerization is widely used to produce water-based PSAs. Currently, commercial water-based PSAs are all negatively charged. Water-dispersible/soluble PSAs have been produced commercially by emulsion polymerization of acrylate and acrylic acid at low pH, which may become water-soluble/dispersible at high pH, such as in the paper recycling process. However, because the commercial water-soluble PSAs are negatively charged, they are unable to adsorb onto wood fibers (wood fibers are negatively charged in most cases). Thus the dissolved/dispersed PSA will accumulate in the paper recycling process water and cause stickies problems.

The copolymerization of hydrophobic and ionic hydrophilic monomers (such as cationic monomer) by emulsion polymerization has been a challenge because

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TABLE I
Basic Recipe

Name	Formula	Amount (g)
Butyl acrylate	$\text{H}_2\text{C}=\text{CHCO}_2(\text{CH}_2)_3\text{CH}_3$	75
MAETAC	$\text{H}_2\text{C}=\text{C}(\text{CH}_3)\text{CO}_2(\text{CH}_2)_2\text{N}(\text{CH}_3)_3\text{Cl}$	Variable
Triton X-405	$4-(\text{C}_8\text{H}_{17})\text{C}_6\text{H}_4-(\text{OCH}_2\text{CH}_2)_n\text{OH}$, $n = 40$	6.43
CHP	$\text{C}_6\text{H}_5\text{C}(\text{CH}_3)_2\text{OOH}$	1.0
TEPA	$\text{HN}(\text{CH}_2\text{CH}_2\text{NHCH}_2\text{CH}_2\text{NH}_2)_2$	1.0
Water	H_2O	115
EGDM	$[\text{H}_2\text{C}=\text{C}(\text{CH}_3)\text{CO}_2\text{CH}_2]_2$	Variable
1-Hexanethiol	$\text{CH}_3(\text{CH}_2)_5\text{SH}$	Variable
Hexadecane	$\text{CH}_3(\text{CH}_2)_{14}\text{CH}_3$	1.5

the ionic monomer resides almost exclusively in the aqueous phase, whereas the hydrophobic monomer resides almost exclusively in the organic phase. Copolymerization of monomers with very different water solubilities by emulsion techniques has been extensively studied.^{3–8} It has been found that the incorporation of water-soluble monomer into hydrophobic polymer backbone is very limited when emulsion polymerization is used, regardless of the initial amount of water-soluble monomer loaded into the system. Brouwer³ studied the emulsion polymerization of styrene (80–100 wt %) and [2-(methacryloyloxy)ethyl]trimethylammonium chloride (MAETAC, 0–20 wt %), and found that the maximum incorporation of MAETAC to the latex was less than 4 wt %. Kim et al.⁴ used a shot process to obtain a copolymer of styrene and sodium styrene sulfonate with a 6 wt % overall composition in a soap-free emulsion polymerization. Recently, Gilbert et al.⁹ proposed that selecting an appropriate initiator system such as cumene hydroperoxide/tetraethylenepentamine (CHP/TEPA) makes it possible to graft hydrophilic monomer onto a hydrophobic polymer in an emulsion process. In our previous work¹⁰ we reported that by using CHP/TEPA as an initiator, the hydrophobic CHP would meet the hydrophilic TEPA at the oil-water interface where radicals would be produced at just the location where hydrophobic and hydrophilic monomer are both present.

Recently, miniemulsion polymerization has been widely studied as a new polymerization process. In comparison with the traditional emulsion polymerization, in miniemulsion polymerization, monomer is pre-dispersed as 50- to 300-nm droplets in the water by applying high shear produced by a homogenizer, microfluidizer, or sonicator. A costabilizer such as hexadecane is used to prevent Ostwald ripening (diffusion of monomer from small droplets to large droplets to reduce the total interfacial energy), and a surfactant is used to prevent coalescence. Because the droplets are so small that they can effectively capture free radicals in water, droplet nucleation becomes dominant. As a result, the miniemulsion polymerization process is

much more robust than traditional emulsion polymerization based on particle nucleation from surfactant micelles. Conventional emulsion (macroemulsion) and miniemulsion polymerization of butyl acrylate with cationic monomer have been investigated.¹¹ In this study we report the development of a new class of novel cationic water-based PSAs, made by miniemulsion polymerization, that are water-dispersible during paper recycling.

EXPERIMENTAL

Materials

All reagents were purchased from Aldrich Chemical (Milwaukee, WI). Butyl acrylate (BA) and ethylene glycol dimethacrylate (EGDM) were purified by three washes with a 5% NaOH solution followed by three washes with DI water. [2-(Methacryloyloxy)ethyl]trimethylammonium chloride (MAETAC, 75 wt % in water) was extracted with ethyl ether three times to remove inhibitors, and the residual ethyl ether was removed in a rotary evaporator at room temperature under reduced pressure. All other reagents were used as received. Cumene hydroperoxide (CHP) and tetraethylenepentamine (TEPA) were used together as redox initiators. Triton X-405 (70 wt % in water) was used as surfactant. Hexadecane (HD) was used as costabilizer in the miniemulsion polymerization. 1-Hexanethiol (HT) was used as a chain transfer agent.

Synthesis

The basic recipe is shown in Table I. A solution of the recipe amounts of MAETAC, DI water, and Triton X-405 was mixed with a solution of the recipe amounts of BA, CHP, HD, and HT under moderate shear. The coarse emulsion was subjected to sonication for 15 min with a 300-W sonic dismembrator at 70% power. Bulk mixing during sonication was provided by a magnetic stirrer. The miniemulsion was added to the reactor and purged with nitrogen for a minimum of 45 min while the reactor temperature was kept at 40°C. The

polymerization was started by injecting the recipe amount of TEPA solution. The polymerization took 3 h. The BA conversion was around 90% and MAETAC conversion was around 92%. BA conversion was determined gravimetrically and MAETAC conversion was measured by NMR, as described in the literature.¹⁰

Characterization

The apparent particle size after polymerization was measured by light scattering with samples diluted with either DI water or filtered 1M NaBr solution. The 1M NaBr solution was used to collapse a hairy layer of poly(BA-co-MAETAC) around the particles to get a hard-sphere particle size. Gel fraction was determined by solvent extraction in a Soxhlet extractor. The dried sample was extracted with ethanol and then with THF for 24 h each. Glass-transition temperatures (T_g 's) were determined on a Perkin-Elmer Pyris-1 differential scanning calorimeter (DSC; Perkin Elmer Cetus Instruments, Norwalk, CT) under helium atmosphere. The sample was heated to 200°C, cooled to -100°C, and then heated to 200°C at the rate of 40°C/min. PSA end-use properties, its repulpability, and effect on paper properties were investigated as described in a previous study.¹

RESULTS AND DISCUSSION

Polymer synthesis and characterization

Generally only very limited amounts of water-soluble monomer can be incorporated into the polymer chain during emulsion polymerization. A special two-component redox initiator system, where one component is hydrophilic and the other is hydrophobic, has been

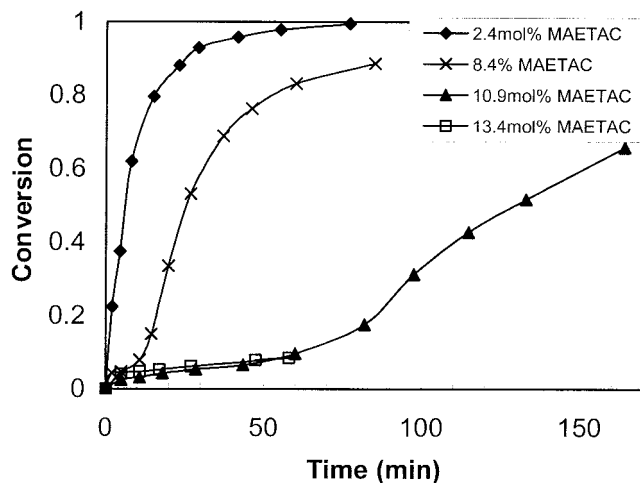


Figure 1 Butyl acrylate conversion in macroemulsion polymerization with varying levels of MAETAC.

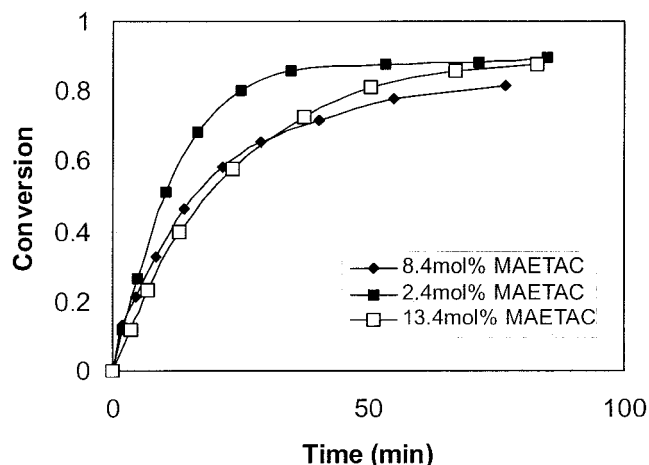


Figure 2 Butyl acrylate conversion in miniemulsion polymerization with varying levels of MAETAC (in feeding).

shown to be useful in promoting copolymerization under these conditions. In the current CHP/TEPA initiator system, CHP is dissolved in the BA monomer droplet, whereas the preponderance of the TEPA resides in the water phase. CHP and TEPA molecules react at the interface of BA droplets and the water phase to form free radicals, and then polymerization is initiated at the interface. We attempted to synthesize poly(BA-co-MAETAC) by macroemulsion and miniemulsion techniques. For macroemulsion polymerization, no hexadecane was added. Figures 1 and 2 show the BA polymerization kinetics. It can be seen that for macroemulsion polymerization, at high MAETAC levels, there is an induction period for BA polymerization. The higher the MAETAC fraction in the feed, the longer the induction period. During the induction period, the MAETAC polymerized in the water phase and formed homopolymer. There was no induction period for BA polymerization in miniemulsion polymerization. This phenomenon is attributed to high levels of aqueous-phase polymerization in the macroemulsion (resulting from the high rate of generation of radicals in the high interfacial area of the micelles), which retards particle nucleation.¹¹ Thus the miniemulsion polymerization technique was used in this study for synthesizing copolymers with high cationic content.

The resulting polymer latex was characterized by dynamic light scattering (DLS), and the results are shown in Figure 3. For copolymers derived from a monomer mix containing with 2.4 mol % MAETAC, the polymer latex particle sizes in deionized water and in 1M NaBr solution are almost the same. With increased MAETAC fraction, the particle size difference in DI water and in 1M NaBr solution increased significantly. For example, in pure water, for copolymer with 15.6 mol % MAETAC, the "apparent average diameter" of the particles was about 600 nm. How-

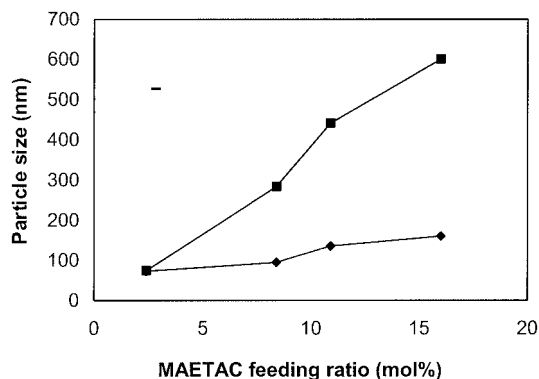


Figure 3 Latex particle size with varying MAETAC recipe levels in DI water (diamonds) and 1M NaBr salt solution (squares).

ever, in the 1M NaBr solution, the diameter would collapse into 160 nm. These results indicated hydrophilic MAETAC units were incorporated into the latex. With increased MAETAC fraction, the content of MAETAC in the particles increases, resulting in highly water swollen particles. Attempts to separate the particles from the serum to analyze the serum for polyelectrolytes were unsuccessful.

The glass-transition temperature of the polymer was measured by DSC, and the results are shown in Figure 4. Apparently all of the copolymers with different cationic monomer fractions have T_g 's around -48°C . By contrast, the T_g for BA homopolymer is around -56°C . These results may indicate that the polymer derived from recipes with high MAETAC fraction in the monomer mix were heterogeneous. The T_g of homopoly-MAETAC was not measured.

The reaction kinetics of the MAETAC were also studied. From Figure 5 it can be seen that for copolymers with 8.4 mol % MAETAC in the monomer mix, 18% of the total MAETAC quickly polymerized in water to form homopolymer, and the rest gradually polymerized to form the polymer latex.

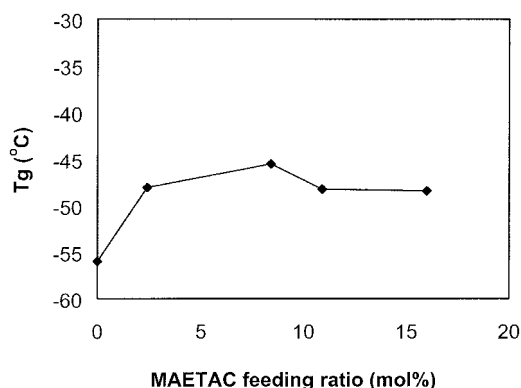


Figure 4 Glass-transition temperature (T_g) for polymers with varying levels of MAETAC (in feeding).

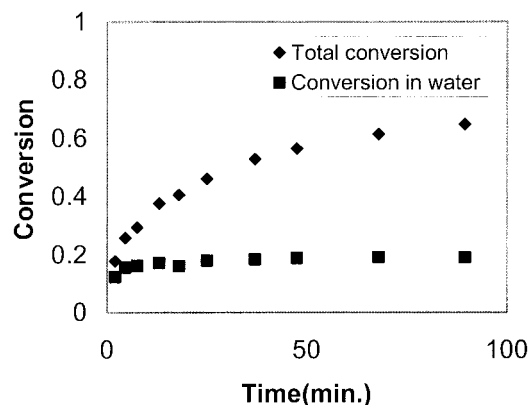


Figure 5 MAETAC conversion in miniemulsion polymerization with 8.4 mol % of MAETAC.

PSA properties and repulpability

This study was intended to develop water-based water-soluble/dispersible cationic PSAs that do not cause stickies problems in paper recycling. For ionic polyacrylate copolymer to be water soluble or dispersible, the copolymer must contain enough hydrophilic units to interact with water and then break the intermolecular bonding. However, the high cationic content may affect the end-use properties of the copolymers as PSAs. The peel adhesion and shear strength of the polymers synthesized from miniemulsion polymerization are shown in Table II. Peel adhesion is the force required to remove a PSA-coated film from a specific test surface under standard conditions (specific angle and rate). The measurement of peel adhesion involves a bonding step and a debonding (or peeling) step. The efficiency of the bonding process is related to the adhesive's ability to exhibit viscous flow. The debonding process involves a rapid deformation of the adhesive mass. Thus, the higher the peel strength, the higher the PSA's ability to resist bond deformation at high strain rates. Peel strength gives a measure of adhesive or cohesive strength, depending on the mode of failure.¹² Shear resistance is measured as the force necessary to pull the PSA material parallel to the surface to which it was affixed with a definite pressure¹³; it measures the cohesion strength of the PSA. All the copolymers (PSA1-PSA3) without chain transfer agent showed very low shear and peel strength. The reason may be that the molecular weight (MW) of the polymer was too high. Satas¹⁴ reported that shear is roughly proportional to molecular weight, up to relatively high MW at which the shear resistance drops off dramatically in some polymers. Peel adhesion typically exhibits a discontinuous behavior, increasing with MW up to moderate MW and then decreasing as the MW further increases. The MW for water-based polyacrylic PSA polymer is normally in the range of 300,000 to 1 million.¹⁵ Generally, the MW

TABLE II
Adhesive Properties of Cationic PSAs

Sample	Feeding composition for variables (mol %)			180° Peel adhesion (10 min dwell) (PSTC-1)		Shear strength (PTSC-7)	
	MAETAC	HT	EGDM	(g/in.)	Failure type ^a	(h)	Failure type ^a
PSA1	8.4	0	0	40.8	A	0	A
PSA2	10.9	0	0	25.3	A	0	A
PSA3	16.0	0	0	10.6	A	0	A
PSA4	10.9	0.1	0	560.5	C	0.1	C
PSA5	10.9	0.14	0	425.3	C	0.1	C
PSA6	10.9	0.1	0.18	339.7	A	6.8	A
PSA7	10.9	0.14	0.18	456.8	A	11.7	A
PSA8	10.9	0.14	0.36	234.2	A	6.0	A
PSA9	16.0	0.13	0.34	180.3	A	0.3	A

^a A, adhesion failure; C, cohesion failure.

for polymer synthesized from emulsion polymerization would be about 1 million. Furthermore, because the copolymer of BA and MAETAC is heterogeneous, the MAETAC-rich portion as well as the MAETAC homopolymer in water may crystallize during drying. The crystals may restrict the molecular flow of the PSA on a substrate surface, thus significantly reducing the adhesion.

The addition of chain transfer agent (CTA) in polymerization significantly increased the peel strength of the PSA (PSA4 and PSA5), but the shear adhesion was still poor. It should be noted that failure of the two PSAs with chain transfer agent added was cohesive failure. The results indicate that PSA bonding to the substrate was stronger than the intermolecular bonding. The reason is that the molecular weight of the PSA polymer is low. The molecular weight of the polymers with chain transfer agent can be estimated based on the Mayo equation¹⁶:

$$\frac{1}{DP_n} = \frac{1}{DP_{n,0}} + C \frac{[CTA]}{[M]}$$

where DP_n and $DP_{n,0}$ are the number-average degree of polymerization with and without chain transfer agent, respectively; C is the chain transfer constant of the CTA; $[CTA]$ is the concentration of the chain transfer agent; and $[M]$ is the concentration of the monomer. Because 1-hexanethiol as transfer agent would stay in the oil phase with the butyl acrylate, the chain transfer would mostly occur with butyl acrylate. The chain transfer constant of 1-hexanethiol for butyl acrylate may be estimated as 0.50.^{17,18} For PSA4, the concentration ratio of 1-hexanethiol to BA is 0.11%. Thus the molecular weight of the polymer with chain transfer agent would be about 189,000. PSA polymer with low molecular weight can flow easily and better wet out the substrate. The result is better bonding between the PSA and the substrate. However, as the

viscous flow ability increases, the cohesion strength of the polymer decreases.

Introducing a small amount of crosslinker into the polymer can increase the polymer cohesive strength, thus increasing the shear strength. The mode of failure under shear becomes adhesive failure for the crosslinked polymers. The effect of crosslinker on peel strength is insignificant. However, a crosslinker content that is too high can significantly limit the ability of the polymer to diffuse on a substrate, thus reducing the peel strength.

From previous work¹¹ we know that the polymer latex resulting from high MAETAC fraction in the recipe has a core-shell structure; we had expected the water-soluble shell to help the PSA film to redisperse in water. However, solubility tests indicated that none of the polymers synthesized by miniemulsion polymerization could redisperse into water once they are dried. A repulping test showed that none of the cationic PSAs was repulpable, even when they formed very thin (<25 μm thickness) PSA film on copy paper. Large sticky particles can clearly be seen from the handsheets made from the recycled pulp. The results indicate that when dry PSA film was formed from the latex emulsion, the water-insoluble MAETAC-poor core of the latex particles coalesced together. During the repulping process, the coalescent film could not be redispersed into colloidal particles.

To increase the redispersability of the cationic PSA, three water-soluble cationic chemicals were added to the emulsion. These included the surfactant cetyltrimethylammonium bromide (CTAB), starch, and water-soluble poly(BA-co-MAPTAC) (BAM) with MAPTAC content of 15.2 mol %. This poly(BA-co-MAPTAC) polymer was made from polymerization in ethanol and itself can be used as a water-soluble cationic PSA as reported in our previous research.¹ The repulpability of the newly formulated PSA was then studied. It was found that neither CTAB nor starch could make

TABLE III
Repulpability of Formulated Cationic PSAs

PSA	PSA composition (wt %)			Repulpability
	Emulsion	BAM	Starch	
PSA10	96 (PSA4)	4	0	No
PSA11	96 (PSA5)	4	0	No
PSA12	96 (PSA6)	4	0	Repulpable
PSA13	96 (PSA6)	3	1	Repulpable
PSA14	96 (PSA7)	4	0	Repulpable
PSA15	96 (PSA7)	3	1	Repulpable

the cationic PSAs redispersible at the 4 wt % addition level. Furthermore, CTAB significantly reduced the tackiness of the PSA, and the starch caused the viscosity of the PSA latex to rise to unacceptable levels. The repulpability of these materials is reported in Table III. However, it was found that the water-insoluble PSA made by miniemulsion polymerization could be made fully water redispersible when it was blended with a cationic PSA by solution polymerization in ethanol (BAM). It was also found that the addition of crosslinker in the PSA latex rendered the PSA repulpable.

For PSA latex without crosslinker, the molecules in the latex may move easily to form an evenly coalescent layer during drying. The addition of poly(BA-*co*-MAPTAC) will not enable the dried PSA film to dissociate in repulping water during the paper recycling process. Conversely, in a PSA latex with crosslinker, the crosslinker can bind the molecules in the latex together. The coalescence of the latices during drying would not be even. With poly(BA-*co*-MAPTAC) added, the BA units, which are hydrophobic, would attach to the hydrophilic units in the PSA latex; and the MAPTAC units, which are hydrophobic, would orient toward the aqueous phase on the latex surface. During drying, poly(BA-*co*-MAPTAC) would form a barrier around the latex particles as a lubricant. In the paper recycling process under high shear, the PSA would easily redisperse into microparticles. The addition of a small amount of starch in some was not aimed to improve the PSA dispersability, but rather to improve the properties of paper made from the recycling process.

To further understand the crosslinking in the polymer latex, the gel content was measured. Ethanol was used as the extraction solvent to remove polymer with high MAETAC content. Tetrahydrofuran (THF) was used to remove polymer with low MAETAC content. From Table IV, it may be seen that most non-crosslinked polymer was removed by the ethanol. This is not surprising because the crosslinker EDGM is hydrophobic, and thus would mainly crosslink inside the latex with butyl acrylate. The crosslinked hydrophobic core of the polymer latex would retard the coalescence during drying.

TABLE IV
Gel Content in the Crosslinked Polymers

Sample	HT (mol %)	EGDM (mol %)	Gel content after solvent extraction	
			Ethanol	Ethanol and THF
PSA6	0.11	0.2	0.736	0.671
PSA7	0.16	0.2	0.708	0.648
PSA8	0.16	0.4	0.769	0.702

The PSA properties of the repulpable (dispersible) PSAs were tested and results are shown in Table V. It was found that the addition of poly(BA-*co*-MAPTAC) had no significant effect on the PSA properties.

PSA adsorption and effect on paper properties

In our previous study¹ we reported that the cationic PSA from solution polymerization could adsorb on fiber surfaces in paper recycling and papermaking. In this study, the adsorption of water-based cationic PSAs made from miniemulsion polymerization is reported. Copy papers with and without 0.5 wt % of cationic PSA12 were repulped. Pulping water was collected. After 0.5 h, the turbidity and charge of the supernatant of the pulping water were measured. No detectable PSA12 was found in the pulping water, which means almost all of the cationic PSA microparticles adsorbed on the fibers.

Because the cationic PSA microparticles would adsorb on fibers, and be brought into the final paper products during the papermaking process, the effect of the PSA on paper properties should be examined. Figure 6 shows the effect of cationic PSA12–PSA15 on the paper tensile strength. It was found that the cationic PSA slightly increased the paper strength. With starch added to the PSA, the increase in the paper strength was greater. Starch is widely used as a strengthening agent in papermaking. The small amount of starch in the PSA should contribute to the strength improvement. Sizing is a measure of the hydrophobicity of the paper. Because the cationic PSA is partly hydrophobic, it may have some effect on the

TABLE V
Adhesive Properties of Water-Dispersible Cationic PSAs

Sample	180° Peel adhesion (10 min dwell) (PSTC-1)		Shear strength (PTSC-7)	
	(g/in.)	Failure type	(h)	Failure type
PSA6	339.7	A	6.8	A
PSA12	376.8	A	10.3	A
PSA13	354.2	A	7.7	A
PSA7	456.8	A	11.7	A
PSA14	480.3	A	15.3	A
PSA15	463.5	A	12.6	A

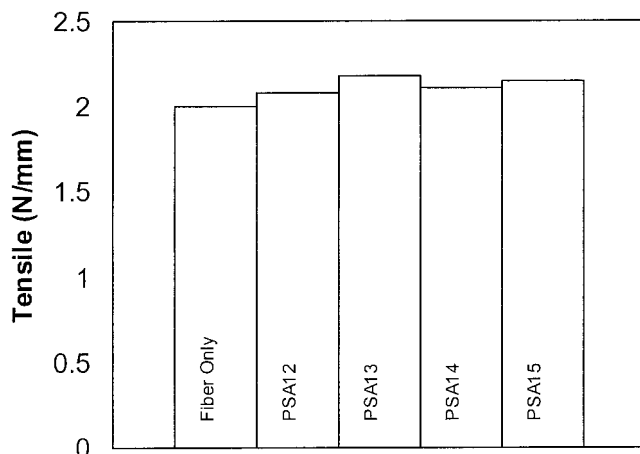


Figure 6 Effect of 0.5% cationic PSA on the tensile strength of paper.

paper sizing properties. The sizing effect of PSA was measure by Hercules sizing test (HST). It was found that with 0.5% addition level of cationic PSA in the paper, the HST increased only marginally. This effect is negligible.

CONCLUSIONS

Water-based cationic PSAs with various compositions were synthesized by miniemulsion polymerization. It was found PSA latex with a high fraction of cationic monomer (MAETAC) in the recipe was very heterogeneous. The cationic PSA itself could not be redispersed in the paper recycling process. However, when the cationic PSA was formulated with cationic water-soluble polymer made by solution polymerization, the PSA was fully dispersible, and did not form stickies during the repulping and papermaking processes. The dispersed PSA could easily be removed from the pa-

permaking water system by adsorbing it onto negatively charged fiber surfaces. Furthermore, the adsorbed colloidal PSA improved the paper strength of the final paper product. Thus, water-based cationic dispersible PSAs, which are more compatible with the paper recycling process, were successfully developed.

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