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**Pressure Sensitive
Adhesives**

Formulations and Technology

Second Edition

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PRESSURE SENSITIVE ADHESIVES

Formulations and Technology

Second Edition

Henry R. Dunning

NOYES DATA CORPORATION

Park Ridge, New Jersey, U.S.A.

1977

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FOREWORD

The detailed, descriptive information in this book is based on U.S. patents issued since 1970 that deal with pressure sensitive adhesives.

This book serves a double purpose in that it supplies detailed technical information and can be used as a guide to the U.S. patent literature in this field. By indicating all the information that is significant, and eliminating legal jargon and juristic phraseology, this book presents an advanced, technically oriented review of pressure sensitive adhesives.

The U.S. patent literature is the largest and most comprehensive collection of technical information in the world. There is more practical, commercial, timely process information assembled here than is available from any other source. The technical information obtained from a patent is extremely reliable and comprehensive; sufficient information must be included to avoid rejection for "insufficient disclosure." These patents include practically all of those issued on the subject in the United States during the period under review; there has been no bias in the selection of patents for inclusion.

The patent literature covers a substantial amount of information not available in the journal literature. The patent literature is a prime source of basic commercially useful information. This information is overlooked by those who rely primarily on the periodical journal literature. It is realized that there is a lag between a patent application on a new process development and the granting of a patent, but it is felt that this may roughly parallel or even anticipate the lag in putting that development into commercial practice.

Many of these patents are being utilized commercially. Whether used or not, they offer opportunities for technological transfer. Also, a major purpose of this book is to describe the number of technical possibilities available, which may open up profitable areas of research and development. The information contained in this book will allow you to establish a sound background before launching into research in this field.

Advanced composition and production methods developed by Noyes Data are employed to bring our durably bound books to you in a minimum of time. Special techniques are used to close the gap between "manuscript" and "completed book." Industrial technology is progressing so rapidly that time-honored, conventional typesetting, binding and shipping methods are no longer suitable. We have bypassed the delays in the conventional book publishing cycle and provide the user with an effective and convenient means of reviewing up-to-date information in depth.

The Table of Contents is organized in such a way as to serve as a subject index. Other indexes by company, inventor and patent number help in providing easy access to the information contained in this book.

15 Reasons Why the U.S. Patent Office Literature Is Important to You —

1. The U.S. patent literature is the largest and most comprehensive collection of technical information in the world. There is more practical commercial process information assembled here than is available from any other source.
2. The technical information obtained from the patent literature is extremely comprehensive; sufficient information must be included to avoid rejection for "insufficient disclosure."
3. The patent literature is a prime source of basic commercially utilizable information. This information is overlooked by those who rely primarily on the periodical journal literature.
4. An important feature of the patent literature is that it can serve to avoid duplication of research and development.
5. Patents, unlike periodical literature, are bound by definition to contain new information, data and ideas.
6. It can serve as a source of new ideas in a different but related field, and may be outside the patent protection offered the original invention.
7. Since claims are narrowly defined, much valuable information is included that may be outside the legal protection afforded by the claims.
8. Patents discuss the difficulties associated with previous research, development or production techniques, and offer a specific method of overcoming problems. This gives clues to current process information that has not been published in periodicals or books.
9. Can aid in process design by providing a selection of alternate techniques. A powerful research and engineering tool.
10. Obtain licenses — many U.S. chemical patents have not been developed commercially.
11. Patents provide an excellent starting point for the next investigator.
12. Frequently, innovations derived from research are first disclosed in the patent literature, prior to coverage in the periodical literature.
13. Patents offer a most valuable method of keeping abreast of latest technologies, serving an individual's own "current awareness" program.
14. Copies of U.S. patents are easily obtained from the U.S. Patent Office at 50¢ a copy.
15. It is a creative source of ideas for those with imagination.

CONTENTS AND SUBJECT INDEX

INTRODUCTION	1
ACRYLICS	3
Copolymer Composition	3
Emulsion Polymerization of Latently Crosslinkable Polymers.	3
Epoxy Substituted Acrylics and Diketene.	5
Copolymers of Fumaric Acid Diesters and Vinyl Compounds.	7
Cyanoacrylates	13
Heterocyclic N-Vinyl Compounds.	14
Hydrolysis of Maleic Anhydride Copolymers	17
Acrylic Acid and Hydroxylated Amide Interpolymers	20
Carboxy Functional Polymers and Polyvalent Metals	21
Aqueous Alkali Removable Adhesives.	23
Polymer-Chelated Titanium Ester Reaction Products	27
Unsaturated Zwitterion Monomers	30
Vinyl Carboxylic Acid-Aminoalkyl Acrylate-Alkyl Acrylate Terpolymer ..	32
Maleic Anhydride-Alkyl Acrylate-Ethylene-Vinyl Acetate Polymer	36
Solvent-Free Adhesive Which Is Melttable and Soluble in Organic Solvents.	38
Pressure Sensitive Adhesive for Velvet-Type Fasteners	40
Carboxybetaine and Sulfobetaine Copolymers	42
Quaternary or Betaine Acrylics Copolymerized with Alkyl Acrylates.	43
Acrylate-Vinyl Carboxylic Acid Copolymer Neutralized with Alkanolamine	45
Alkyl Acrylate-Emulsifier Latex	46
Polymers of Carbocyclic Alkyl Acrylates and Vinyl Esters	47
Copolymerizing with Tertiary Amine and Organometallic Salt	49
2-Acrylamido-2-Methylpropanesulfonic Acid Interpolymers.	51
Acrylate-Oxypropylacrylamide-Acrylic Acid Interpolymers	53
Anaerobics	56
Acryloyloxyalkyl Acrylates.	56
Thermoplastic Polymer Plasticized with Anaerobic Adhesive	56
High-Molecular Thermoplastic Polymer.	60

Organic Nitrone Stabilizers	62
Additives.	63
Fatty Alkyl Amines to Improve Adhesion.	63
Amine Plasticity Modifiers	65
Hydrolyzed Gluten and Acrylates.	67
Phenol-Formaldehyde to Improve Thermal Stability.	68
Polyfunctional Aziridines	70
Carboxylated Monoether of Polyglycol.	72
Crosslinking Techniques	74
Isocyanate Prepolymers.	74
Polymerizable Isocyanates.	76
Polyfunctional Amines	79
3-Methacryloxypropyltrimethoxysilane	81
Electron Beam Curing.	83
Irradiation Treatment of Hot Melts.	85
Irradiation of Monomer Layer.	86
Ionizing Radiation.	90
Acid-Epoxy Self-Curing Interpolymer.	92
Other Processes.	95
Flame Retardant.	95
Aqueous Emulsion Adhesive for Vinyls.	97
Impregnation of Nonwoven Fabric Base	99
ELASTOMERS	101
Polymer Compositions	101
Low Styrene Butadiene-Styrene Copolymers	101
Styrene-Isoprene Block Copolymers	103
ABA Block Copolymer and Poly(α -Methylstyrene).	105
Polypentenomers.	108
Branched Rubbery Block Copolymer	110
Blend of Amorphous Polyolefin, Rubbery Block Polymer, Polybutylene, Polyisobutylene and Crystalline Polypropylene.	113
Styrene-Butadiene, Styrene-Isoprene and Isoprene-Piperylene Copolymers.	115
Polydodecene	117
α -Olefin Copolymers	120
Isotactic Polyolefins	122
Cyclic Monoolefin Copolymers	125
Carboxyl-Containing Chloroprene Polymer.	127
Water-Soluble Polymers from Epoxidized cis-1,4-Polybutadiene.	129
Acid-Grafted Polyolefins.	132
Sulfo and Thiouronium Derivatives.	133
Ethylene-Vinyl Acetate-Diolefin-Ester Copolymer	135
Polyisocyanate Modified Rubber.	137
Functionally Terminated Polybutadiene-Isocyanate Reactions.	140
Isocyanates and Hydroxylated Styrene-Butadiene Copolymers.	145
Tackifiers	147
Alkylated Polystyrene Resins	147
Piperylene-2-Methyl-2-Butene Copolymers	148
Styrene-Isobutene- β -Pinene Terpolymer	150
Hydrogenated Petroleum Fraction	151
Blend of Solid and Liquid Tackifiers.	153
Low-Molecular Hydrocarbon Resin or Liquid Natural Ester Resin	154

Rosin and Polyterpene Resins	156
Rosin Ester and Olefin-Diolefin Copolymer	157
Zinc Resinates of Disproportionated Rosin	160
Phenol-Diene-Phenol Resin Adduct	162
Tackifier Containing Carboxy, Hydroxy or Hydroxymethyl Groups	166
Alkylated Melamine-Aldehyde Resin	169
Polyalkene Tackifier Modifier	170
Other Additives and Curing Techniques	172
Acid-Modified Resin	172
Hydroxycarboxylic Acid or Ester as Thixotropic Agent	175
Naphthenic Process Oil Plasticizer	177
Finely Divided Silica	178
Aminoorganosilane	180
Phenol-Formaldehyde Curing Agents	181
Brominated Phenol-Formaldehyde and Zinc Resinate	185
Alkaline-Catalyzed Phenol-Formaldehyde Resin	186
OTHER PRESSURE SENSITIVE POLYMERS	189
Polyvinyl Ethers	189
Moisture-Vapor-Permeable Sheet	189
Transparent Water-Soluble Adhesive	191
Silicones	195
Curable Polyorganosiloxane	195
Alkylarylsiloxane or Alkylarylalkylsiloxane Extender	197
Polyurethanes	198
Hydroxylated Elastomer-Polyurethane Reaction Products	198
In Situ Polymerization and Crosslinking with Polyamines	199
Adhering with Polyesters and Polyisocyanates	200
Highly Branched Capped Polyurethanes	202
Polyether-Polyol and Diisocyanatodiurethane	202
Bonding Polyurethanes and Polyethylene Oxide	203
Self-Adhesive Coat	204
3-(Isocyanatomethyl)-3,5,5-Trimethylcyclohexyl Isocyanate	206
Moisture-Resistant Polyurethanes	208
Polyesters	210
Soft Thermoplastic Segmented Copolyesters	210
Adhesive from Waste Polyethylene Terephthalate	210
Other Polymer Materials	212
Polyvinyl Alcohol and Metal Curing Agent	212
Adducts of N-Substituted Aziridines and Maleic Anhydride	214
High-Temperature Bis(Epoxyalkyl)Carborane Adhesives	216
Aromatic Polysulfones	218
Polyamide Resin	221
Epihalohydrin Polymer and Polyketone	222
Isethionic Acid Ester Matrix Builder	224
Polythioethers	226
Aldehyde-Phenolic Condensation Copolymer	227
Lignin-Sulfonate Extender for Urea-Formaldehyde Resin	229
Ethers of N-Methylolamide	231
WEB AND BACKING CONSTRUCTION AND RELEASE COATINGS	233
Backing Construction	233
Nitrous Oxide Treatment of Polypropylene	233

Longitudinal Stretching of Polyolefinic Sheet	234
Tape Substrate with Increased Splitting Resistance	235
Extensible and Elastic Backing of ABA Block Copolymers	236
Flocked Covering	238
Transfer-Proof Ink on Web	238
Cellulose Acetate Butyrate and Copolyester and/or Acrylate Copolymer	241
Continuous Self-Sealing Webs for Forming Booklets	242
Polyurethane Foam Backing with Fluoroaliphatic Stabilizer	242
Heterocyanoacrylate Ultraviolet Absorbers	245
Silicone Release	246
Polypropylene Liner and Polysiloxane Coating	246
Hydroxy-Terminated Polysiloxane Formulations	249
Hydroxy-Terminated Polysiloxanes, Titanates and Organic Polymers	252
Polysiloxane and Alkyl Vinyl Ether-Maleic Anhydride Copolymers	253
Reaction Product of Isocyanate and Organosilicone	255
Corona Treatment of Silicone Surfaces	258
Nitrogen-Containing Release Coatings	260
Acylated Polyethyleneimines	260
Stearyl Methacrylate-Acrylonitrile Copolymers	262
Stearic Acid-Morpholine Release Agents	264
Alkyl Sulfide, Alkyl Sulfoxide and Alkyl Sulfone Terminated Oligomers	266
Other Release Coatings	267
Polyvinyl Alcohol and Werner Complex Fluorocarbon	267
Gelled Cellulose Triacetate	269
Printable Release Coat	270
SPECIALTY TAPES	275
Electrical	275
Self-Fusing Tape Based on EPDM Elastomer	275
Linerless Tape Having Elastomeric Backing	278
Polyimides Containing Inorganic Flakes	281
Acrylic Polymers	283
Acrylic Vinyl lactam Copolymers	286
Highly Aromatic Polyester Resin	288
Conductive Carbon Black and Elastomers	291
Electrically Conductive Projections	293
Conductive Tape Using Metal Mesh and Silver Particles	294
Fluorocarbon Film for Silicone Adhesive	296
Direct Bonding of Coatings Without the Use of Primer	298
Medical	301
Primer Applied to Opposite Side of Backing	301
Foamed Tape	303
Polyurethane Foam	305
Woven Spandex	307
Micropleated Web Using Wood Pulp, Hemp and Rayon Fibers	309
Pervious Tape	310
Breathable Tape Containing Hydroxyethylcellulose	316
Zinc Resinate	319
Polyisoprene, Block Copolymers and Natural Rubber	320
Nonwoven Fabric Coated with Natural and Synthetic Elastomers	322
Corticosteroid Treatment of Dermatological Lesions	326

Adhesive Containing 5-Fluorouracil	328
Retinoic Acid Dermatological Tape.	330
Transfer.	332
Noncontinuous Adhesive Patterns.	332
Copy Sheet for Patterns	334
Recording Paper for Dental Pantograph	336
Recording Sheet of Polyolefin Fibers	341
Translucent Sheet of Printing Characters.	342
Correction Tape for Hectographic Stencils	344
Transfer Adhesives for Rolled Paper Products.	344
Dry Transfer	347
Other Applications	350
Facing Lens Abrading Tool	350
Waterproof Tape.	350
Low Temperature Tar Tape for Pipe Wrapping	351
Encapsulated Epoxy Sealant for Mechanical Fasteners	352
Gold Reflective, Polyimide Based Tape.	354
Tear Tape	355
Two-Color Embossable Tape.	355
Sewing Tape with Lines of Perforations	357
Nongumming Sewing Tape	357
Adhesive Zipper Tapes	358
Shipping Documents Tape.	358
CONTAINERS, LABELS AND LAMINATES.	359
Containers.	359
Reusable Composite Sealing Tapes	359
Bread Wrapper Sealing.	359
Polyamide Films	360
Polyethylene Coatings for Multiwall Bags	362
Closure for Hermetically Sealed Cans	363
Mail Envelope	366
Labels	367
Recording Label	367
Dry Decals.	367
Adhesive Coated Label with Silicone Release Coating.	369
Polyurethane Elastomer in Label Sheet Stock.	370
Self-Sticking Material of EVA-VC Graft Polymer	373
Wash-Off Labels	375
Electrophotographic Reproduction Label	377
Laminates	377
Asbestos-Foam Products.	377
Asbestos Laminates.	378
Slidable Laminate	379
Polyisocyanate-Polyvinyl Alcohol Barrier Layer	380
Bituminous Adhesive.	381
Bisimide-Polyamine Reaction Products	383
Polyimide Film Coated with Acrylic Resin	385
Bonding Using Strings of Adhesive	386
Polyvinyl Acetate-Impregnated Plastic Laminate.	387
Adhesive Cement Containing Sulfobutyl Rubber	387
Decorative Adhesive Laminate	389
Laminate for Tear-Resistant Labels.	391
Sprayable Latex Adhesives	391

OTHER END USES	393
Disposable Diapers	393
Integral Fasteners	393
Loop-Type Adhesive Fastener	393
Primary and Secondary Tab Fasteners	394
Pleated Tab Fastener	394
Tab Fastener with Zone-Coated Adhesive	394
Applying Adhesive Tape to Pads	395
Other Specific Applications	395
Wall Covering Employing Encapsulated Adhesive	395
Wallpaper	397
Self-Supporting, Non-Load-Bearing Resilient Tape Sealant	399
Adhering Preformed Resins to Architectural Surfaces	401
Lint Remover	402
Tape Splice Connection	403
Photograph Mounting Sheet	405
Protective Sheet for Metal Working Plates	406
Disposable Headrests	408
Solar Control Film for Window Glass	409
Vehicle Sun Blindness Eliminator	410
Road Markings	410
Sealing Strips for Highways	410
Reusable Bulletin Board	411
Display Board	413
Protecting Hulls of Marine Vessels from Fouling	413
Self-Sealing Roof Shingle	414
General Processes	415
No Preformed Core	415
Reusable Tape	416
Slit Vinyl-Backed Tape	416
Light-Reflecting Markings on Tape	416
Removing Adhesive	417
Joining Fibers or Metals	418
Composite of Isocyanate-Coated Elastomer and Metal	419
COMPANY INDEX	420
INVENTOR INDEX	422
U.S. PATENT NUMBER INDEX	426

INTRODUCTION

Pressure-sensitive adhesives generally combine a high degree of tackiness, for instantaneous bonding, with a high cohesive strength to facilitate removal of tape from a surface without leaving any residue. Typically the adhesive is based on a film-forming elastomeric material such as SBR, polychloroprene, polybutadiene and acrylics. Commonly, blends of various materials are needed to optimize initial tack, cohesive strength and, of course, the actual adhesive properties for a given substrate. Considerable research effort has been expended in finding tackifiers for these systems and terpene resins are used in many formulations.

Pressure-sensitive tapes are composed of the following compounds: (a) the adhesive, (b) a primer coating which is applied to the plastic or metal foil surface, (c) a release coating on the backing to allow unrolling of the tape and (d) a suitable backing material such as plastic, metal foil, paper and, in recent years, nonwoven textiles.

In view of the wide utility and consumer acceptance of these pressure-sensitive adhesives, a high level of basic research and product development has evolved over the past 15 years and is continuing. This interest is broadly based throughout the resin industry, tape and container producers, surgical bandage suppliers and in every conceivable segment of the consumer product field.

Thus, the purpose of this book is not only to describe the synthetic resin processes which are so basic to this technology but to provide hundreds of starting formulations for developing new and improved adhesives and to describe in some detail as wide a variety of end uses as possible.

The first two chapters are largely devoted to a consideration of the developments in adhesive resins and formulations, particularly acrylics, styrene-butadiene, modified polybutadiene, polyvinyl ethers and silicones. The construction and use of various backings, which is extremely important where high tensile strength, good tear and flexibility are required, are described in the next chapter, which also describes silicone and many fatty nitrogen-containing compounds, which are used as release agents.

The following chapter describes specific formulations and production techniques for electrical, medical and other end uses. The use of pressure-sensitive adhesives for containers, labels, laminates and other applications, all the way from decorative wall coverings to disposable diapers are described in the last chapter.

Overall, some 280 processes and over 1,000 formulations, as described in the patent literature since 1970 are included in this book. Where possible, for continuity the individual processes are covered with reference to a specific adhesive or application, although it is recognized that many of these pressure-sensitive adhesive systems have broad utility.

ACRYLICS

COPOLYMER COMPOSITION

Emulsion Polymerization of Latently Crosslinkable Polymers

C.M. Samour and D. Satas; U.S. Patent 3,492,260; January 27, 1970; assigned to The Kendall Company describe an aqueous emulsion polymerization system for the preparation of an emulsion of latently crosslinkable polymers where one of the comonomers such as an amic acid or acid ester is in salt form and one of the essential comonomers is selected from the group consisting of acrylamide, methacrylamide, acrylonitrile, methacrylonitrile, cyanoethyl acrylate and cyanoethyl methacrylate. The polymer produced may be coagulated from the dispersion in its uncrosslinked state and then subsequently crosslinked by heating. The preferred crosslinking monomers are the polyesters of polyalcohols and acrylic or methacrylic acid. Of these, the polyester of acrylic or methacrylic acid and polyethylene glycol is preferred.

The carboxylic acid monomer, N-t-C₁₂ maleamic acid, in the following examples is a mixture of maleamic acid monomers prepared by the amidation of maleic anhydride with a complex mixture of t-alkyl primary amines. The t-alkyl groups are highly branched alkyl groups. Primene 81-R is one of a series of t-alkyl amines containing such amines as t-butyl amine; 1,1,3,3-tetramethylbutyl amine; 1,1,3,3,5,5-hexamethylhexyl amine; 1,1,3,3,5,5,7,7-octamethyloctyl amine; 1,1,3,3,5,5,7,7,9,9-decamethyldecyl amine; and 1,1,3,3,5,5,7,7,9,9,11,11-dodecamethyldodecyl amine. Primene 81-R consists principally (90%) of C₁₁ to C₁₄ branched alkyl amines. The neutral equivalent of Primene 81-R is 191 corresponding to an alkyl group of about 12 carbon atoms.

The following examples represent preferred examples of the process for preparation of crosslinkable pressure-sensitive adhesive polymers. The lauryl methacrylate and acrylic acid are optional comonomers which may be added to the reaction mixture in an amount no greater than 20 parts per 100 parts total of the ester and carboxylic acid monomeric constituents (1) and (2).

Example 1:

Monomer composition:

Monomeric constituents (1) and (2):

Ethyl acrylate, ethylhexyl acrylate and N-t-C ₁₂ maleamic acid in the molar proportions of 1:4:1	100.0 g
--	---------

Cross-linking constituent (3):

Polyethylene glycol dimethacrylate	5.0 g
------------------------------------	-------

Amide constituent (4):

Acrylamide	1.5 g
------------	-------

Other comonomers:

Lauryl methacrylate	10.0 g
---------------------	--------

Acrylic acid	5.0 g
--------------	-------

Water	325.0 g
-------	---------

Base: Ammonium hydroxide

Catalyst system:

Hydrogen peroxide (3% in water)	10.0 g
---------------------------------	--------

Reductant	10.7 cc
-----------	---------

The monomers were thoroughly mixed together for ½ hour. The water and ammonium hydroxide were separately mixed in a reaction vessel and vigorously stirred while flushing the vessel with N₂ gas. The monomer composition was added and the mixture stirred with formation of an emulsion. The amount of the ammonium hydroxide in the mixture was sufficient to neutralize all of the acrylic acid and 70% of the maleamic acid monomers. Accordingly, the reaction mixture contains 18 parts of the ammonium salt of the N-t-C₁₂ maleamic acid monomer.

After flushing for ½ hour with nitrogen, the emulsion was then cooled in an ice bath to 12°C. The peroxide was added all at once and the reductant added dropwise until an increase in temperature of the reaction mixture was observed. The temperature rose to a maximum of 15.5°C during the reaction. The reductant was a mixture of 0.5 gram ascorbic acid and 0.02 gram ferrous ammonium sulfate in 10 cc of water. When no further rise in temperature was observed, the ice bath was removed. Stirring was continued at room temperature for one hour.

The emulsion was then poured into dilute hydrochloric acid to neutralize the base and convert the salt carboxylate groupings in the polymer to their free acid form. The emulsion was thus broken and the polymer coagulated. The coagulum was washed water. A 20% solution of the polymer coagulum in toluene was prepared.

This solution was spread upon polyethylene terephthalate film. The coated film was air dried at ordinary room temperatures. Samples of the resulting pressure-sensitive adhesive tape were then tested for adhesive properties without any further treatment. Other samples were cured by heating at 150°C for 3 minutes and then tested for the same properties. The gel content and swelling index (SI) of the gel in the adhesive polymer for both the cured and uncured samples were also determined. The gel in the uncured sample formed during the solvent drying step. The polymer from the reaction vessel dissolved in the toluene. The toluene solution contained no gel.

Example 2: The polymerization reaction of Example 1 was repeated, with the following changes in the recipe. Acrylic acid was omitted from the monomer composition. Ammonium hydroxide was present in an amount equivalent to 60% neutralization of the maleamic acid monomers. 4.9 cc of the reductant was added to the reaction mixture. The maximum temperature reached during the reaction was 21.5°C. The polymer was coagulated, a toluene solution of the polymer was prepared and pressure-sensitive adhesive tapes made as in Example 1.

Example 3: The polymerization reaction of Example 1 was repeated, with the following recipe variations. Double the amount of acrylamide was employed. The amount of ammonium hydroxide employed was sufficient to neutralize all of the acrylic acid and only 52% of the maleamic acid monomers. 3.8 cc of the reductant was added to the reaction mixture. The temperature rose a maximum of 5.5°C during the reaction. A small amount of prepolymer formed and separated. Tapes were prepared from the polymer as described in Example 1. The test data for these examples is given in the table below. The letters NC indicate that there was no measurable movement of the tape during at least 100 hours of testing.

Example Adhesion Data Splitting				Creep, hours Gel Data:	
	At 75°F	°F	Ounces	Tack		Percent	SI
1: No cure	82	77	94	555	24	41	17
1: Cured	46	85	87	785	90	71	19
2: No cure	120	65	168	1,078	.01	5	8
2: Cured	84	98	92	1,156	NC	63	20
3: No cure	24	150	35	330	NC	70	7
3: Cured	17	190	27	400	NC	77	6

Epoxy Substituted Acrylics and Diketene

G.W.H. Lehmann and H.A.J. Curts; U.S. Patent 3,563,953; February 16, 1971; assigned to Beiersdorf AG, Germany describe pressure-sensitive adhesives from curable copolymers formed by polymerization of a mixture of (a) one or more primary alkyl acrylates and (b) a minor amount of monomers containing reactive epoxide groups. The monomer mixture for the production of the copolymers may also contain minor amounts of monomers with acid anhydride groups or monomers with carboxyl groups or diketene. When heated for a short time at 50° to 150°C, preferably 60° to 100°C, in the presence of a catalyst the copolymers become crosslinked, whereby solvent-, heat- and weather-resistant pressure sensitive adhesive compositions are obtained. The low temperature curing property is particularly useful in the production of pressure sensitive adhesive coated films and foils.

Example 1: 490 parts by weight of 2-ethylhexyl acrylate, 490 parts by weight of n-butyl acrylate and 20 parts by weight of glycidyl methacrylate were mixed, with the addition of 2 parts by weight of dibenzoyl peroxide, with 306 parts by weight of acetone and 123 parts by weight of petrol, and the mixture was heated to 65°C while stirring in a nitrogen atmosphere in a vessel surrounded by a heating and cooling bath provided with a reflux condenser. After a short time polymerization set in. It was continued for 20 hours while the mixture was gradually diluted with petrol.

The solid content of the copolymer solution obtained was 22 to 24%. The K value according to Fikentscher (*Cellulosechemie* 13, 1932, p 58) measured from a 1% solution of the copolymer in toluene, was 97 to 100. To determine the K value, the solution of the copolymer obtained in the manner described was spread out in a thin film on a glass plate, dried at room temperature and then dissolved in toluene in the proportion required to produce a 1% solution.

1 part by weight of zinc chloride to each 100 parts by weight of solid content was added in the form of a suspension in acetone to the solution of the copolymer and spread on a polyethylene terephthalate film 0.025 mm thick, which had previously been provided with a thin priming coat of an acrylate copolymer as an anchorage coat for the adhesive composition, in a coating thickness of 25 g/m² with the use of a conventional coating device. To evaporate the solvent, the coated film was passed through a heated drying conduit and, after the removal of the solvent, it was then heated for 3 minutes to 100°C to cure and crosslink the copolymer.

The coating, drying and curing processes are advantageously carried out with the use of a machine with a drying conduit of appropriate length. The finished adhesive films or adhesive tapes can then be cut into strips of the desired width and wound on rollers. For this purpose, the backs of the sheets, films or tapes may be provided with adhesive-repellent coats beforehand, or the adhesive side of the support can be covered with paper, which is provided with an adhesive-repellent coat, for example, a silicone separating layer. The resulting pressure-sensitive adhesive tapes showed a good bond strength on steel of 300 g/cm (strip-off angle 180°, strip-off speed 200 mm/min) and very good thermal stability.

To test the holding power at an elevated temperature, a steel plate measuring 4.8 x 10 cm, 1.6 mm thick was polished with abrasive paper in a longitudinal direction and cleaned with hot petrol. An adhesive tape 15 cm long and 2 cm wide was stuck to the steel plate so that, from the edge of one narrow side, a length of 2.54 cm (=1 inch) lay on the steel plate and the attached part of the adhesive tape was pressed on by rolling a roller weighing 2 kg slowly to and fro once. The free end of the tape was loaded by vertical suspension with an 800 gram weight.

The test was carried out at various temperatures and the time until the tape together with the weight fell off was measured. The holding power at elevated temperatures determined in this manner was over 20 hours at all temperatures up to 150°C. Comparison tests on nonvulcanized rubber self-adherent compositions and noncrosslinked polyacrylate self-adherent compositions, on the other hand, yielded values, with the same load, of 3 to 10 minutes at 100°C and 10 to 40 minutes at 50°C.

By quick-stick is meant the immediate adhesiveness of a pressure-sensitive adhesive tape at the moment of contact between the self-adherent composition and the surface to which it is to adhere. To determine the quick-stick, an adhesive tape 150 cm long and 2 cm wide, with the adhesive side upwards, was stuck to a bar bent concavely downwards with a given radius of curvature. A steel ball weighing 63.6 g was then caused to roll downwards over the sticking surface of the tape, the difference in height between the starting point of the ball and the end point of the 150 cm long adhesive tape being 250 mm. The distance from the starting point to the halting point of the ball, measured in centimeters, gave