Pressure sensitive adhesives of acrylic polymers containing functional monomers

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Pressure sensitive adhesives possessing good cohesive strength have been prepared by copolymerizing n-butyl acrylate with various functional monomers. The effect of the polar nature and of the concentration of comonomer on tack, adhesion and cohesion was studied. The cohesive strength of the adhesive was determined on various surfaces. A comparative study was made to assess the effectiveness of a particular comonomer on the basis of the results obtained.

Keywords Alkyl acrylate; hydroxy alkylacrylate; cohesion; adhesion; tack and performance index

INTRODUCTION

The use of homopolymers containing long-chain alkyl acrylates for pressure-sensitive adhesive (PSA) applications has long been recognized. Although these polymers are quite tacky, they lack the necessary cohesive strength. One of the ways to enhance the cohesive integrity is to modify long-chain alkyl acrylate monomers with various short alkyl acrylate monomers[1]. Recently Heilman *et al.*[2] have shown that the cohesive strength can be increased by copolymerizing iso-octyl acrylate with various polar functional monomers.

To optimize the adhesive performance characteristics of the tape, we attempted to incorporate monomers possessing functional groups into poly(n-butyl acrylate) or (n-butyl acrylate–acrylic acid) copolymer. With this objective in mind, the co- and ter-polymerizations of n-butyl acrylate with 2-hydroxypropyl methacrylate, 2-hydroxyethyl acrylate, 2-hydroxyethyl methacrylate and acrylic acid were carried out to evaluate the tack, adhesive and cohesive characteristics of the products.

EXPERIMENTAL

Materials

n-Butyl acrylate (fluka) was purified by vacuum distillation, while other comonomers were all commercial samples (fluka) and used without further purification. The comonomers used were (1) 2-hydroxy ethyl methacrylate (HEMA), (2) 2-hydroxy propyl methacrylate (HPMA), (3) 2-hydroxy ethyl acrylate (HEA) and (4) acrylic acid (AA).

Copolymerization

Copolymerizations were carried out in solution using ethyl acetate as the solvent and benzoyl peroxide as the free radical initiator at 60°C[3]. The copolymerization reactions were carried out until 95% conversion (theoretical) was achieved.

Tape tests

Pressure sensitive adhesive (PSA) are characterized by fulfilling the following basic requirements, which are referred to as balanced properties[2,4].

(i) the ability to resist shearing loading (cohesion).

(ii) the ability of the bonds to sustain high peeling load (adhesion).

(iii) stretchiness and elasticity (tack).

These three properties were studied by tape evaluation, where the copolymers were coated to polyester film to a thickness of 0.3 mm and dried in an oven at 88°C for 30 min to ensure the removal of all solvents. The various evaluation methods are described below.

Tape cohesion test

This was carried out using a standard shear strength test method[5], where a 500 gram load was suspended from a 14.6 cm^2 contact area. The time required for the tape to be separated from the steel plate was measured and recorded in minutes as the average of 4 trials.

Tape adhesion test

The 180° peel adhesion was carried out using an 'INSTRON 3710-016' apparatus at a cross head speed of 500 mm min^{-1}.

Glass surface was used as the substrate instead of stainless steel. The data reported are the average of two trials.

Tape tack test

The tape tack test was carried out by employing the standard procedure[6], the contact area of the stainless steel probe being 0.02 cm^2 and the withdrawing velocity being 1 cm s^{-1}. The data given are the average of 15 trials.

Performance index (*PI*)

'Balance of properties' of a PSA implies the high value desired for each of the experimentally determined parameters, where a quantitative assessment is required to evaluate the effectiveness of a particular comonomer as a function of its concentration. The concept of PI is to make such comparison. The PI concept is very empirical and is computed in the following manner.

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$$PI = \left[\begin{matrix}\text{Copolymer}\\ \text{Adhesion}\end{matrix} - \begin{matrix}\text{BA}\\ \text{homopolymer}\\ \text{Adhesive}\end{matrix}\right] + \left[\begin{matrix}\text{Copolymer}\\ \text{Tack}\end{matrix} - \begin{matrix}\text{BA}\\ \text{homopolymer}\\ \text{Tack}\end{matrix}\right] + \left[\begin{matrix}\text{Copolymer}\\ \text{Cohesion}\end{matrix} - \begin{matrix}\text{BA}\\ \text{homopolymer}\\ \text{Cohesion}\end{matrix}\right]$$

It should be noted that PI represents a dimensionless value, as adhesion and tack are measured in grams while cohesion is measured in minutes.

RESULTS AND DISCUSSION

Effect of various polar comonomers

The results of the tape study are summarized in *Table 1*. From the data it was apparent that n-butyl acrylate–acrylic acid copolymer showed greater cohesive and adhesive strength than poly(n-butyl acrylate). This may be due to intermolecular dipole–dipole interactions due to –COOH groups. Further terpolymers monomers showed n-butyl acrylate–acrylic acid-hydroxy functional monomers showed excellent properties indicating the significance of the hydroxyl groups and their possible hydrogen bonding with COOH groups. The high values for HEA containing terpolymer with a lesser concentration of HEA is attributed to the stronger interchain hydrogen bonding due to the absence of α-methyl group. The poor PI for the HPMA containing terpolymer may be related to the weak intermolecular interactions of secondary hydroxyl groups.

Since terpolymers showed higher PI than the copolymers, i.e. BA–AA and BA–HPMA, it may be said that the increase in cohesion and adhesion in the former is due to preferential intermolecular hydrogen bonding between the –OH of hydroxy functional monomer and COOH or AA.

Effect of HPMA concentration on PI of BA–AA–HPMA terpolymer

The results of the cohesion, adhesion and tack with respect to increase in concentration of HPMA are listed in *Table 2*. Increase in HPMA concentration increases the adhesive strength. A close examination of the cohesive strength reveals that the strength is maximum at a particular concentration of HPMA (0.0014 mols) and beyond that the cohesive strength drops. This may be related to the predominance of intramolecular hydrogen bonding between terminal-OH and –COOH groups over intermolecular interactions. Tack values did not show marked variation with respect to HPMA concentration.

Effect of AA concentration on PI of BA–AA–HPMA terpolymer

The results of the tape study of these polymers are given in *Table 3*. Examination of the data reveals an increase in the cohesive force with increase in proportion of AA up to 0.0083 mols. Further increase in AA concentration results in a significant drop in cohesive strength.

These observations indicate that it is necessary to choose proper concentrations of AA and HPMA to attain the optimum balance of properties desired for a PSA.

The polymers cured with pyrromellitic dianhydride showed significant improvement in the tape performance. Detailed investigations on the cohesive, adhesive and tack measurements of polymers cured with various cross linking agents are in progress.

Table 1 Effect of various comonomers on tape performance of PSA

Polymer composition	Cohesive failure (minutes)	Adhesive failure (load in grams)	Tack (grams)	PI
1 BA(.078)	25	700	63	–
2 BA(.076) + AA(.004)	59	1400	78	749
3 BA(0.076) + HPMA(.002)	79	1100	76	467
4 BA(.074) + AA(.004) + HPMA(.0013)	125	2000	106	1433
5 BA(.074) + AA(.004) + HEMA(.0015)	115	3500	110	2937
6 BA(.075) + AA(.004) + HEA(.0004)	300	1900	105	1547

Table 2 Effect of concentration of HPMA on tape performance

Polymer composition	Cohesive failure (minutes)	Adhesive failure (grams)	Tack (grams)	PI
1 BA(0.075) + AA(.004) + HPMA(.007)	87	1600	95	994
2 BA(.074) + AA(.004) + HPMA(.0014)	125	2000	106	1443
3 BA(.073) + AA(0.004) + HPMA(.002)	100	2150	112	1574
4 BA(.072) + AA(.004) + HPMA(.003)	96	3200	116	2586
5 BA(.071) + AA(.004) + HPMA(.004)	31	3900	114	3257

Table 3 Effect of AA concentration on tape performance

Polymer composition	Cohesive failure (minutes)	Adhesive failure (grams)	Tack (gram)	PI
1 BA(.074) + AA(.004) + HPMA(.0014)	125	2000	106	1443
2 BA(.072) + AA(.008) + HPMA(.0014)	142	2900	113	2367
3 BA(.070) + AA(0.012) + HPMA(.0014)	102	3200	115	2629

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