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"D-optimal experimental design" analysis in preparing optimal polyisobutylene based pressure sensitive adhesives

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ABSTRACT

The aim of this work was to model the mechanical properties of different blends of polyisobutylene (PIB) pressure sensitive adhesives (PSAs) with different molecular weights (36,000, 51,000, 75,000, 400,000, and 1,100,000). The mechanical properties of PSAs are usually described by tack, peel, and shear strength which are strongly depended on the bulk viscoelastic properties of the adhesive system. It is assumed that the blends of high and low molecular weight PIB could affect these properties. According to D-optimal design of Design Expert software, various blends of five different molecular weights of PIB have been selected for study in this investigation. Using manual regression analysis, the quadratic model generated for three responses (tack, peel, and shear) was found to be statistically significant ($p < 0.05$). It was found that tack increases with decreasing molecular weight. It was also found that the presence of B10 and B12 as low molecular weight polymers and B50 as a high molecular weight polymer in increasing peel strength was more dominant compared with B15 and B100. Furthermore, shear strength was found to increase with an increasing concentration of the low molecular weight PIB, B15. The results have shown that statistical analysis meets theoretical expectations and they suggest desired blends of polymer for different aims.

1. Introduction

Pressure sensitive adhesives (PSAs) are viscoelastic materials that can adhere strongly to solid surfaces upon application of a light contact pressure for a short contact time. Abroad range of PSAs has been used in medical, cosmetic, and in labeling products, commercially. The performance characteristics of the PSAs such as drug flux and skin friendliness depends upon a balance of adhesive properties (tack, peel, and shear resistance), release force and cohesive strength as well as adhesive formulation. The balance between these properties must be changed according to the specific end use of the PSA. A PSA must be able to perform effectively under a wide range of temperature, humidity, and application frequency (from 24 h for some products to one week for others) conditions [1–3]. The viscoelastic properties of the bulk adhesive as well as the surface energies of the adhesive and adherent can affect the PSAs properties [4].

Tack is determined as the ability of an adhesive to instantaneously stick to a substrate under low pressure and remove by adhesive separation (without leaving any residue on the substrate surface) [5]. It is inversely proportional to elasticity modulus. It should be high enough that the adhesive surface achieves bonding from merely contacting and pressing to a substrate [6]. Tack is not a simple material's property such as density. It is a combined response of the chemical and physical properties of the adhesive bulk to stick to another material. Molecular weight, temperature and morphology also have significant effects on the tack behavior of polymers [7]. PSA tack and bond formation, ultimately involves molecular interactions at the adhesive/ adherent interface. The peeling characteristics of PSAs are determined by bulk rheological properties and the surface properties of the adherent as well as the mechanical properties of the substrate [3]. Also the adhesion properties are primarily influenced by the inherent properties of the polymer such as molecular weight [8], whereas shear strength depends on the miscibility and viscoelastic properties of the PSA [2].

Most PSAs are blends of numerous components including elastomers, tackifiers, plasticizers and fillers. Due to the complexity of these materials, it can be difficult to quantify the effect of a single variable such as molecular weight on the adhesive properties [9].

PSAs tend to be elastomeric in nature and the most widely used elastomer is natural rubber; although synthetic rubber, acrylic and silicone based PSAs are common. Polyisobutylene (PIB) as a synthetic rubber is the cheapest and simplest material to produce PSAs and is well-known for its excellent biostability and biocompatibility [10,11].

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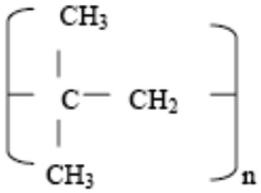

Scheme 1. Chemical structure of PIB.

PIB, which is a vinyl polymer made from the monomer isobutylene (IB) by cationic polymerization has the structure shown in Scheme 1.

PIB has unique properties: very low air, moisture, and gas permeability; good thermal and oxidative stability; chemical resistance and; high tack in adhesive formulations. PIB is a colorless to light-yellow, elastic, semisolid or viscous substance; odorless, tasteless, and nontoxic. Due to their highly paraffinic and non-polar nature, PIBs are soluble in aliphatic and aromatic hydrocarbon solvents and insoluble in polar solvents. Solubility generally decreases with increasing molecular weight of the polymer and increasing size of the aliphatic portion of the solvent molecule. PIB possesses great flexibility and is completely amorphous with a glass transition temperature of −65 °C. PIB high molecular weight grades are strong and elastic, however, low molecular weight PIBs are very soft and liquid-like making them suitable as tackifiers. The amorphous characteristics and low glass transition temperature of PIB impart high flexibility and permanent tack. Despite the favorable tack property, the adhesion of PIBs on many surfaces is weak because of their low polarity. PIBs are usually classified into two groups according to molecular weight. High molecular weight PIBs have a weight average molecular weight(Mw)of 500,000 to 1,100,000 g/mol, preferably between 650,000 and 850,000 g/mol and low molecular weight PIBs have a weight average between 40,000 and120,000 g/mol [5,8,10].

Numerous studies have investigated the adhesion properties of different blends of PIBs. Schulz et al. prepared a patch matrix containing crospovidone, acrylic adhesive and a blend of high, medium, and low molecular weight PIBs. They showed that crospovidone can improve the adhesive properties of PIB patches. Also, in vitro and in vivo investigations showed that ≤ 30% crospovidone content had no effect on the adhesive properties of the PIB patches [12]. O'Connor and Willenbacher focused on the effect of molecular weight and temperature on the adhesive behavior of blends of low and high molecular weight PIBs as model systems. They investigated cavitation and fibrillation on adhesive properties and showed that heterogeneities, for example air cavities, impair the strength of the adhesive bond by limiting contact [6]. Fujita et al. have investigated the effects of molecular weight on the peel strength and shear resistance of epoxidized natural rubber (ENR 25) based pressure sensitive adhesives. Results showed that peel strength and shear strength increases up to an optimum molecular weight of 6.5 * 10^4 of ENR 25 [2].

A limited number of studies have investigated the effect of different factors on adhesion properties by experimental design software. Taghizadeh et al. used a three level response surface methodology approach to evaluate the effect of different penetration enhancers on buprenorphine patches. They showed that levulinic acid was the best chemical skin enhancer [13]. Kajtna and Krajnc used experimental design methods to investigate the effect of four different acrylic PSA crosslinking agents on different PSA responses including tack, peel and shear strength [14]. Kardar et al. used a mixture method for experimental design and investigated the effect of the chemical structure of monomers on some of the physical and mechanical properties of resins and their films such as viscosity, glass transition temperature (T_g), hardness and scratch resistance [15]. Pichavant et al. published a study concerning the influence of composition and processing parameters on the properties of UV cured films. They used screening and quantification tools of experimental design methodology [16]. The selected methods were found to be a useful tool to minimize the number of experiments.

Table 1
Molecular weight of BASF PIBs.

	Trade name	Molecular weight
Low Molecular Weight	B10	36,000
	B12	51,000
	B15	75,000
Medium/High Molecular Weight	B50	400,000
	B100	1,100,000

In spite of the number of investigations conducted on the effects of various parameters on PIB adhesion properties, there have been no reports as far as we are aware on the use of experimental design software to predict the effect of PIB molecular weight on the tack, peel, and shear properties of PSAs. The objective of this work was to study the effect of molecular weight on the adhesive behavior of PIB- PSAs without any additives by D-optimal experimental design and to predict tack, peel, and shear properties.

2. Materials and methods

2.1. Materials

Different molecular weights of PIB(36000, 51000, 75000, 400000, and 1100000) were purchased from BASF AG (Ludwigshafen, Germany). The trade names of the different molecular weight PIBs are given in Table 1. CoTran 9705 as a backing layer was purchased from 3M Co. (St. Paul, MN, USA) and graft paper SC-45 as a liner layer was gifted from Bordar Shib Co. (Tehran, Iran).Toluene used for preparing adhesive solution was analytical grade, available from Merk (Darmstadt, Germany).

2.2. Formulation optimization of adhesives

D-optimal design of mixture from Design Expert software 7.1.3 (Version 7.1.3, Stat-Ease Inc., Minneapolis, MN, USA), was used to obtain different formulations. Five numerical factors (weight percent of B10, B12, B15, B50, and B100 in blends), three responses (tack, peel, and shear) and 5 replicating points were considered to obtain 25 runs. The amount of polymer with low and high molecular weight was considered to be 60–70% and 30–40% of total blend weight in the adhesive mixture, respectively. Table 2 shows the compositions of 25 different blends obtained by the software. Five run groups are replicating points: (run 1 and run 7), (run 4 and run 6), (run 10 and run 12), (run 3 and run 20), and (run 16 and run 22).

2.3. Film preparation

As indicated in Table 2, different blends of PIBs were prepared by dissolving PIBs in toluene with a rotator mixer (MIX-HTR-02, Shanghai, China) at room temperature for approximately 24 h. The adhesives were considered to be adequately prepared when the PIBs were completely dissolved in toluene and the solution was visually homogeneous. The adhesive film was prepared by solvent evaporation. An Elcometer-3580SPRl casting knife film applicator (Manchester, UK) was used to obtain a uniform thickness layer adhesive on a liner layer and maintained at room temperature for 20 min. The dried film was then placed in an oven at 60 °C for 1 h to remove residual solvent. The dried film was laminated onto a baking layer using a standard 4.5 kg roller. Finally, adhesive thickness was measured by a Micrometer Mitutoyo 156-101 (Mitutoyo, Japan) [17].