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Tarikh : 2 Disember 2011

Prof. Madya Poh Beng Teik
Pusat Pengajian Teknologi Industri
Universiti Sains Malaysia

Tuan,

LAPORAN AKHIR SKIM GERAN PENYELIDIKAN FUNDAMENTAL (FRGS)
Tajuk Projek : Effect of Molecular Weight, Phase Separation and Molecular Interaction on the Adhesion Properties of NR and ENR - Based Pressure Sensitive Adhesives
No. Akaun : 203/PTEKIND/6711101

Dengan hormatnya perkara di atas dirujuk.

2. Terlebih dahulu saya ucapkan ribuan terima kasih di atas satu salinan laporan akhir untuk projek penyelidikan seperti tajuk di atas.

3. Adalah dimaklumkan walaupun projek ini telah selesai, kerjasama Jabatan Bendahari dipohon untuk menguruskan penutupan akaun projek pada selewat-lewatnya **31 Disember 2011**. Tempoh ini bertujuan untuk menyelesaikan semua urusan tuntutan dan bayaran yang telah dibelanjakan di dalam tempoh projek. Walau bagaimanapun, tuan dinasihatkan supaya tidak mengeluarkan borang-borang pesanan baru di dalam tempoh ini.

4. Selanjutnya sila ambil perhatian terhadap perkara-perkara berikut sekiranya berkaitan:

(i) Semua penerbitan harus merakamkan penghargaan kepada **Skim Geran Penyelidikan Fundamental (FRGS)** dan tuan dipohon mengemukakan satu salinan ke Pejabat ini.

(ii) Bahagian Penyelidikan & Inovasi boleh/akan mengagihkan semula peralatan yang telah dibeli menggunakan peruntukan geran ini seandainya terdapat penyelidik lain yang memerlukan peralatan tersebut.

5. Akhir sekali, tahniah di atas usaha dan kejayaan pihak tuan dapat menyelesaikan projek ini dengan jayanya.

Sekian, terima kasih.

"BERKHIDMAT UNTUK NEGARA"
'Memastikan Kelestarian Hari Esok'

Yang menjalankan tugas,

(AMRA OTHMAN)
Penolong Pendaftar
Unit Pengurusan Geran & Kontrak

HAH, HAR, SM

LAPORAN AKHIR SKIM GERAN PENYELIDIKAN FUNDAMENTAL (FRGS)

Tajuk Projek : Effect of Molecular Weight, Phase Separation and Molecular Interaction on the Adhesion Properties of NR and ENR - Based Pressure Sensitive Adhesives

No. Akaun : 203/PTEKIND/6711101

s.k. Dekan Penyelidikan
Pelantar Sains Fundamental
Pejabat Pelantar Penyelidikan
Universiti Sains Malaysia

Dekan
Pusat Pengajian Teknologi Industri
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Timbalan Dekan
(Pengajian Siswazah & Penyelidikan)
Pusat Pengajian Teknologi Industri
Universiti Sains Malaysia

Ketua Pustakawan
Perpustakaan Hamzah Sendut
Universiti Sains Malaysia

} Disampaikan satu salinan laporan akhir projek untuk simpanan Perpustakaan

Penolong Bendahari Kanan
Unit Kumpulan Wang Penyelidikan
Jabatan Bendahari
Universiti Sains Malaysia

} Mohon kerjasama pihak puan untuk menguruskan penutupan akaun projek selewat-lewatnya pada <u>31 Disember 2011</u> dan mohon kemukakan satu salinan penyata kewangan terakhir ke Pejabat ini untuk tujuan rekod

Pegawai Sains
Pelantar Sains Fundamental
Pejabat Pelantar Penyelidikan
Universiti Sains Malaysia

F0260

2007

FINAL REPORT
FUNDAMENTAL RESEARCH GRANT SCHEME (FRGS)
Laporan Akhir Skim Geran Penyelidikan Asas (FRGS) IPT
Pindaan 1/2010

A

RESEARCH TITLE *Tajuk Penyelidikan* : Effect of molecular weight, phase separation and molecular interaction on the adhesion properties of NR and ENR-based pressure sensitive adhesives.

PROJECT LEADER *Ketua Projek* : Prof. Madya Poh Beng Teik

PROJECT MEMBERS (including GRA) *Ahli Projek* :
1. Dr. Issam Ahmed Mohammed
2. Yong Ai Thing
3. Deepa a/p Thiagarajan
4. Lim Ye-Wheen
5. Dr. Imran Khan

PROJECT ACHIEVEMENT *(Prestasi Projek)*

B

ACHIEVEMENT PERCENTAGE			
Project progress according to milestones achieved up to this period	**0 - 50%**	**51 - 75%**	**76 - 100%**
Percentage			95

RESEARCH OUTPUT		
Number of articles/ manuscripts/ books *(Please attach the First Page of Publication)*	**Indexed Journal**	***Non-Indexed Journal***
	15	1
Conference Proceeding *(Please attach the First Page of Publication)*	**International**	***National***
	1	1
Intellectual Property *(Please specify)*		

HUMAN CAPITAL DEVELOPMENT					
Human Capital	**Number**				**Others** (please specify)
	On-going		**Graduated**		
Citizen	Malaysian	Non Malaysian	Malaysian	Non Malaysian	
PhD Student					1 Post-Doctoral Fellow
Master Student	2				
Undergraduate Student	4		21		
Total					

EXPENDITURE *(Perbelanjaan)*

C

Budget Approved *(Peruntukan diluluskan)* : **RM 88,000.00**
Amount Spent *(Jumlah Perbelanjaan)* : **RM 63,370.15 (Penyata 30/11/2010)**

Balance *(Baki)* : **RM 24,629.85**
Percentage of Amount Spent *(Peratusan Belanja)* : 72.01%

ADDITIONAL RESEARCH ACTIVITIES THAT CONTRIBUTE TOWARDS DEVELOPING SOFT AND HARD SKILLS *(Aktiviti Penyelidikan Sampingan yang menyumbang kepada pembangunan kemahiran insaniah)*

D

International

Activity	Date (Month, Year)	Organizer
4th USM-JIRCAS Joint International Symposium	18-20 January 2011	USM, JIRCAS and FFPRI

National

Activity	Date (Month, Year)	Organizer
4th Life Sciences Postgraduate Conference USM	18-20 June 2008	USM

PROBLEMS / CONSTRAINTS IF ANY *(Masalah/ kekangan sekiranya ada)*

E

-

RECOMMENDATION *(Cadangan Penambahbaikan)*

F

-

RESEARCH ABSTRACT – Not More Than 200 Words *(Abstrak Penyelidikan – Tidak Melebihi 200 patah perkataan)*

G

The effect of molecular weight of natural rubber (NR) and epoxidized natural rubber (ENR) on viscosity, loop tack, peel strength and shear strength of rubber-based adhesives was studied. Coumarone-indene resin, gum rosin and petro resin were used as the tackifiers whereas toluene was chosen as the solvent. A polyethylene terephthalate (PET) film substrate was coated with the adhesive using a SHEEN hand coater. Viscosity of adhesive was measured using a HAAKE Rotary Viscometer. Loop tack, peel strength and shear strength were determined by a Llyod Adhesion Tester. Results indicate that the viscosity increases with increasing molecular weight of rubber. Maximum values of adhesion properties were obtained at a molecular weight of 8.5×10^4, an observation which is attributed to maximum wettability of adhesive on the substrate. The effect of interaction between rubber and filler was investigated using magnesium oxide, barium chloride and silica. Viscosity of adhesive increases with increasing filler loading. Loop tack, peel strength and shear strength shows a maximum value with filler content due to the maximum wettability and compatibility of adhesive on the substrate. Addition of antioxidant decreases the viscosity and shear strength of the adhesives. Tack and peel strength, however, exhibits the reverse behaviour.

Date : 8-2-2011
Tarikh

Project Leader's Signature:
Tandatangan Ketua Projek

COMMENTS, IF ANY/ ENDORSEMENT BY RESEARCH MANAGEMENT CENTER (RMC)
(Komen, sekiranya ada/ Pengesahan oleh Pusat Pengurusan Penyelidikan)

H ....

....

....

Name:
Nama:

Signature:
Tandatangan:

Date:
Tarikh:

Effect of Molecular Weight of Epoxidized Natural Rubber on Shear Strength of Adhesives

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Received 12 March 2009; accepted 17 June 2009
DOI 10.1002/app.30978
Published online 19 August 2009 in Wiley InterScience (www.interscience.wiley.com).

ABSTRACT: The dependence of shear strength of epoxidized natural rubber (ENR)-based adhesives on molecular weight of the rubber is studied using coumarone–indene resin, gum rosin, and petro resin as tackifiers. The adhesive was coated on polyethylene terephthalate (PET) film substrate using a SHEEN hand coater at various coating thickness. The shear strength of adhesives was determined by a Texture Analyzer. Results show a maximum at 6.63 $\times 10^4$ and 4.14 $\times 10^4$ for ENR 25 and ENR 50, respectively, after which the shear strength decreases with further increases in molecular weight for all the coating thickness. This observation is attributed to varying degree of cohesiveness which culminates at the respective optimum molecular weight of ENR. © 2009 Wiley Periodicals, Inc. J Appl Polym Sci 114: 3976–3979, 2009

Key words: shear strength; molecular weight; adhesive; ENR

INTRODUCTION

In formulating a rubber-based pressure-sensitive adhesive, an elastomer provides the elastic component, whereas a low-molecular-weight tackifying resin imparts the viscous component. Scientific research on rubber-based adhesives—particularly natural rubber—seems scarce. Kraus et al.[1,2] have studied the adhesive behavior of the styrene-diene-based pressure-sensitive adhesives. Leong et al.[3] have investigated the viscoelastic properties of natural rubber pressure-sensitive adhesive using acrylic resin as a tackifier. It was discovered that for a good pressure-sensitive adhesive, the ratio of storage modulus at high frequencies to low frequencies should be high. Fujita et al.,[4] on the other hand, reported the effects of miscibility and viscoelasticity on shear creep resistance of natural-rubber-based pressure-sensitive adhesives. They found that holding time of miscible pressure-sensitive adhesive systems tended to decrease as the tackifier content is increased, whereas the holding time of an immiscible adhesive system varies with the tackifier used. However, with respect to epoxidized natural rubber (ENR)-based pressure-sensitive adhesive, very few research works has been published. Thongnuanchan et al.[5] have carried out a study on epoxidized natural rubber-bonded para rubber wood particleboard. The adhesion of the epoxidized natural rubber adhesive with parawood sawdust was observed to improve by reducing the molecular weight of epoxidized natural rubber molecules. It was suggested that lower molecular weight exhibited greater ability to wet or cover the wood particle surfaces. Recently, we have carried out several studies on the adhesion behavior of ENR-based pressure-sensitive adhesives. These include the effect of zinc oxide, calcium carbonate, and rubber blends on the viscosity, tack, and peel strength of ENR-based pressure-sensitive adhesives.[6–8] We have also investigated the shear property of ENR-based adhesives.[9] It is found that shear strength decreases with increasing coumarone–indene resin for all the coating thickness studied. The rate of decrease is greatest for the thicker coating sample. ENR 25 shows higher shear strength than ENR 50 due to the greater flexibility and compatibility with resin in the former system. The previous study was carried out using unmasticated ENR. No investigation was reported so far on the dependence of shear strength on the molecular weight of ENR. In view of the lack of scientific research in this field of interest, we have carried out a systematic research on the effect of molecular weight of ENR on the shear strength of the adhesive in the presence of various tackifiying resins.

EXPERIMENT

Materials

ENR 25 and ENR 50 having 25 mol % and 50 mol % of epoxidation, respectively, were used as the elastomers, which were supplied by Rubber Research Institute of Malaysia (RRIM). The gel content of ENR is estimated to be less than 5%. Coumarone–indene

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Contract grant sponsor: Universiti Sains Malaysia (FRGS).

Effect of Silica on Viscosity, Tack, and Shear Strength of Epoxidized Natural Rubber-Based Pressure-Sensitive Adhesives in the Presence of Coumarone-Indene Resin

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Received 13 January 2010; accepted 25 April 2010
DOI 10.1002/app.32686
Published online 14 July 2010 in Wiley InterScience (www.interscience.wiley.com).

ABSTRACT: The viscosity, loop tack, and shear strength of silica-filled epoxidized natural rubber (ENR 25 and ENR 50 grade) adhesive were investigated using coumarone-indene as the tackifying resin. Silica loading was varied from 10–50 parts per hundred parts of rubber (phr), whereas the coumarone-indene concentration was fixed at 40 phr. Toluene was used as the solvent throughout the study. Polyethylene terephthalate substrate was coated at various adhesive coating thicknesses, i.e., 30, 60, 90, and 120 μm using a SHEEN Hand Coater. Viscosity of the adhesive was determined by a HAAKE Rotary Viscometer whereas loop tack and shear strength were measured by a Llyod Adhesion Tester operating at 30 cm/min. Result shows that viscosity of the adhesive increases gradually with increase of silica loading due to the concentration effect of the filler. Both loop tack and shear strength show maximum value at 40 phr silica for ENR 25. However, the respective values for ENR 50 are 20 and 40 phr of filler. This observation is attributed to the maximum wettability and compatibility of adhesive on the substrate at the respective silica loadings. © 2010 Wiley Periodicals, Inc. J Appl Polym Sci 118: 3439–3444, 2010

Key words: silica; viscosity; tack; shear strength; rubber

INTRODUCTION

The structure of filled rubber compounds on a molecular scale, in particular when silica is used as filler, is not very well known. Silica has a number of hydroxyl groups on the surface, which results in strong filler–filler interactions and adsorption of polar materials by hydrogen bonds.[1–3] Mixing of silica with rubber is a challenge, as these materials are not very compatible in various aspects: surface energy, solubility, structure of the filler and viscoeslastic properties. Silica has been used as an important reinforcing agent in a rubber compound together with carbon.[1]

Most pressure-sensitive adhesives (PSAs) are blends of rubbery polymers and oligomeric tackifier resins. PSA tapes can paste on various adherends under light pressure in very short time without heating or heavy pressure. The bonds formed are strong enough to use temporarily. However, PSAs sometimes need durability for a long time, especially the resistance to shear force. For example, PSAs are used for bookbinding, upholstery, and pasting papers on the wall.[4]

Epoxidized natural rubber (ENR) is prepared by peroxyacetic acid epoxidation of natural rubber (NR) latex. The epoxidation reaction is a random process. The glass transition temperature of ENR 25 and ENR 50 is −45 and −20°C, respectively. ENR undergoes strain-induced crystallization. The curing characteristics and mechanical properties of epoxidized natural rubbers (ENR) have been widely studied.[5–14] Ishak and coworkers[15] has studied the curing characteristics and mechanical properities of ENR with filler silica and has reported that cure characteristics could be related not only to the fillers characteristics, i.e., particle size, structure, and aspect ratio, but also to the filler–rubber interaction. All these studies involve the bulk property of ENR. The solution property of ENR, especially its use in PSA was not widely reported. Recently, we have investigated the shear property of ENR-based adhesives[16] and the effect of zinc oxide, calcium carbonate, and rubber blends on the adhesion properties of ENR-based PSAs.[17–19] In view of the lack of scientific research in this field of interest, we have carried out a systematic research on the effect of silica on ENR on the viscosity, tack, and shear strength of the adhesive in the presence of coumarone-indene resins.

Correspondence to: B. T. Poh (btpoh@usm.my).
Contract grant sponsor: Universiti Sains Malaysia (FRGS).

EXPERIMENTAL

Materials

Epoxidized natural rubber, ENR-25 and ENR-50 having 25 and 50 mol % of epoxidation, respectively

Effect of Molecular Weight and Testing Rate on Peel and Shear Strength of Epoxidized Natural Rubber (ENR 50)-based Adhesives

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DOI 10.1002/app.33492
Published online 00 Month 2010 in Wiley Online Library (wileyonlinelibrary.com).

ABSTRACT: The dependence of peel strength and shear strength of epoxidized natural rubber (ENR-50)-based pressure sensitive adhesive on molecular weight and rate of testing was investigated using coumarone-indene as the tackifying resin. Toluene and polyethylene terephthalate were used as the solvent and substrate respectively, throughout the study. A SHEEN hand coater was used to coat the adhesive on the substrate at a coating thickness of 120 μm. All the adhesion properties were determined by a Llyod Adhesion Tester operating at different rates of testing. Result shows that peel strength and shear strength increases up to an optimum molecular weight of 4.2×10^4 of ENR 50. For peel strength, the observation is attributed to the combined effects of wettability and mechanical strength of rubber at the optimum molecular weight, whereas for the shear strength, it is ascribed to the optimum cohesive and adhesive strength which enhances the shear resistance of the adhesive. Peel strength and shear strength also increases with increase in rate of testing, an observation which is associated to the viscoeslastic response of the adhesive. DSC and FTIR study confirms the miscibility of tackifier and the ENR 50. © 2010 Wiley Periodicals, Inc. J Appl Polym Sci 000: 000–000, 2010

Key words: molecular weight; rate of testing; adhesion; epoxidized natural rubber

INTRODUCTION

Epoxidized natural rubber (ENR) is a chemically modified form of the *cis*-1,4-polyisoprene rubber, whereby some of the unsaturation is converted into epoxides groups, which are randomly distributed along the polymer chain.[1] It is known as a compatibilizer for incompatible blends and a processing aid. The incorporation of ENR-50 into the rubber blends has improved process ability, stiffness, resilience and oil resistance, reduced air permeability, good damping, and wet grip performance.[2] The concept of physically blending two or more existing polymers to obtain a new product has not been developed as fully as the chemical approach to blending, but the physical approach is now attracting widespread interest and is being used commercially. The main aim of blending the rubber is to improve the physical and mechanical properties as well as modify processing characteristics and reduce the cost of the final product.

The adhesion properties are characterized via measurements of two basic applicative properties: peel strength (the ability to resist removal by peeling), and shear resistance (the ability to resist flow when shear forces are applied). The adhesion properties are primarily influenced by the inherent properties of the polymer such as molecular weight. They have an influence on the polymer properties directly and as well as indirectly through their influence on the physical properties (e.g., T_g).[3] FTIR study has been used to see differences in peaks with changes in molecular weight of ENR 50 and variation of coumarone-indene resin content. Our previous study focuses on the dependence of adhesion property on molecular weight at different coating thickness.[4–7] The aim of this study is to investigate the effect of molecular weight and testing rate on adhesion property of ENR. We have also studied effects of zinc oxide,[8] calcium carbonate,[9] silica,[10] magnesium oxide,[11] kaolin,[12] and sodium sulfate[13] on the adhesion properties of ENR-based pressure-sensitive adhesives. This work is novel as no author has studied adhesion property with molecular weight and testing rate.

The end-use properties of the PSAs will depend upon balance of peel strength and shear resistance and the balance between these properties must be changed according to the specific end use of the PSA.[14] Commercial use of PSAs covers abroad range of label, medical, and cosmetic products.[15,16] Fujita

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Contract grant sponsor: Universiti Sains Malaysia (FRGS).

Viscosity and Shear Strength of Natural-Rubber-Based Adhesives in the Presence of Gum Rosin and Petroresin

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Received 14 December 2007; accepted 29 July 2008
DOI 10.1002/app.29012
Published online 23 September 2008 in Wiley InterScience (www.interscience.wiley.com).

ABSTRACT: The viscosity and shear strength of pressure-sensitive adhesives based on natural rubber (standard Malaysian rubber grade L) were studied with gum rosin and petroresin as the tackifying resins. Effects of the concentration of the tackifying resin and the molecular weight of rubber on the two properties were systematically investigated. Toluene was used as the solvent throughout the study to prepare the adhesives. The viscosity and shear strength of the adhesives were determined with a rotary viscometer and a texture analyzer, respectively. For the shear test, a hand coater was used to coat the adhesives on the release paper substrate to provide coating thicknesses of 60 and 120 μm. The results indicated that the viscosity increased with the resin loading and molecular weight of rubber increasing. The viscosity of the adhesive prepared from petroresin had a higher value than that of the gum-rosin-based adhesive. The shear strength of the adhesives decreased gradually with increasing resin content for both tackifying resins and coating thicknesses, and this observation was attributed to the decrease in the cohesive strength due to the dilution effect of the resins. However, the shear strength passed through a maximum at a molecular weight of rubber of 8.5×10^4 for both resins. The gum-rosin-based adhesive consistently showed higher shear strength than that of the petroresin/natural rubber adhesive because of the better cohesiveness and compatibility of the former system. J Appl Polym Sci 110: 4079–4083, 2008

Key words: adhesives; resins; rubber; shear; viscosity

INTRODUCTION

We have recently carried out several studies on the viscosity and adhesion properties of natural-rubber-based pressure-sensitive adhesives with coumarone–indene resin as the tackifying resin.[1–5] The results have shown that the viscosity and tack of the adhesives increase with the resin content because of the concentration effect of the tackifying resin. The peel strength increases with the resin content up to a maximum value of 40 phr resin, at which optimum wettability of the substrate is achieved. However, the shear strength decreases gradually with increasing resin content, and this observation has been attributed to the decreasing cohesive strength of the adhesives as the resin loading is increased.[3,4] We have also extended our study to rubber blend systems using coumarone–indene resin as the tackifier.[6,7] There have been very few studies on the adhesion properties of natural-rubber-based pressure-sensitive adhesives prepared from tackifiers other than coumarone–indene resin. To understand better the effects of other tackifiers, here we report our findings on the effects of the tackifier concentration and molecular weight of rubber on the viscosity and shear strength of natural-rubber-based adhesives using two other tackifier types, that is, gum rosin and petroresin.

EXPERIMENTAL

Materials

Standard Malaysian rubber grade L (SMR L) was used as the natural rubber to prepare the pressure-sensitive adhesives in this study. The technical specifications of SMR L[8] are given in Table I. Gum rosin and petroresin were freshly supplied by Euro-Chemo-Pharma Co. (Prai, Penang, Malaysia). Toluene was used as the solvent to prepare the adhesives throughout the experiments.

Determination of the molecular weight of rubber

Five rubber samples were obtained by mastication with a two-roll mill. A viscometric method was used to determine the molecular weight of each masticated sample. The intrinsic viscosity ([η]) was measured according to the method described by Billmeyer.[9] The viscosity-average molecular weight (M_v) of the rubber was computed with the Mark–Houwink equation:[10]

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Effect of Molecular Weight of Epoxidized-Natural Rubber on Viscosity and Tack of Pressure-Sensitive Adhesives

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School of Industrial Technology, Universiti Sains Malaysia, Penang 11800, Malaysia

Received 22 December 2008; accepted 25 July 2009
DOI 10.1002/app.31185
Published online 15 September 2009 in Wiley InterScience (www.interscience.wiley.com).

ABSTRACT: The effect of molecular weight of rubber on viscosity and loop tack of rubber-adhesives were studied using two grades of epoxidized-natural rubber, i.e., ENR 25 and ENR 50. Coumarone–indene resin, gum rosin, and petro resin were used as tackifiers. Toluene was used as the solvent throughout the experiment. The adhesive was coated on polyethylene terephthalate (PET) substrate using a SHEEN hand coater. Viscosity was determined by a HAAKE Rotary Viscometer, whereas loop tack was measured by a Llyod Adhesion Tester operating at 10 cm/min. Results show that viscosity increases gradually upto a critical molecular weight of 6.8 × 10^4 and 3.9 × 10^4 for ENR 25 and ENR 50, respectively, before a rapid increase in viscosity is observed. Loop tack indicates maximum value at the respective critical molecular weights for the three tackifiers investigated suggesting the culmination of wettability. For both rubbers, loop tack increases with coating thickness due to the concentration effect of adhesive. © 2009 Wiley Periodicals, Inc. J Appl Polym Sci 115: 1120–1124, 2010

Key words: viscosity; tack; rubber; molecular weight

INTRODUCTION

Recently, we have reported the effect of molecular weight of rubber on the viscosity and shear strength of SMR L-based adhesives in the presence of gum-rosin and petro-resin tackifiers.[1] For both tackifying resins, viscosity increases gradually with molecular weight upto 9.7 × 10^4 after which it increases rapidly with further increase in molecular weight of rubber due to the effect of entanglement of rubber chains. On the other hand, shear strength of the SMR L-based adhesive increases with molecular weight upto 8.5 × 10^4, after which it drops with further increase in molecular weight of rubber for the two coating thickness investigated.

The effect of molecular weight of rubber on tack and peel strength of SMR L-based pressure-sensitive adhesives using gum rosin and petro resin as tackifiers were also investigated.[2] Maximum loop tack and peel strength were observed at a molecular weight of 8.5 × 10^4 of SMR L. Several studies on the adhesion properties of epoxidized-natural rubber (ENR) were studied, including the effect of coumarone–indene resin, zinc oxide, and calcium carbonate on adhesion property of ENR-based pressure-sensitive adhesives.[3–5] However, the effect of molecular weight of ENR on the adhesion behavior of ENR-based adhesives is unknown. In view of the absence of research in this field of interest, we have carried out a systematic study on the dependence of viscosity and loop tack of adhesives on molecular weight of epoxidized-natural rubber using two grades of ENR, i.e., ENR 25 and ENR 50.

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Contract grant sponsor: Universiti Sains Malaysia (FRGS).

EXPERIMENTAL

Materials

Two grades of epoxidized-natural rubber, i.e., ENR 25 and ENR 50 having 25 mol % and 50 mol % of epoxidation, respectively, were used as the elastomers. The technical specifications of ENR[6] are shown in Table I. The rubbers were supplied by Rubber Research Institute of Malaysia (RRIM). Coumarone–indene resin, gum rosin, and petro resin were freshly supplied by EuroChemo-Pharma Company (Malaysia). Commercial grade toluene was used as the solvent throughout the experiment.

Molecular weight determination

ENR 25 and ENR 50 were masticated on a two-roll mill for 5, 10, 15, and 20 min to obtain different molecular weights of rubber. Viscometric method was adopted to determine the molecular weights of masticated- and unmasticated-rubber. For each rubber sample, five different concentrations (C) of dilute rubber solutions were prepared in toluene and the respective flow times (t) were determined using an

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DOI: 10.1080/00218460903040941

Dependence of Peel Adhesion on Molecular Weight of Epoxidized Natural Rubber

B. T. Poh and A. T. Yong
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The effect of molecular weight of two grades of epoxidized natural rubber (ENR)—i.e., *ENR 25 and ENR 50—on the peel strength of an adhesive is studied using coumarone-indene resin, gum rosin, and petro resin as tackifiers. Toluene and polyethylene terephthalate (PET) film acted as the solvent and substrate, respectively. A SHEEN hand coater was used to coat the adhesive on the substrate to give coating thicknesses of 30, 60, 90, and 120* μm. *The peel strength of adhesive was determined using a Lloyd Adhesion Tester operating at 30 cm/min. Results show that peel strength has a maximum value at a molecular weight of* 6.8×10^4 *and* 3.9×10^4 *for ENR 25 and ENR 50, respectively, an observation which is attributed to the combined effects of wettability and mechanical strength of the rubber at the respective optimum molecular weight of ENR. Peel strength increases with coating thickness for all the tackifiers investigated, with a gum rosin-based adhesive exhibiting the highest peel strength.*

Keywords: Adhesive; Molecular weight; Peel strength; Rubber

INTRODUCTION

Recently, we have carried out several studies on the adhesion behavior of epoxidized natural rubber (ENR)-based pressure-sensitive adhesives using unmasticated rubber. Results show that the maximum peel strength of ENR 25 and ENR 50-based adhesives occurs at 40 parts per hundred parts of rubber (phr) of coumarone-indene resin [1]. The shear strength shows a gradual decrease with increasing tackifier loading due to the decrease in cohesive strength of adhesive. On the other hand, viscosity and loop tack of ENR 25-based adhesive increases with

Received 9 January 2009; in final form 30 April 2009.
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Effect of Kaolin on Adhesion Property of Epoxidized Natural Rubber-based Pressure-sensitive Adhesive Using Gum Rosin as the Tackifier

B. T. POH* AND C. E. CHEW

School of Industrial Technology, Universiti Sains Malaysia 11800 Penang, Malaysia

ABSTRACT: The viscosity, loop tack, and peel strength of kaolin-filled epoxidized natural rubber (ENR 25 grade) adhesive was investigated using gum rosin as the tackifying resin. Kaolin loading was varied from 10–50 parts per hundred parts of rubber (phr), whereas the gum rosin concentration was fixed at 40 phr. Toluene was used as the solvent throughout the study. Polyethylene terephthalate substrate was coated at various adhesive coating thicknesses, i.e., 30, 60, 90, and 120 μm using a SHEEN hand coater. A HAAKE Rotary Viscometer Viscosity was used to measure the viscosity of the adhesive. Loop tack and peel strength were determined by a Llyod Adhesion Tester operating at 30 cm/min. Results show that viscosity of the adhesive increases gradually with increase of kaolin loading due to the concentration effect of the filler. Loop tack and peel strength, however, show maximum value at 20 phr and 30 phr kaolin, respectively, an observation which is attributed to the maximum wettability and compatibility of adhesive on the substrate.

KEY WORDS: adhesion, kaolin, rubber, adhesive.

INTRODUCTION

IN FORMULATING A rubber-based pressure-sensitive adhesive, an elastomer provides the elastic component, while a low molecular

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Dependence of Adhesion Property of SMR L-based Adhesives on Antioxidants

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ABSTRACT: The viscosity, tack, peel, and shear strength of Standard Malaysian Rubber (SMR L grade)-based pressure-sensitive adhesives was studied in the presence of two types of antioxidants, that is, 2,6-di-tert-butyl-*p*-cresol (BHT) and 2,2,4-trimethyl-1,2-dihydroquinoline (TMQ) using coumarone-indene resin and toluene as the tackifier and solvent, respectively. The concentration of antioxidant was varied from 1 to 4 parts per hundred parts of rubber (phr). A SHEEN hand coater was used to coat the adhesive on polyethylene terephthalate substrate to give a coating thickness of 90 μm. Viscosity was determined by a HAAKE Rotary Viscometer. Tack, peel, and shear strength were measured using a Lloyd Adhesion Tester operating at 30 cm/min. Results indicate that the viscosity and shear strength decreases with increasing antioxidant concentration, an observation that is attributed to the plasticizing effect of the antioxidants. However, tack and peel strength shows the reverse behavior, a phenomenon that is associated to the increasing wettability and compatibility of adhesive on the substrate as antioxidant concentration is increased. Except for the shear strength, BHT antioxidant consistently exhibits a higher viscosity, tack, and peel strength compared to TMQ antioxidant.

KEY WORDS: antioxidant, rubber, adhesive, adhesion.

INTRODUCTION

THE VISCOSITY, TACK, peel, and shear strength of natural rubber-based pressure-sensitive adhesives have been investigated recently [1–3]

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Effect of Sodium Sulfate on the Viscosity, Tack, and Adhesion Properties of SMR 10-based Pressure-sensitive Adhesive

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ABSTRACT: The effect of sodium sulfate as filler on viscosity, tack, and adhesion properties of Standard Malaysian Rubber (SMR 10)-based pressure-sensitive adhesive were studied. Coumarone-indene resin and toluene were used as the tackifier and solvent respectively throughout the experiment. The results show that viscosity of adhesive increases with addition of sodium sulfate. For rolling ball tack test, the distance traveled by rolling ball decreases with increasing sodium sulfate concentration indicating the increase of tack value with sodium sulfate. The distance traveled by the rolling ball decreases with coating thickness of adhesive. For cross-hatch adhesion test, result shows that maximum adhesion occurs between 30 and 40 parts per hundred parts of rubber (phr) of sodium sulfate content, an observation which is attributed to the maximum of wettability of adhesive. The 60 μm of adhesive coating thickness shows good performance of adhesion property of SMR 10-based pressure-sensitive adhesive.

KEY WORDS: viscosity, tack, adhesion properties, sodium sulfate, natural rubber.

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Effect of Molecular Weight of Rubber on Tack and Peel Strength of SMR L-Based Pressure-Sensitive Adhesives using Gum Rosin and Petroresin as Tackifiers

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Loop tack and peel strength of natural rubber (SMR L)-based pressure-sensitive adhesive were studied using five different molecular weights of SMR L. Gum rosin and petroresin were used as tackifiers, whereas toluene was chosen as the solvent throughout the experiment. A SHEEN hand coater was used to coat the adhesive on a polyethylene terephthalate (PET) substrate at a coating thickness of 30, 60, 90 and 120 μm. Loop tack and peel strength were determined by a Llyod Adhesion Tester operating at 30 cm/min. Results show that maximum values of loop tack and peel strength were obtained at a molecular weight of 8.5×10^4, an observation which is attributed to maximum wettability of adhesive on the substrate. Loop tack and peel strength increases with coating thickness for all molecular weight of rubber and tackifiers studied.

Keywords: Adhesive, rubber, tackifier, molecular weight

1 Introduction

Pressure-sensitive adhesives are commonly prepared from natural rubber. In order to achieve the desired tack, peel adhesion and shear properties, tackifiers are added to the rubber (1). The rubber provides the elastic component whereas the tackifier imparts the viscous component. Systematic studies on the adhesion properties of rubber-based adhesives were scarcely reported. Kraus et al. (2) have reported the effect of entanglement plateau on the adhesive behavior of the styrene-diene-based pressure-sensitive adhesives. They (3) have also studied the structural changes in melts of butadiene-styrene and isoprene-styrene block polymer-based adhesives. Leong et al. (4) have studied the viscoelastic properties of natural rubber pressure-sensitive adhesive using acrylic resin as a tackifier. Fujita et al. (5), on the other hand, reported the effects of miscibility and viscoelasticity on shear creep resistance of natural rubber-based pressure-sensitive adhesives. Besides the rubber-based adhesives, other adhesive systems have also been investigated (6–7). Recently, we have reported several studies on the adhesion properties of Standard Malaysian Rubber (SMR L, SMR 10 and SMR 20 grades)-based pressure-sensitive adhesives (8–10). Results have shown that viscosity, peel strength and tack of SMR-based adhesives generally increases with an increase of tackifier loading, an observation which is attributed to the increased wettability and formation of mechanical interlocking, and anchorage of adhesive in pores and irregularities in the substrate. However, shear strength decreases gradually with increasing tackifier concentration due to the decreasing cohesive strength of adhesive as resin loading is increased. We have also carried out a systematic study on the adhesion properties of adhesives prepared from styrene-butadiene rubber (SBR)/Standard Malaysian Rubber (SMR L) blends (11–12). It is observed that the viscosity of the SBR/SMR L-based adhesive decreases with increasing % SBR. Loop tack of the rubber blend-based adhesive passes through a maximum value at 20% SBR composition for all resin loadings investigated. For the coumarone-indene resin adhesive, peel strength exhibits maximum value at 40% SBR whereas for the phenol-formaldehyde resin system, maximum peel strength occurs at 60% SBR composition. With regard to the effect of molecular weight of rubber on the adhesion property of pressure-sensitive adhesives prepared from SMR rubber, there is virtually no study published so far in this field of interest. Owing to the scarcity of data in this area of research, we have conducted a systematic investigation on the dependence of the loop tack and peel strength of SMR L-based adhesive on molecular weight of rubber using gum rosin and petroresin as the tackifying resins.

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ORIGINAL PAPER

Effect of Hybrid Tackifiers on Adhesion Properties of Epoxidized Natural Rubber-Based Pressure-Sensitive Adhesives

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Abstract Viscosity, peel and shear strength of epoxidized natural rubber (ENR)-based pressure-sensitive adhesive was studied by using hybrid tackifiers consisting of a mixture of coumarone-indene resin and petro resin. The coumarone-indene resin concentration was fixed at 40 parts per hundred parts of rubber (phr). The concentration of petro resin, however, was varied from 20 to 80 phr. Toluene and polyethylene terephthalate (PET) film were used as the solvent and coating substrate respectively throughout the experiment. Viscosity of adhesive was determined by a HAAKE Rotary Viscometer whereas peel and shear strength was measured by a Lloyd Adhesion Tester. Results show that viscosity and shear strength decreases with increasing petro resin concentration. However, peel strength exhibits a maximum value at 40 phr petro resin, an observation which is attributed to maximum wettability and compatibility of adhesive on the substrate. ENR 25-based adhesive exhibits higher viscosity and peel strength but lower shear strength compared to the ENR 50 adhesive system.

Keywords Adhesive · Adhesion · Rubber · Tackifier

Introduction

In formulating a rubber-based pressure-sensitive adhesive, an elastomer provides the elastic component while a low molecular weight tackifying resin imparts the viscous component. Natural rubber alone has a very low tack and adhesion to surfaces [1]. Therefore, it is necessary to add tackifying resins to the elastomer to produce the required balance of tack, peel adhesion and resistance to shear forces. Recently, we have carried out several studies on the adhesion behavior of rubber-based pressure-sensitive adhesives using a single tackifier system. These investigations include effect of coumarone-indene resin on the viscosity, tack, peel and shear strength of natural rubber [2–5] and epoxidized natural rubber (ENR) [6–8]. The effect of phenol formaldehyde resin on the adhesion properties of styrene-butadiene rubber (SBR)/Standard Malaysian Rubber (SMR L)-based adhesives was also studied [9]. On the other hand, Leong et al. [10] have reported the viscoelastic properties of natural rubber pressure-sensitive adhesive using acrylic resin as a tackifier. However, the effect of mixture of tackifiers (i.e. hybrid tackifier) on the adhesion properties of rubber-based adhesives is not published so far. Owing to the importance of tackifiers in rubber-based adhesives, we have carried out a systematic study on the effect of hybrid tackifier involving coumarone-indene resin and petro resin on the viscosity and adhesion behavior of ENR-based pressure-sensitive adhesives.

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Experimental

Materials

ENR 25 and ENR 50 having 25 and 50 mol% of epoxidation respectively were used as the elastomers. Two ENRs were used in order to study the effect of hybrid tackifiers on ENRs of different degree of epoxidation. The respective technical specifications of ENR are shown in Table 1. The rubbers were supplied by Rubber Research Institute of

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Effect of Barium Chloride Filler on the Adhesion Properties of Epoxidized Natural Rubber (ENR 25)-Based Adhesives

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The adhesion properties of epoxidized natural rubber (ENR 25)-based adhesive were studied using barium chloride, coumarone-indene resin and toluene as the filler, tackifier and solvent respectively. Viscosity was determined by a Brookfield Viscometer whereas tack, shear and peel strength was measured by a Llyod Adhesion Tester. Results show that viscosity of adhesive increases gradually with increasing barium chloride loading. Loop tack, shear and peel strength indicates a maximum value at 10 parts per hundred parts of rubber (phr) of barium chloride, an observation that is attributed to the maximum wettability and compatibility of adhesive on the substrate.

Keywords Adhesion property; Adhesive; Barium chloride; Viscosity

INTRODUCTION

In formulating a rubber-based pressure-sensitive adhesive, an elastomer provides the elastic component, while a low molecular weight tackifying resin imparts the viscous component. Several studies on the adhesion properties of rubber-based adhesives have been reported. These include the investigation of entanglement plateau and structural changes of pressure-sensitive adhesives of styrene-diene block polymer-based pressure-sensitive adhesives[1–2], the viscoelastic properties of natural rubber pressure-sensitive adhesive using acrylic resin as a tackifier[3]. Recently, we have reported several studies on the adhesion properties of natural rubber (NR) and epoxidized natural rubber (ENR)-based pressure-sensitive adhesives using coumarone-indene resin as the tackifying resin[4–6].

Results indicate that the adhesion properties, except shear strength, of the adhesive generally increase with an increase in tackifier concentration. Shear strength, on the other hand, decreases gradually with increasing resin content. We have also extended our study on the effect of filler on the adhesion properties of epoxidized natural rubber-based adhesives. Viscosity and loop tack increases with increasing zinc oxide concentration whereas peel strength exhibits maximum value at 30–40 phr of zinc oxide depending on the tackifier loading used[7]. Similar behavior was observed in the case of calcium carbonate-filled ENR adhesives[8].

Our recent study indicates that loop tack and peel strength show maximum value at 20 phr and 30 phr kaolin, respectively, an observation that is attributed to the maximum wettability and compatibility of adhesive on the substrate[9]. The growing interest of filler on the adhesion property of rubber-based adhesive has prompted us to extend our investigation on the effect of barium chloride on the adhesion properties of adhesives prepared from ENR 25. Results obtained are discussed with respect to the dependence of viscosity, tack, shear and peel strength on the concentration of barium chloride in the adhesive system.

EXPERIMENTAL

Materials

Epoxidized natural rubber (ENR 25 grade), having 25 mol% of epoxidation was used as the elastomer. It was supplied by Rubber Research Institute of Malaysia (RRIM). Coumarone-indene resin which has a molecular weight of 1000–3000 and specific gravity of 1.07 was obtained from Mukmin Enviro Company (Malaysia). Barium chloride and toluene were used as the filler and solvent respectively in this study.

Preparation of Adhesive

ENR 25 was masticated on a 2-roll mill for 10 minutes. For each adhesive formulation, 5 g of the shredded rubber was dissolved in 30 ml of toluene and the rubber solution was tightly closed. It was kept for 24 hours to ensure complete dissolution of the rubber. A fixed amount of 2 g of pulverized coumarone-indene resin was then slowly added to the rubber solution. After the complete dissolution of the resin, 5 different weights of barium chloride, i.e., 0.5, 1, 1.5, 2 and 2.5 g corresponding to 10, 20, 30, 40 and 50 phr of filler were then added to the rubber adhesive with constant stirring. For comparison purposes, 1 control sample without barium chloride was also prepared.

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The Effect of Silica on the Peel Adhesion of Epoxidized Natural Rubber-Based Adhesive Containing Coumarone-Indene Resin

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The peel strength of silica filled on two grades of epoxidized natural rubber (ENR), i.e., ENR 25 and ENR 50 adhesive were investigated using coumarone-indene as the tackifying resin. Toluene was used as the solvent throughout the study. Result shows that peel strength increases with increase in silica loading due to the concentration effect of the filler. Peel strength, however, shows maximum value at 40 phr silica for both ENR 25 and ENR 50 an observation that is attributed to the maximum wettability and compatibility of adhesive on the substrate.

Keywords Adhesion; Adhesive; Epoxidized natural rubber; Peel strength; Silica

INTRODUCTION

Epoxidized natural rubber (ENR) is a chemically modified natural rubber. Adhesion is the bond strength measurement of a coating to a substrate. Adhesion testing is often associated with adhesives, sealants, laminates, electronics, cosmetics, medical device packaging, general packaging seal strength and applications where bond strength measurement is critical for research and quality control applications. Peel strength is generally used to measure the adhesive bond strength of a material, typically and adhesive. The demand for pressure sensitive adhesives (PSA) has been increasing over the last several years. PSAs are almost indispensable in everyday life as they are used for labels, tapes, films and other special adhesive applications such as bookbinding, upholstery, and pasting papers on the wall[1].

When silica is used as filler, the structure of filled rubber compounds on a molecular scale, in particular, is not very well known. Silica has a number of hydroxyl groups on the surface, which results in strong filler–filler interactions and adsorption of polar materials by hydrogen bonds[2–4]. Mixing of silica with rubber is a challenge, as these materials are not very compatible in various aspects: surface energy, solubility, structure of the filler and viscoeslastic properties. Silica has been used as an important reinforcing agent in a rubber compound together with carbon[2].

The curing characteristics and mechanical properties of epoxidized natural rubbers (ENR) have been widely studied[5–14]. Sung-Seen Choi et al.[15] studied the effect of filler–filler interaction on rheological behavior of natural rubber compounds filled with silica and found abnormal rheological behavior is due to the strong filler–filler interactions of silica since silica has a strong density of silanol groups on the surface and aggregates tightly by strong hydrogen bond. Ishak et al.[16] has studied the curing characteristics and mechanical properties of ENR with filler silica and has reported that cure characteristics could be related not only to the fillers characteristics, i.e., particle size, structure, and aspect ratio, but also to the filler–rubber interaction. All these studies involve the bulk property of ENR. The solution property of ENR, especially its use in pressure-sensitive adhesive (PSA) was not widely reported.

Recently, we have investigated the effect of zinc oxide[3] and kaolin[2] on the peel property of ENR-25-based pressure sensitive adhesive. To alleviate the dependency of silica on the dependence of peel adhesion of epoxidized natural rubber and the lack of scientific research in this field of interest, we have carried out a systematic research on the effect of silica on ENR on the peel strength of the adhesive in the presence of coumarone-indene resins.

EXPERIMENTAL

Materials

Epoxidized natural rubber, ENR-25 and ENR-50 having 25 mol% and 50% of epoxidation respectively were used as the elastomer for the preparation of the pressure sensitive adhesive. The rubbers were supplied by Rubber Research Institute of Malaysia. Silica was used as the filler, whereas toluene and polyethylene terephthalate (PET) film were chosen as the solvent and substrate respectively. Silica loading was varied from 10–50 parts per hundred parts of rubber (phr), whereas the coumarone-indene concentration

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Viscosity and Peel Strength of Magnesium Oxide-Filled Adhesive Prepared from Epoxidized Natural Rubber (ENR 25)

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Viscosity and peel strength of epoxidized natural rubber (ENR 25)-based adhesive was investigated in the presence of magnesium oxide. Petro resin and toluene were used as the tackifier and solvent, respectively. Viscosity was determined by a HAAKE Rotary Viscometer whereas peel strength was measured by a Lloyd Adhesion Tester. Results show that viscosity of adhesive increases with increasing magnesium oxide loading. However, for the peel strength, a maximum is obtained at 20 phr of magnesium oxide where maximum wettability and compatibility occurs. For a fixed filler concentration, peel strength increases with coating thickness for the three modes of peel tests.

Keywords Adhesive; Magnesium oxide; Peel strength; Viscosity

INTRODUCTION

Natural rubber has been widely used to prepare pressure-sensitive adhesives[1]. In formulating a rubber-based pressure-sensitive adhesive, an elastomer provides the elastic component while a low molecular weight tackifying resin imparts the viscous component. Kraus et al.[2–3] have studied the entanglement plateau and structural changes of pressure-sensitive adhesives prepared from styrene-diene block polymer-based pressure-sensitive adhesives. Leong et al.[4] has investigated the viscoelastic properties of natural rubber pressure-sensitive adhesive using acrylic resin as a tackifier. It is found that blends with good pressure-sensitive adhesives have higher loss tangent at higher frequencies.

Recently, we carried out several studies on the adhesion properties of natural rubber (NR) and epoxidized natural rubber (ENR)-based pressure-sensitive adhesives using coumarone-indene resin as the tackifying resin[5–8]. Results indicate that viscosity, tack and peel strength of the adhesive generally increases with an increase in tackifier concentration, an observation which is attributed to the better wettability of adhesive on the substrate with increasing tackifer loading. However, shear strength decreases gradually with increasing resin content. Addition of zinc oxide in the ENR-shows that peel strength passes through a maximum value at 30–40 phr of zinc oxide depending on the tackifier loading used[9]. Research involving the use of filler in rubber-based adhesives is not widely reported. Due to this scarcity of research data in this field of interest, it is thus the aim of this article to report some of our study on the viscosity and peel strength of ENR 25-based pressure-sensitive adhesives in the presence of magnesium oxide which acts as a filler in the adhesive system.

EXPERIMENTAL

Materials

Epoxidized natural rubber, ENR 25, having 25 mol% of epoxidation was used as the elastomer for the preparation of the pressure-sensitive adhesive. The rubber was supplied by Rubber Research Institute of Malaysia (RRIM). Petro resin, Nisseki Neopolymer grade 120 with softening point of 120°C and molecular weight of 1500 was chosen as the tackifying resin. Magnesium oxide and toluene were used as the filler and solvent respectively throughout the experiment.

Preparation of Adhesive

ENR 25 was masticated on a two-roll mill for 10 minutes to facilitate dissolution in the solvent. 5 g of the masticated rubber was then dissolved in 20 ml of toluene and the rubber solution was closed and kept for 24 hours to ensure complete dissolution of ENR 25. A fixed amount of 2 g of petro resin—corresponding to 40 phr of resin—was slowly added to the rubber solution. This was followed by the addition of five different weights, i.e., 0.5, 1, 1.5, 2, and 2.5 g – corresponding to 10, 20, 30, 40, and 50 phr – of magnesium oxide. One control sample without magnesium oxide was also prepared. The adhesive was left for 2 hours at room temperature (30°C) prior to testing.

Measurement

Viscosity. The viscosity of the adhesives was measured using a HAAKE Rotary Viscometer (Model PK 100) at room temperature (30°C). The spindle head (PK1;1°) was cleaned with acetone to prevent any impurities from affecting the viscosity results. It was then fixed to the viscometer.

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Dependence of adhesion properties of SMR L based adhesive on molecular weight of rubber

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The viscosity, loop tack, peel and shear strength of five different molecular weights of standard Malaysian rubber based pressure sensitive adhesive were studied. Coumarone–indene resin and toluene were used as the tackifier and solvent respectively throughout the experiment. The adhesive was coated on polyethylene terephthalate substrate at a coating thickness of 30, 60, 90 and 120 μm using a Sheen hand coater. Viscosity of the adhesive was determined by a Haake rotary viscometer whereas loop tack, shear and peel strength was measured by a Llyod adhesion tester. Results show that viscosity of adhesive increases with increasing molecular weight of rubber. However, loop tack, shear and peel strength exhibit maximum value at a molecular weight of $8{\cdot}5\times10^4$ where maximum wettability of adhesive on the substrate occurs. The adhesion property increases with coating thickness at this optimum molecular weight of rubber.

Keywords: Adhesive, Rubber, Coatings, Molecular weight

Introduction

Natural rubber has been widely used to prepare pressure sensitive adhesives. In order to produce the required balance of tack, peel adhesion and resistance to shear forces, the addition of tackifying resins to the rubber solution is required because natural rubber alone has a very low tack and adhesion to the surface of substrate.[1] The tackifying resin enhances the adhesion of non-polar elastomers by improving the wettability, increasing polarity, and altering the viscoelastic properties of the adhesive mass. However, systematic studies on the adhesion properties of rubber based adhesives were not extensively carried out. Kraus *et al.*[2,3] have studied the adhesive behaviour of the styrene–diene based pressure sensitive adhesives and the structural changes in melts of butadiene–styrene and isoprene–styrene block polymer based adhesives. Fujita *et al.*[4] have investigated the effects of miscibility and viscoelasticity on shear creep resistance of natural rubber based pressure sensitive adhesives. On the other hand, Leong *et al.*[5] have reported on the viscoelastic properties of natural rubber pressure sensitive adhesive using acrylic resin as a tackifier. Recently, the authors have reported the viscosity, tack, peel and shear strength of standard Malaysian rubber (SMR) based pressure sensitive adhesives.[6–8] Results have shown that viscosity, peel strength and tack of SMR based adhesives generally increases with an increase in coumarone–indene resin loading which acts as a tackifying resin. On the other hand, shear strength decreases gradually with increasing tackifier content. However, with respect to the effect of molecular weight of SMR L on the adhesion property of pressure sensitive adhesives using coumarone–indene resin as the tackifier, no study has been reported so far in this field. It is thus the aim of this paper to report the authors' findings on the effect of molecular weight of rubber on the viscosity, tack, shear and peel strength of SMR L based pressure sensitive adhesive.

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Experimental

Materials

Standard Malaysian rubber L grade natural rubber was used as the elastomer for the preparation of pressure sensitive adhesive. The technical specification of SMR L is given in Table 1. Coumarone–indene resin, freshly supplied by EuroChemo-Pharma Company (Malaysia), and commercial grade toluene was used as the tackifier and solvent respectively throughout the study.

Molecular weight determination

The molecular weight of the five masticated rubber samples was determined by viscometry. The method of determining the intrinsic viscosity $[\eta]$ as described by Billmeyer[9] was adopted in this study. The viscosity average molecular weight M_v of the rubber was calculated from the Mark–Houwink equation[10] as below

$$[\eta]=kM_v^a$$

where $k=5{\cdot}00\times10^{-4}$ dL g^{-1} and $a=0{\cdot}67$ in toluene.

Adhesive preparation

Five grams of each masticated rubber sample was dissolved in 30 mL of toluene. The rubber solution was left in a conditioned room for 24 h before adding 2 g of coumarone–indene resin which corresponds to 40 parts per hundred parts of rubber. The rubber solution

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Adhesion property of epoxidized natural rubber (ENR)-based adhesives containing calcium carbonate

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Abstract. The adhesion property (i.e. viscosity, loop tack and peel strength) of epoxidized natural rubber (ENR 25 and ENR 50 grade)-based pressure-sensitive adhesive was studied in the presence of calcium carbonate. The range of calcium carbonate loaded was from 10 to 50 parts per hundred parts of rubber (phr). Coumarone-indene resin was used as the tackifier and its concentration was fixed at 80 phr. Toluene was chosen as the solvent throughout the investigation. The substrates (PET film/paper) were coated with the adhesive using a SHEEN hand coater at a coating thickness of 60 μm. Viscosity of the adhesive was measured by a HAAKE Rotary Viscometer whereas loop tack and peel strength were determined by a Llyod Adhesion Tester operating at 30 cm/min. Results show that viscosity of ENR-based adhesives increases gradually with increase in calcium carbonate loading due to the concentration effect of the filler. However, for loop tack and peel strength, it passes through a maximum at 30 phr calcium carbonate, an observation which is attributed to the optimum wettability of adhesive on the substrate at this adhesive composition. ENR 25-based adhesive consistently exhibits higher adhesion property than ENR 50 for all calcium carbonate loadings studied.

***Keywords:** adhesion, calcium carbonate, coatings, rubber*

1. Introduction

Recently, we have carried out a systematic study of the viscosity, tack, peel and shear strength of natural rubber (SMR L, SMR 10 and SMR 20)-based pressure-sensitive adhesives [1–3]. The study shows that viscosity and tack of the adhesive increases with an increase in coumarone-indene resin loading. For the peel strength, it generally indicates an increasing trend with resin loading, an observation which is associated to the increasing wettability of adhesive on the substrate as tackifier is increased. However, shear strength decreases gradually with increasing resin content. Our recent study on the adhesion properties of SBR/SMR L-based adhesives indicates that the viscosity of adhesive decreases with increasing % SBR whereas loop tack passes through a maximum value at 20% SBR composition [4]. Leong *et al.* [5] on the other hand have reported the viscoelastic properties of natural rubber pressure-sensitive adhesive using acrylic resin as a tackifier. Higher loss tangent at higher frequencies is obtained for good pressure-sensitive adhesives. With respect to the adhesive prepared from epoxidized natural rubber, systematic investigation on its adhesion property is scarce. We have reported that peel strength of ENR-based adhesive passes through a maximum value at 40 phr coumarone-indene resin [6]. Also, a gradual drop of shear strength with increasing tackifier loading is also observed. Recently, we have carried out a study on the effect of zinc oxide on the viscosity, tack and peel strength of ENR 25-based pressure-sensitive adhesives [7]. It is shown that viscosity and loop tack of adhesive increases with

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Effect of Molecular Weight and Testing Rate on Adhesion Property of Pressure-Sensitive Adhesives Prepared from Epoxidized Natural Rubber (ENR 25)

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The dependence of peel strength and shear strength of epoxidized natural rubber (ENR-25)-based pressure sensitive adhesive on molecular weight and rate of testing was investigated using petro resin as the tackifying resin. Toluene and polyethylene terephthalate (PET) were chosen as the solvent and substrate respectively throughout the study. A SHEEN hand coater was used to coat the adhesive on the substrate at the coating thickness of 120 μm. All the adhesion properties were determined by a Llyod Adhesion Tester operating at different rates of testing. Result shows that peel strength and shear strength increases up to an optimum molecular weight of 6.5×10^4 of ENR 25. For peel strength, the observation is attributed to the combined effects of wettability and mechanical strength of rubber at the optimum molecular weight, whereas for the shear strength, it is ascribed to the increasing amount of adhesive present in the coating layer which enhances the shear resistance of the adhesive. Peel strength and shear strength also increases with increase in rate of testing, an observation which is associated to the viscoeslastic response of the adhesive.

Keywords: Molecular Weight, Rate of Testing, Adhesion, Epoxidized Natural Rubber

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BR01

ADHESION PROPERTIES OF SMR L-BASED PRESSURE-SENSITIVE ADHESIVE IN THE PRESENCE OF PETRO RESIN

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Viscosity, peel and shear strength of natural rubber (SMR-L grade)-based pressure-sensitive adhesive was studied by using petro resin as the tackifier. The resin concentration was varied from 0-80 parts per hundred of rubber (phr). Toluene was used as the solvent throughout the experiment. A SHEEN hand coater was used to coat the adhesive on substrate to give a coating thickness of 30, 60, 90 and 120μm. The viscosity of adhesive was determined by a HAAKE Rotary Viscometer whereas the peel and shear strength were measured by a Lloyd Adhesion Tester. Results show that the viscosity of the adhesive decreases with increasing petro resin concentration. For the peel strength, it increases up to 40 phr of resin concentration and drops with further tackifier loading. This observation is attributed to the wettability of the adhesive on the substrate. However, the shear strength shows a gradual decrease with increasing resin concentration and this observation is attributed to the decrease amount of rubber content which weakens the cohesive strength. In general, peel strength increases with increasing coating thickness while shear strength decreases with increasing coating thickness.

Keywords: Adhesion, adhesive, petro resin, rubber, coating

BORANG FRGS – P3(R)

Kod Projek	:	FRGS/FASA1-2009/(Fizik)/(USM)/(203/PFIZIK/67167)

FINAL REPORT
FUNDAMENTAL RESEARCH GRANT SCHEME (FRGS)

Laporan Akhir Skim Geran Penyelidikan Asas (FRGS) IPT

Pindaan 1/2009

A

RESEARCH TITLE : Dielectric and Dynamic Properties of hetero-structured Ferroelectric Systems
Tajuk Penyelidikan

PROJECT LEADER : Prof. Madya Dr. Ong Lye Hock
Ketua Projek

PROJECT MEMBERS (including GRA) *Ahli Projek* :
1. Lee Thong Yan (from July 2008)
2. Prof. Junaidah Osman (retired on 30 June 2008)

PROJECT ACHIEVEMENT *(Prestasi Projek)*

B

ACHIEVEMENT PERCENTAGE			
Project progress according to milestones achieved up to this period	0 - 50%	51 - 75%	76 - 100%
Percentage			80%

RESEARCH FINDINGS		
Number of articles/ manuscripts/ books	Indexed Journal	*Non-Indexed Journal*
	1	2
Paper presentations	International	*National*
	2	1
Others (Please specify)	A paper titled: "Phase Transitions Of Strained Barium Titanate Epitaxial Films" is submitted to Journal of Applied Physics.	

HUMAN CAPITAL DEVELOPMENT

Human Capital	Number		Others (Please specify):
	On-going	Graduated	
PhD Student			
Masters Student	1		
Undergraduate Students			
Temporary Research Officer			
Temporary Research Assistant			
Total	1		

EXPENDITURE *(Perbelanjaan)*

C

Budget Approved *(Peruntukan diluluskan)*	:	RM 61720.00
Amount Spent *(Jumlah Perbelanjaan)*	:	RM 61549.83
Balance *(Baki)*	:	RM 170.17
Percentage of Amount Spent *(Peratusan Belanja)*	:	99.72 %

ADDITIONAL RESEARCH ACTIVITIES THAT CONTRIBUTE TOWARDS DEVELOPING SOFT AND HARD SKILLS
(Aktiviti Penyelidikan Sampingan yang menyumbang kepada pembangunan kemahiran insaniah)

D

International

Activity	Date (Month, Year)	Organizer
1. Third International Meeting on Frontiers of Physics (IMFP 2009)	12-16th January 2009	Malaysian Institute of Physics and University of Malaya
2. 12th International Meeting on Ferroelectrics and 18th IEEE International Symposium on Application of Ferroelectrics (IMF-ISAF-2009)	23-27 August 2009	Xi'an Jiaotong University, China

National

Activity	Date (Month, Year)	Organizer
Persidangan Fizik Kebangsaan 2009 (PERFIK 2009)	December 2009	Universiti Teknologi Mara Kampus Shah Alam

PROBLEMS / CONSTRAINTS IF ANY *(Masalah/ Kekangan sekiranya ada)*

E

The problems that we faced in undergoing this project are:

i) We managed to get an R.O. who is also the master's degree student which is trained under the same project from July 2008. His progress is slow as he needs to be trained on the basic knowledge of ferroelectric as well as computational skills. Hence, in term of manpower utility, he has not been able to involve directly on the project. He is now almost ready to take part in the computational work. Prof. Junaidah Osman had retired in June 2008.

RECOMMENDATION (*Cadangan Penambahbaikan*)