International Journal of Adhesion & Adhesives 50 (2014) 65–69

Contents lists available at ScienceDirect

International Journal of Adhesion & Adhesives

journal homepage: www.elsevier.com/locate/ijadhadh

Release of a cohesively strong, general purpose hot-melt pressure sensitive adhesive from a silicone liner

Ahmed F. Moustafa [a,b,*]

[a] ETI Converting, 1490H Rue Nobel, Boucherville, Montreal, QC, Canada J4B 5H3
[b] National Research Center, Polymers Department, Tahrir St., Dokki, Cairo, Egypt

ARTICLE INFO

Article history:
Accepted 28 October 2013
Available online 27 December 2013

Keywords:
UV curable polysiloxanes
Label dispensing
Release build up
Hot melt PSA
Styrene isoprene styrene block copolymer
C5 Hydrocarbon resin
Rosin ester

ABSTRACT

Rubber based block copolymers styrene isoprene styrene (SIS) was used as rubber vehicles for the adhesive system. Wingtack ET (C5 hydrocarbon resin) in addition to rosin ester resin was the tackifier system. Naphthenic oil plasticizer was an additional processing aid of the adhesive system. Hydrophobic fumed nano-silica was used as flow hindering agent for the adhesives. The hotmelt adhesives were mixed in electrically heated sigma blender at 90 °C for 1 h. The adhesive system was coated with ETI Cohesio™ Machine at 177 °C onto Boise facestock at 20 g/m^2, and then laminated with Boise liner which was coated online with UV curable acrylated poly dimethyl siloxane (silicone) at 1.6 g/m^2. Laminates were cut and left to cool down and condition at room temperature for 48 h before testing. Thermally aged and humidity aged release forces were measured at 180° angle. Loop tack and 90° peel from corrugated card board surface were measured at room temperature. Dynamic mechanical thermal analysis was carried out from -100 to 100 °C at 1 Hz and 0.25% strain for formulations 5 and 6 which correspond to unacceptable and acceptable release force build up respectively.

1. Introduction

The force required to peel a label from its liner, is called release force or simply release, which is usually expressed as grams per inch or grams per 25 mm^2. When a hot melt adhesive is laminated with siliconzed liner; a weak van der Waal forces will be initiated causing the primary release value. As time passes the rubber polymers reorient, rotate then build stronger physical interactions with silicone surface. As the aging continues the polymers entangle across the relatively rough interface causing higher release values. Finally after prolonged contact time the physical interaction strengthen by numbers, or by direct chemical bonds developed between the rubber phase and the siliconzed liner surface causing scrap rolls of labels. The adhesive cohesive strength and functionality, silicone cure level, complete coverage and functionality, in addition to moisture absorption of facestock and liner, are the main factors which accelerate such kind of release build up. Other factors are playing important role, such as, silicone surface smoothness, silicone and/or adhesive surface contamination, and physical degradation of materials. The release force build up is not critical for label hand application, however seriously critical for machine automatic applications, once the release value goes above 20 g/25 mm^2, the label may not machine automatically released into the product substrate causing huge loss of materials and time.

Due to the nature of pressure sensitive hot melt adhesives [1,2]; which is lightly physically cross-linked by high and low T_g molecular domains; these crosslink sites become labile upon aging due to difference in cohesive energy density of adhesive ingredients. The molecules will continue to flow at high temperature and humidity, forming more bonds with the silicone surface [3,4]. This problem is not significant with acrylic adhesives due to higher elastic modulus and high cohesive molecular interactions within the material hindering the free flow of acrylic molecular chains [5,6].

Label converters when started to confront this technical problem, they thought that they can increase silicone coat weight from 1.5 to 2 g/m^2 to hide any site of future interaction with the rubber based adhesives, which never happened because of the difficulty of curing the UV acrylated silicone system to completion. Our industrial production studies showed that the highest possible cure level of silicone system is 83% due to radical disproportionation and increase of silicone oil viscosity upon crosslinking polymerization reaction which in turn decreases the radical mobility and probability of radical-double bond collision. This means that 17% of silicone is free to contaminate the adhesive surface and reduce adhesion properties in addition to reduction of label optical clarity. The moisture absorption of facestock and liner are affecting the release value because, heavier or different flexural rigidity [7] papers will have different micropeel angles from the liner. As shown in Fig. 1, the peel angle of label changes the release force

---

* Correspondence address: ETI Converting, 1490H Rue Nobel, Boucherville, Montreal, Canada J4B 5H3.
E-mail address: amoustafa1971@gmail.com

0143-7496/$ - see front matter © 2013 Elsevier Ltd. All rights reserved.
http://dx.doi.org/10.1016/j.ijadhadh.2013.12.014

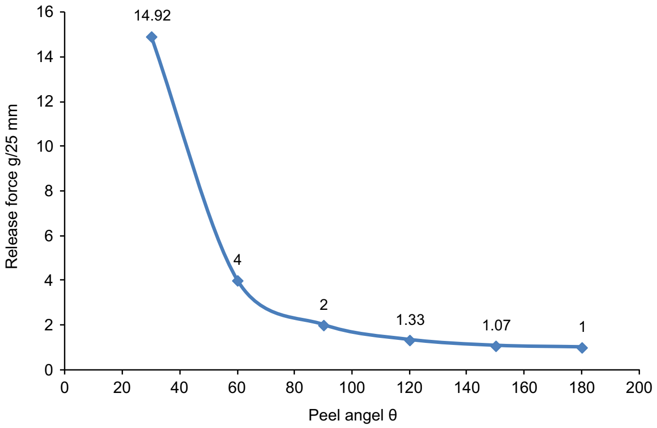

Fig. 1. Effect of peel angel onto release force.

according to Equation1 [8].

$$Ga = (1 - \cos\theta)Fr \tag{1}$$

where Ga is the adhesive failure energy, θ is the peel angle and Fr is the release force per unit width. Since the silicone cure level is limited and we cannot prevent moisture absorbance of paper at a reasonable price, i.e., apply protective paper coatings; the formulation of an adhesive which does not build up release force was the target of many adhesive companies.

The increase in release force at a given peel rate is due to an increase in the work of adhesion [9]. A contact mechanical approach based on the Johnson–Kendall–Roberts (JKR) theory [10] is currently used for the direct estimation of the work of adhesion and thus the surface free energy. The radius of a contact area a generated between an elastic spherical surface and a flat surface compressed by a normal force P is described by Eq. 2 [10].

$$a^3 = \frac{R}{K}\left\{P + 3\pi GR + \left[6\pi GPR + (3\pi GR)^2\right]^{1/2}\right\} \tag{2}$$

where R is the radius of curvature of the sphere, K is the elastic constant, and G is the energy release rate. If thermodynamic equilibrium conditions established by the initial contact of the surfaces are sustained over the course of the JKR experiment, the measured energy release rate during the loading stage GL corresponds to the work of adhesion.

In my study, i will eliminate all variables which affect the release values; keeping the same paper; Boise face and liner for all samples, standard silicone formulation at 1.6 g/m^2 and coat constant hot melt adhesive coat weight at 20 g/m^2.

2. Materials and methods

2.1. Styrene isoprene styrene block copolymer.

See Table 1.

2.2. Tackifiers

See Table 2.

2.3. Antioxidant, oil and filler

Antioxidant, Irganox 1010 (Pentaerythritol Tetrakis (3-(3, 5-di-tert-butyl-4-ydroxyphenyl) propionate) CAS No (6683-19-8) was supplied by Ciba Geigy (BASF) USA. Naphthenic oil (Solespec 500) was a product of Total, Exton, PA, USA, molecular weight 720, pour point −10 °C, color Gardner 5.5, and aniline point 124 °C. Silica was a product of Evonik, AEROSIL, Parsippany, NJ, USA. R 202 is fumed silica treated with a poly dimethyl siloxane, surface area 100 m^2/g and carbon content 3.5%.

Table 1
Physical properties of SIS and SBS block copolymers.

Physical property	Kraton D 1113
Tensile strength (MPa)	4
300% Modulus (MPa)	0.3
Hardness, Shore A, (10 s)^3	23
Melt flow index 10 min (200C/ 5Kg)	24
Styrene rubber ratio	16/84
Diblock %	55

Table 2
Physical properties of C5 hydrocarbon wingtack ET resin.

Physical property	Value
Appearence	Yellow flakes
Ash content (wt)%	0.0
Color gardner	2
FT-IR ratio (aromatic)	0.52
FT-IR ratio (olefinic)	0.32
Molecular weight, M_n (g/mol)	1000
Molecular weight, M_w (g/mol)	1600
Softening point (°C)	95
Specific gravity at 25 °C	0.96
T_g (midpoint) (°C)	50
T_g (onset) (°C)	44

2.4. Release and adhesion tester [11]

TMI release and adhesion tester, Testing Machine Inc., Ohio, USA was used for release and adhesion measurements at 180° and 90° respectively at 400 points/s data acquisition rate.

2.5. Loop tack tester [12]

Loop Tack tester LT 1000 was from Chemistruments, Mentor OH, USA. This compact tack testing machine meets the ASTM and TLMI specification.

2.6. Sigma blender

The NH-10D, ETI equipments, Shanghai Ltd., Shanghai, China. Electric horizontal heating kneading machine with two axes. It mainly consists of kneading body, driving system, heating system and electric control system. Blender mixing pot is filled with all SIS rubber and 10% weight of tackifier and antioxidant at 90 °C and 50 rpm. After 20 min the tackifiers and antioxidant were added portion wise over 60 min. Then the temperature is reduced to 75 °C with portion wise addition of filler and oil over 60 min. Finally the temperature is raised again to 100 °C to unload the batch into siliconized box.

2.7. Facestock and liner

Boise liner, 55 g/m^{2,} gloss 75%, Gurley porosity 2100 s/100 ml. Boise facestock, semi gloss, 80 g/m^2, gloss 75%, Boise Inc, Tillsonburg, ON, Canada.