Journal of Adhesion Science and Technology

ISSN: 0169-4243 (Print) 1568-5616 (Online) Journal homepage: http://www.tandfonline.com/loi/tast20

Adhesion properties of eco-friendly PVAc emulsion adhesive using nonphthalate plasticizer

Youn-Mee Choi , Byoung-Ho Lee , Ji-Won Park , Hyun-Joong Kim , Young Geun Eom , Sung-Wook Jang & Young-Kyu Lee

To cite this article: Youn-Mee Choi , Byoung-Ho Lee , Ji-Won Park , Hyun-Joong Kim , Young Geun Eom , Sung-Wook Jang & Young-Kyu Lee (2013) Adhesion properties of eco-friendly PVAc emulsion adhesive using nonphthalate plasticizer, Journal of Adhesion Science and Technology, 27:5-6, 536-550, DOI: 10.1080/01694243.2012.705481

To link to this article: https://doi.org/10.1080/01694243.2012.705481

Published online: 09 Aug 2012.

Journal of Adhesion Science and Technology, 2013
Vol. 27, Nos. 5–6, 536–550, http://dx.doi.org/10.1080/01694243.2012.705481

Adhesion properties of eco-friendly PVAc emulsion adhesive using nonphthalate plasticizer

Youn-Mee Choi[a,b], Byoung-Ho Lee[b], Ji-Won Park[b], Hyun-Joong Kim[b]*, Young Geun Eom[c]*, Sung-Wook Jang[a] and Young-Kyu Lee[d]

[a]*R&D Center, OKONG Corp, Incheon 402-023, Republic of Korea;* [b]*Laboratory of Adhesion & Bio-Composites, Program in Environmental Materials Science, Seoul National University, Seoul 151-921, Republic of Korea;* [c]*Department of Forest Products & Biotechnology, College of Forest Science, Kookmin University, Seoul 136-702, Republic of Korea;* [d]*National Instrumentation Center for Environmental Management, Seoul National University, Seoul 151–921, Republic of Korea*

(*Received 30 November 2011; final version received 8 February 2012*)

An eco-friendly poly (vinyl acetate) emulsion adhesive was synthesized without phthalate. Four types of eco-friendly plasticizers for use in these adhesives were selected to confirm their primary properties by injecting the eco-friendly plasticizer without any prior change to its processing or cost. The four types of eco-friendly plasticizers used were dibutyl phthalate DBP-based product, dialkyl ester, acetyl tributyl citrate, and pentandiol-di-isobutyrate. Their properties were determined by comparison with the existing (DBP)-based product. As a result, an emulsion adhesive was produced without addition of phthalate or need for additional additives, resulting in a significant decrease in cost. However, the low temperature characteristics of the eco-friendly plasticizers were slightly inferior to those of DBP. These adhesives containing eco-friendly plasticizer were studied and their characteristics for adhesion strength, water resistance, ability for low temperature film formation, excellent storage stability, and lack of volatile organic compounds productions (including phthalate) were confirmed.

Keywords: poly (vinyl acetate); nonphthalate; plasticizer; eco-friendly adhesive; low temperature film formation

1. Introduction

Adhesives have a wide range of applications and poly (vinyl acetate) (PVAc) emulsion adhesive formulations are affordable and are regarded as being nontoxic. In addition, they are easy to handle. PVAc resin, which was first manufactured in Germany in 1924, is widely used in adhesives, paints, and film resins due to its excellent viscosity properties, even though it cannot be used as a forming material because its glass transition temperature is close to room temperature [1]. The characteristics of emulsion adhesives are determined not only by the polymerized monomer, but are also greatly affected by various additives which are selected based on their type, quantity, and the need as a protective colloid, surface active agent, or catalyst. In particular, the type and quantity of an added plasticizer have a significant effect on the viscosity of an adhesive. In addition to their use in wood products such as furniture,

*Corresponding authors. Email: hjokim@snu.ac.kr

building materials, musical instruments, and toys, adhesives are also used for bookbinding and nonwoven fabrics. Adhesives copolymerized with ester acrylate are used for bonding soft vinyl sheets to plywood or paper and for joining vinyl faced paper [2]. PVAc emulsions are mainly used in woodworking and paper packaging. However, the properties of PVAc emulsions are substantially temperature dependent with worsening viscosity and usability at low temperatures during winter. Accordingly, phthalate plasticizers have been used to overcome these drawbacks.

The flexibility of a PVAc adhesive can be controlled by external plasticization, normally by addition of dibutyl phthalate, tritolyl phosphate, or mixtures of these compounds, even though a wide variety of plasticizers are available. These plasticizers are normally stirred directly into the hot PVAc emulsion, where they become absorbed into the emulsion particles. Plasticization with increasing solids content tends to increase the viscosity. Although the addition of a plasticizer will reduce the T_g (glass transition temperature), the addition of small amounts (~5%) will often increase film transparency at low temperature. On the other hand, it will also increase ‘creep,’ even though this can be avoided to some extent by increasing the molecular weight to a maximum permissible level [3]. However, the addition of a phthalate plasticizer can weaken the adhesion strength and decrease the heat resistance, and has the problem of increase the dosage to volume of use in the winter. In addition, it also stimulates an increase in the viscosity of the product, which decreases the product’s usability and delays hardening. Furthermore, it also provokes separation and precipitation of the product, which worsens product’s stability during storage.

Plasticizers have been used for centuries to improve the processability and flexibility of a variety of materials. At the dawn of civilization, water was used to plasticize clay for the production of pottery and clay tablets. Today, the most common plasticizers are phthalates, adipates, and ester derivatives; the majority of which are used to produce flexible poly (vinyl chloride) (PVC) products. The widespread use of plasticizers in these applications over the last 40 years has resulted in their toxicology profiles being investigated and understood. As a consequence of these investigations, many official bodies and legislative authorities have concluded that plasticizers pose no significant hazards to humans or the environment. Plasticizers will, therefore, continue to be used to improve the quality of life through the production of a wide variety of PVC items ranging from medical components to waterproof clothes and floor coverings [4].

Various phthalic acid esters (PAEs) are used in a wide range of products. However, PAEs and their metabolites produce reproductive and developmental toxicity in laboratory animals. These findings have raised concerns regarding the possibility of PAEs contributing to adverse reproductive and developmental effects in humans [5].

Building and finishing materials and consumer products are important sources of formaldehyde and other volatile organic compounds (VOCs) as well as endocrine (phthalate, alkyl phenol ethoxylate, etc.) in the indoor environment. There is increasing concern regarding the effects of human exposure to these indoor pollutants. The slow release of VOCs and endocrine agent from polymeric materials can affect the performance and durability of products as well as adversely affect the indoor air quality (IAQ) and ‘well being’ of the occupants [6].

In this study, an environment-friendly PVAc emulsion was produced using an environment-friendly plasticizer rather than a phthalate, to generate a nonphthalate PVAc emulsion.

Products made from this experiment results also showed that it meets all the other spec with the improved basic adhesion properties. The products were then evaluated for their characteristics at low temperatures, adhesive properties, water resistance, film transparency, storage stability, and stability of the PVAc emulsion without phthalate.

2. Experimental

2.1. Materials

PVAc was obtained from 88% saponification (P24, P20, OCI Chemical, Korea) with the polymerization of a 2400, 2000; emulsifier, polyoxyethylene nonylphenyl ether (NP1050, Dongnam, Korea). Vinyl acetate (Samsung BP, Korea) with a purity of 99.5% was used as the monomer. Dibutyl phthalate (DBP, Junsei Chemical, Japan) and ammonium persulfate (APS, 98.0%, Junsei Chemical, Japan) were used as the plasticizer and initiator, respectively. Diethylene glycol n-butyl ether (DEGBE) was purchased from Junsei Chemical. Table 1 lists the six different eco-friendly plasticizers used in this study.

2.1.1. PVAc synthesis

PVAc was synthesized using the following process: Water (330 mL) was placed into a reactor equipped with a stirrer, followed by addition of 13 g of PVA P20, 9 g of P24, and 0.7 g of the emulsifier (HLB = 17.8). The reaction mixture was heated to 80 °C and stirred at 130 rpm to dissolve the contents. After complete dissolution, APS was added to the reaction vessel while 220 g of vinyl acetate monomer was added drop-wise over a 4 h period. The contents were then cooled to a temperature of $\leqslant$50 °C.

2.1.2. Blends of nonphthalate plasticizers with PVAc

Six different samples were prepared using the nonphthalate plasticizers. The blending process was similar to that used in the synthesis method reported in Section 2.1.1. The following nonphthalate plasticizers were then added: (4-cyclohexane 1,2-dicarboxyl acid dialkyl ester), (diethylene glycol dibenzoate, dipropylene glycol dibenzoate, blend), (dipropylene glycol dibenzoate [DPGDB]), (diethylene glycol ester), (acetyl tributyl citrate), and (2,2,4-tri-1,3-pentandiol diisbutyrate). These samples are subsequently referred to as EFP-1, EFP-2, EFP-3, EFP-4, EFP-5, and EFP-6, respectively.

2.2. Methods

2.2.1. Physical property measurements

A viscometer (Brookfield RV-1+) was used for the viscosity measurement and the measurement temperature was set to 25 °C. The solids content (%) was determined by drying the mixture at 150 °C for 30 min and weighing the residue.

Table 1. Composition of products (nonphthalate plasticizers).

Notation	Chemical name	Boiling point (°C)	Freezing point (°C)	Solubility in water
DBP	Dibutyl phthalate	340	−35	0.04%
EFP-1	4-Cyclohexane 1,2-dicarboxyl acid dialkyl ester			
EFP-2	Diethylene glycol dibenzoate/dipropylene glycol dibenzoate blend	232–234	<16	Insoluble
EFP-3	Dipropylene glycol dibenzoate (DPGDB)	232	−40	<0.01%
EFP-4	Diethylene glycol ester			
EFP-5	Acetyl tributyl citrate (ATBC)	410		Insoluble
EFP-6	2,2,4-Tri-1,1,3-pentandioldiisobutyrate	280	−40	0.42%

The particle size was determined using a laser diffraction particle size analyzer (Shimadzu SALD 1100). The size of the microparticles was determined in paraffin oil (viscosity 5 mPa s) as a nondissolving dispersion medium. The particles were suspended by saponification and magnetic stirring during the measurements.

2.2.2. *Test sample preparation for adhesion strength*

Wood timber with a size of 25 mm × 30 mm and a specific gravity of ⩾ 0.5, which had been dried to a moisture content of 15%, was used as the test sample. Approximately 100 g/m^2 of premixed adhesive was applied to the timber, which was then compressed for 10 min with a load of 5–10 kg/cm^2 and left to stand at 20 ± 2 °C for 24 h. After removing the pressure, the sample was left to stand for 48 h and tested for its wet adhesion strength.

2.2.3. *Adhesion strength*

The lab shear strength of the test sample was measured using a universal test machine (UTM, Shimadzu AG-2000A). A total of 360 specimens were subjected to the shear test in a 10-ton UTM in accordance with ASTM D 905-98 standard. The loading speed of the machine during the experiment was adjusted to 12.7 mm/min. The load at the moment of separation (breaking off) of the parts from each other was read from the dial and recorded. The values obtained were used in the following equation to calculate the shear strength of each piece.

$$\sigma_M = \frac{P_{max}}{A}$$

where σ_M is the maximum shear strength (N/mm^2), P_{max} is the maximum load at breaking point (N), and A is the bond surface area (mm^2).

2.2.4. *Wet adhesion strength*

After dipping the test sample in water at 20 ± 1 °C for 10 min, followed by 3 h immersion in water at 30 ± 1 °C, the compression shear strength was checked using a universal test machine (UTM, Shimadzu AG-2000A).

2.2.5. *Minimum film forming temperature*

To demonstrate the adhesive strength of a water-based adhesive (emulsion type), the adhesive needs to be bonded to the substrate by forming a film as the water evaporates. However, whitening of the film is frequently observed in winter because the water is not volatilized and is trapped in the film. Therefore, the adhesive strength of a water-based adhesive is closely related to the temperature of the adhesive. This whitening condition appears at low temperatures where water is not easily volatilized and begins to freeze, which is a limitation of water-based adhesives. The minimum film forming temperature (MFT) is an important criterion that determines whether a water-based adhesive can demonstrate its adhesive strength by forming a film at a specific low temperature. Previously, it was reported [3] that a resin emulsion with a low T_g has superior low-temperature MFT properties.

After applying the resin emulsion to one side of the sample on a clean glass slide, a 0.1–0.3 mm thick uniform film was produced by drying it at a 4 °C. The dried sample was then examined to determine if the film had whitened.

2.2.6. Differential scanning calorimetry (DSC)

The glass transition (T_g) values of the blends were determined by DSC with a TA Instruments Q-1000 equipment, at a scan rate of 10 °C/min over a temperature range from −50 to 100 °C. T_g was determined from the second scan. The T_g was taken as the midpoint of the 2nd transition region.

2.2.7. VOC emissions

The main chamber was made of stainless steel (SUS304) and contained an air control unit. Although there are larger chambers, the 20 L chamber was used because it has been standardized in Korea. The air control system consisted of an air supply unit, humidifier, and pumps. The 20 L chamber was set up in a temperature-controlled climate chamber. Purified air was used for ventilation. A stainless steel seal box was used to prevent the cut edge effect, which allowed chemical emission only from one surface of the test piece. When two of these seal boxes were used, the total surface area and loading was 0.044 m^2 and 2.2 m^2/m^3, respectively.

Before setting up the chamber and seal boxes, they were washed with water and baked in an oven at 260 °C to eliminate any contamination from the chamber itself. The 20 L small chamber was supplied with purified and humidified air at a given ventilation rate. The temperature and relative humidity inside the chamber were constant at 25 ± 1 °C and 50 ± 5%, respectively. The chamber was ventilated at 0.5/h. Aldehydes were analyzed by high performance liquid chromatography (HPLC), and thermo desorption system (TDS)/gas chromatograph–mass spectrometer (GC–MS) was used for analysis of the VOCs. Total volatile organic carbon (TVOC) was defined by converting the areas of the peaks between C6 and C16 to concentrations using the toluene response factor. A peak area of <10 was defined as the detection limit. The sample gas was taken by Tenax TA and a 2,4-DNPH cartridge seven days after the sample specimens had been placed into the 20 L small chamber, according to the regulations of the Ministry of Environment, Korea.

3. Results and discussion

3.1. Properties of PVAc blended with nonphthalate plasticizers

3.1.1. General properties

The addition of plasticizers to an emulsion has the effect of slightly increasing the particle size. It is seldom necessary to prepare an emulsion system independent of plasticizer. However, some plasticizers, e.g. tritolyl phosphate, enter the polymer particles much more slowly than, for example, dibutyl phthalate. Various proprietary plasticizers, usually the polyester type, may be included in PVAc compositions, especially when a particular property is required, e.g. the absence of migration into the substrate [7].

The plasticizing efficiency of ethylene is quite high: ∼3-fold that of 2-ethylhexyl acrylate on a weight basis. Because of this, only low proportions of ethylene, ∼5% on the basis of vinyl acetate, are needed to obtain flexible copolymer films at ambient temperature [7].

A wide range of compatible plasticizers have been used. For many years, triethylene glycol-bis (2-ethyl butyrate) was the universally used plasticizer for poly (vinyl butyral).

Recently, this has been supplanted by adipates tetraethylene glycol derivates, butyl sebacate, ricinoleates, and others. Rahman and Brazel summarize the major categories of plasticizers, giving specific examples and some common end uses for plasticized materials [8].

Six types of eco-friendly plasticizers were selected in this study and EFP-6 (2,2,4-tri-methyl-1,3-pentandiol diisbutyrate) had a higher volatility compared to DBP because it had 60% of the S.C.(solid contents), which was used as a scale to measure the properties of the adhesive. EFP-5 (acetyl tributyl citrate) is sometimes used in toys in Europe and other countries, but it is very expensive.

Figure 1 shows the solids content of each plasticizer alone and Figure 2 shows the solids contents after adding each type of plasticizer into the PVAc adhesive. EFP-6 demonstrated the highest volatility when in the emulsion, which confirmed its effect on the solids content. Volatility is one of the basic properties of a product which may cause the generation of volatile organic compounds, which is used as a barometer to measure the environmentally-friendly nature of a compound or product.

The T_g value indicates that the low temperature film forming characteristic of EFP-1 is slightly better than those of DBP, EFP-2, and EFP-3, similar to level of EFP-5 and EFP-6 and much higher for EFP-4.

Experiments were carried out using 4 kinds of eco-friendly plasticizers except EFP-6 (low solid content) and EFP-5 (high cost)

3.1.2. Low temperature film formation

For a water-based adhesive (emulsion type) to demonstrate its adhesive strength, it needs to be bonded to the adherend by forming a film as water evaporates. However, whitening is frequently observed when the adhesive is used in winter. This defect occurs when water is not volatilized and is trapped in the film [7].

The formation of a film from a solution is a simple process. The evaporation of the solvent causes the liquid film, which is applied by a brush, sprayer, roller, or by dipping, to form a concentrate, resulting in a gradual increase in viscosity until all the solvent has evaporated. At this point, the characteristic properties are obtained depending on the nature of the polymer that is dissolved.

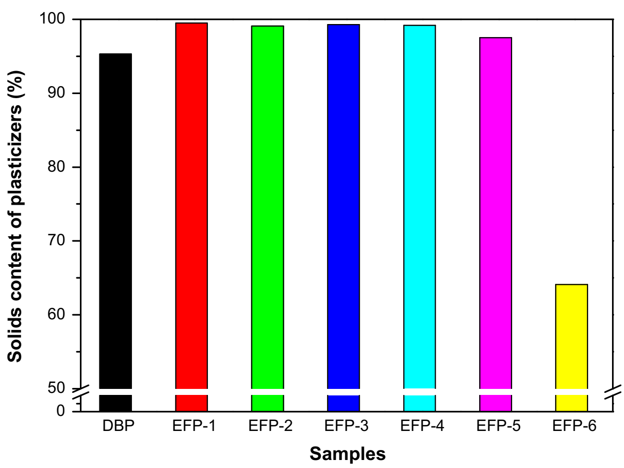

Figure 1. Solids content of each plasticizer.

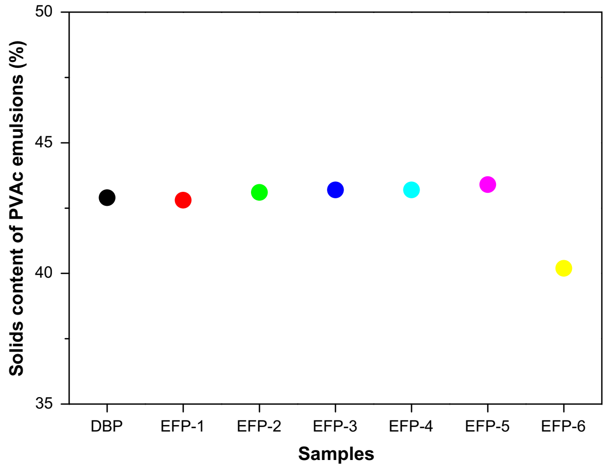

Figure 2. Solids content after adding each type of plasticizer to the adhesive.

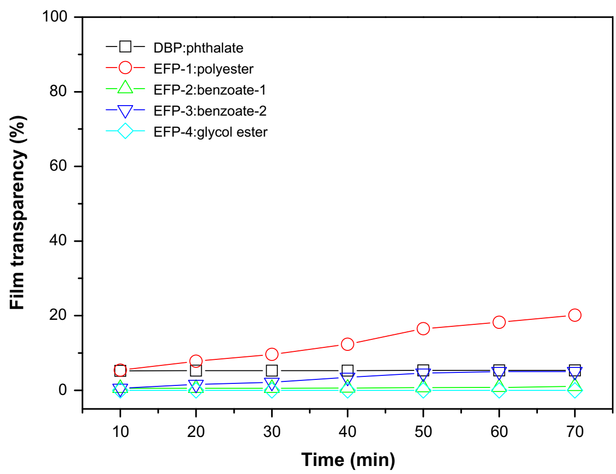

Figure 3. Transparency of PVAc film as a function of time after using 8% nonphthalate plasticizers.

If a PVAc emulsion is plasticized with dibutyl phthalate, micro photography has shown that the plasticizer is absorbed into the polymer before 30 min of stirring. This process takes much longer tritolyl phosphate is the preferred plasticizer. Therefore, film formation is rapid for most plasticized PVAc emulsion products [3].

A concentration of ~5% of plasticizer based on the weight of polymer is desirable for obtaining sufficient plasticization for a continuous film formation at ambient temperature. Accordingly, PVAc emulsion adhesives are often plasticized to a slight degree to increase their film strength and adhesive strength, even though this might cause a slight increase in cold flow [7].

Figures 3 and 4 show the film transparency at each temperature, to display the low temperature properties that result when combining the different contents of each eco-friendly plasticizer. Measurements of the low temperature film transparency for the amount of plasti-

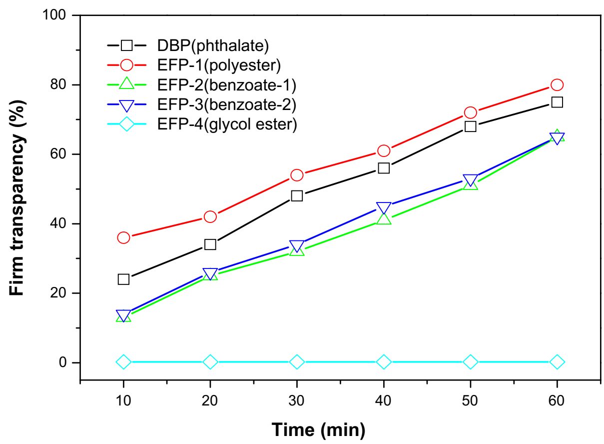

Figure 4. Transparency of PVAc film as a function of time after using 12% nonphthalate plasticizers.

cizer, for each plasticizer type, revealed EFP-1 to have a lower MFT, resulting in an outstanding low temperature film forming ability. However, other eco-friendly plasticizers were shown to have lower temperature film capability than DBP. The possibility of a defect in winter was confirmed when using the eco-friendly plasticizers alone. In particular, the eco-friendly plasticizer, EFP-4, would have poor low temperature film characteristics with a very unclear state, even if the plasticizer content increased.

Usually, the T_g of nonplasticized PVAc is ~35–40 °C, but will be lowered if a plasticizer is added. Greater amounts of plasticizer produced lower T_g values, but it is not possible to make PVAc with a T_g of unlimited low value. When T_g decreases, minimum film forming temperature (MFT) will also decrease. It appears that an eco-friendly plasticizer less capable of forming a film because, if the same amounts of DBP and eco-friendly plasticizer are used, the eco-friendly plasticizer is less effective in decreasing T_g than is DBP. Low temperature film formation is one of the important roles of a plasticizer. For an eco-friendly plasticizer, however, due to lack of its capability of plasticization, which helps to decrease the T_g, low temperature film formation is more difficult than that with DBP. Therefore, there is a need for additives to supplement low temperature film characteristics. A significant improvement was achieved when even a small amount of diethylene glycol n-butyl ether (DEGBE) additive was added. In addition, a small amount of glycol ester additive was added to each of the eco-friendly plasticizers and their low temperature properties were measured.

Figures 5 and 6 show the low temperature film transparency at 4 °C in the incubator, after coating the PVAc emulsion with each eco-friendly plasticizer on a glass plate. In the case of the existing DBP, it was confirmed that the transparency of the film improved without the whitening phenomenon with the addition of 8 or 12% plasticizer, based on the vinyl acetate monomer.

Among the eco-friendly plasticizers used, EFP-1 showed better characteristics than did DBP. However, the other products showed poorer low temperature film formation at lower temperatures than did DBP when added at concentrations even up to 12%. In particular, the whitening phenomenon with EFP-4 was quite severe. The low temperature whitening phenomenon associated with eco-friendly adhesives might deteriorate adhesion due to a lack of drying if the temperature at the worksite or temperature of the adherend is low (i.e. in winter).

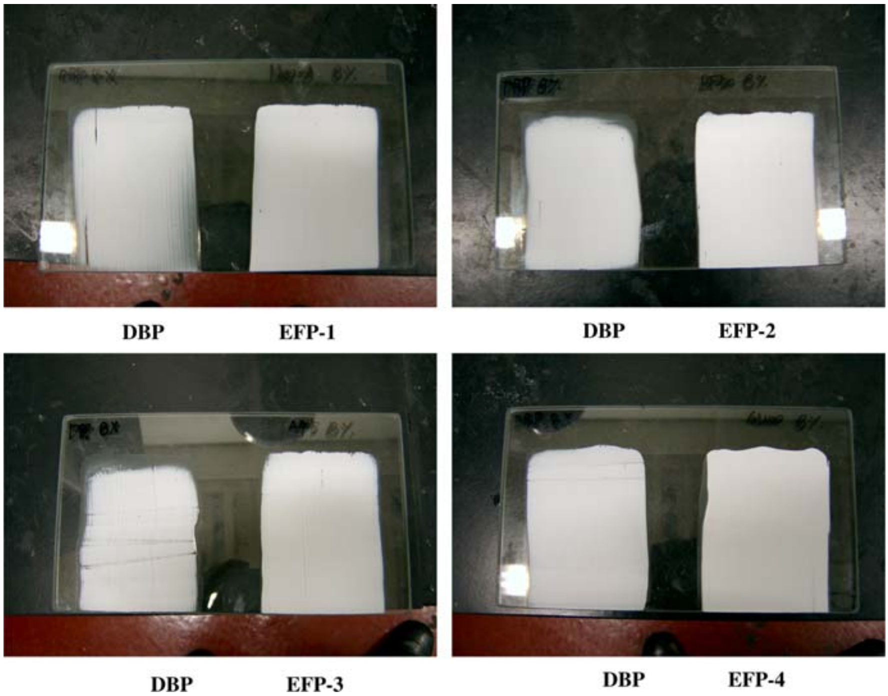

Figure 5. PVAc film transparency using 8% DBP and nonphthalate plasticizers at 4 °C.

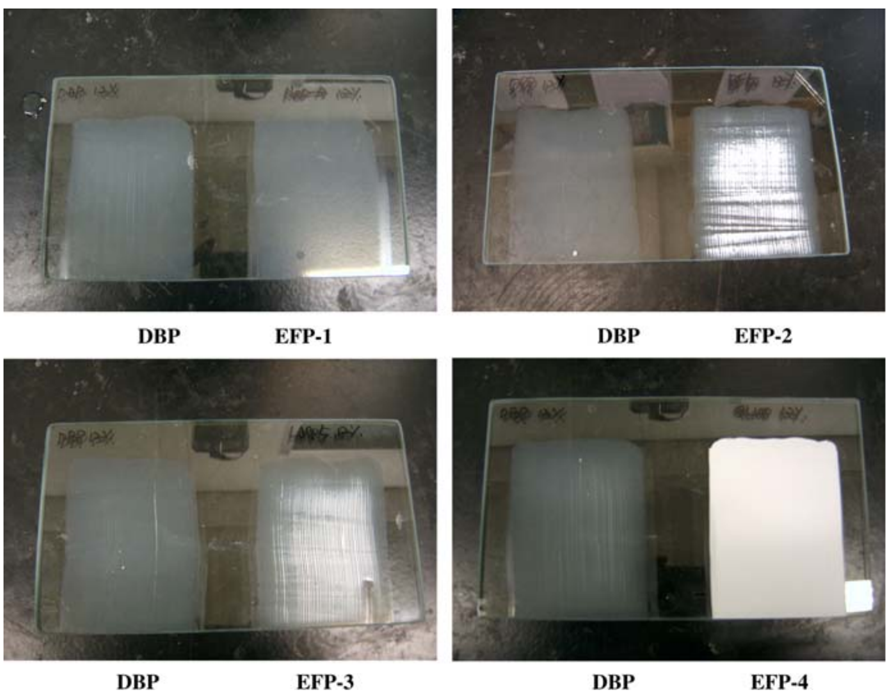

Figure 6. PVAc film transparency using 12% DBP and nonphthalate plasticizers at 4 °C.

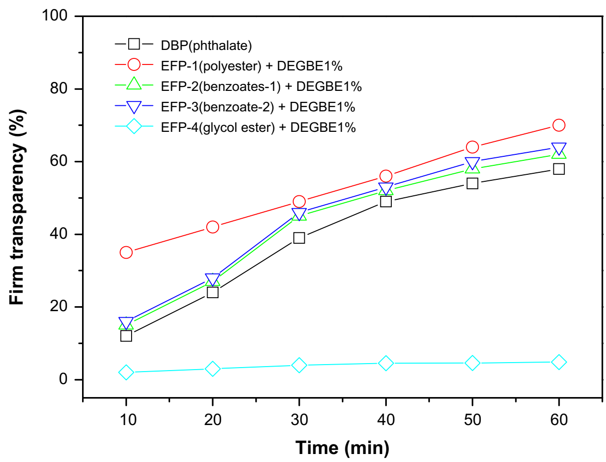

Figure 7. PVAc film transparency as a function of time after using 7% of the plasticizer and 1% of DEGBE.

3.1.3. *Low temperature properties using the auxiliary additive*

Figures 7 and 8 present the film transparency when using the auxiliary additive in the eco-friendly plasticizer. Because of applying several adhesives to improve the film formation ability at low temperature, the low temperature characteristics improved in accordance with the small addition of the glycol ester derivate or the glycol ether derivate. However, the low temperature film characteristics did not improve in the case of the eco-friendly plasticizer EFP-4, making it unsuitable for use in a PVAc emulsion.

3.1.4. *Adhesion strength*

Adhesion strength is the most frequently used indicator for changes caused by plasticization. As the plasticizer must plasticize the polymer, the tensile strength of the plasticized material

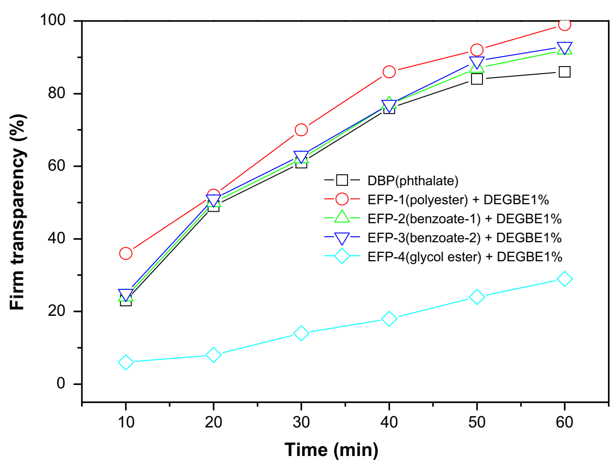

Figure 8. PVAc film transparency as a function of time after using 11% of plasticizer and 1% of DEGBE.

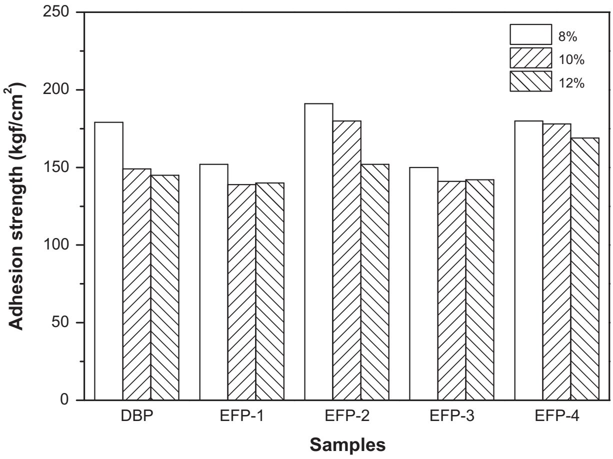

Figure 9. Comparison of the adhesion strength of PVAc emulsion adhesive using different plasticizer types and contents (at 25 °C).

should decrease with increasing amounts of the plasticizer. In addition, the hardness decreases with increasing amounts of plasticizer [9].

While external plasticization is a simple process, in many cases, an adhesive film may embrittle and fail due to plasticizer migration into the substrate. This is particularly the case when PVAc in the plasticized state is used as an adhesive for a ‘plastic’ film. In other cases, with a synthetic resin film, which is used as a laminating component, plasticizer migration softens the film and causes a loss of adhesion strength [3].

When substrates are flexible, it may be used to improve the adhesion and match the characteristics of the adhesive film to more closely resemble those of the substrate, particularly where this is flexible [10].

In Figures 9 and 10, when a comparison is made with DBP for each type of eco-friendly plasticizer, the usage compared to the VAM used was made different to display the adhesion

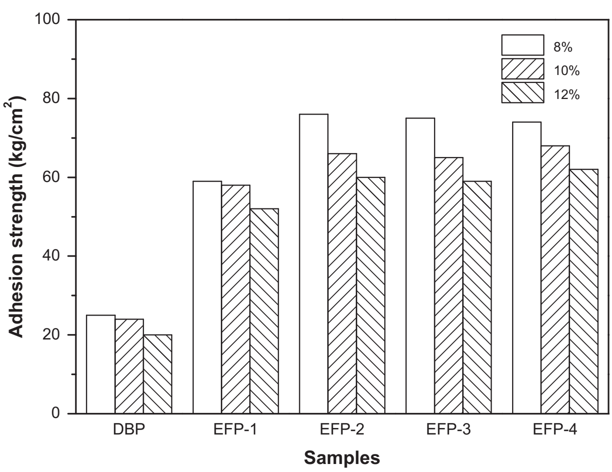

Figure 10. Comparison of the wet adhesion strength of PVAc emulsion adhesive using different plasticizer types and contents (at 25 °C).

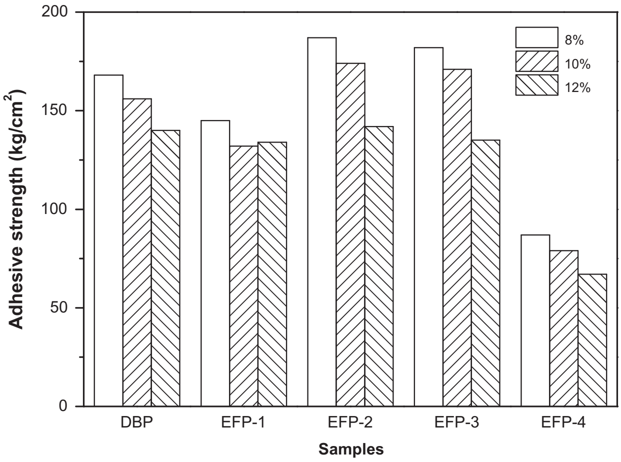

Figure 11. Comparison of the adhesion strength at low temperature (2 °C) using different plasticizer types and contents.

strength and wet adhesion strength. In the case of this eco-friendly plasticizer, there was almost no decrease in adhesion strength, and the eco-friendly plasticizer in EFP-2 and EFP-4 showed higher adhesion strength than that with DBP. In the case of wet adhesion strength of the PVAc adhesive, most of the eco-friendly plasticizers were superior to DBP.

Figure 11 shows that the low temperature adhesion strength of the lower measurement value for EFP-1 resulted in the most outstanding low temperature film capability, and EFP-4 exhibited poor low temperature film transparency with lower low temperature film capability, but outstanding wet adhesion.

Adhesion strength can be viewed when particles of the emulsion fuse to form a film. Film formation results were confirmed with film transparency. Measurements of low temperature adhesion strength showed that nontransparent particles cannot perform as an adhesive because these particles hardened without forming a film. PVAc without plasticizer has weak adhesion strength because of its lack of film formation capability. Adding a certain amount of plasticizer decreases the T_g below the room temperature and helps in film formation; as a result, low temperature adhesion strength is increased. However, an excess amount of plasticizer will decrease the hardness and cause poor adhesion strength. Except for EFP-4, eco-friendly plasticizers, which do not decrease T_g, show the same adhesion strength as resulted after adding plasticizers such as DBP. Addition of supplementary additives can be used to improve the poor low temperature film formation capability, and low temperature characteristics and low temperature adhesion strength can also be improved.

3.1.5. Adhesion strength after addition of supplementary additive

Figures 12 and 13 show the adhesion strength of adhesives according to plasticizer type and content in normal/wet states. The low temperature characteristics were clearly improved by the supplementary additive, but the normal status and degree of wet strength decreased slightly. Nevertheless, with the exception of EFP-1, the adhesives showed outstanding adhesion strength compared to DBP.

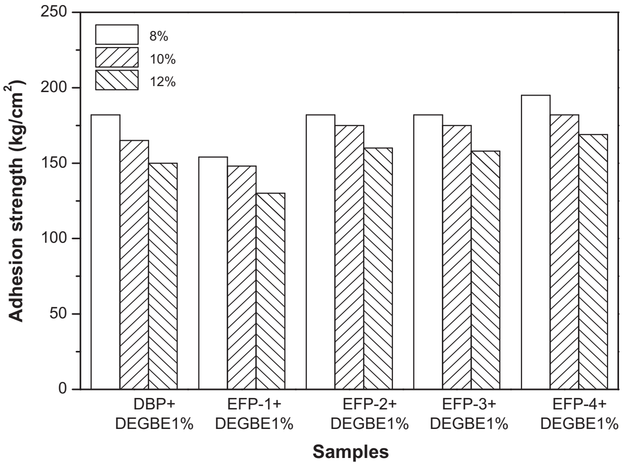

Figure 12. Comparison of the adhesion strength of PVAc emulsion adhesive using different plasticizer types and contents (when adding 1% DEGBE, at 25 °C).

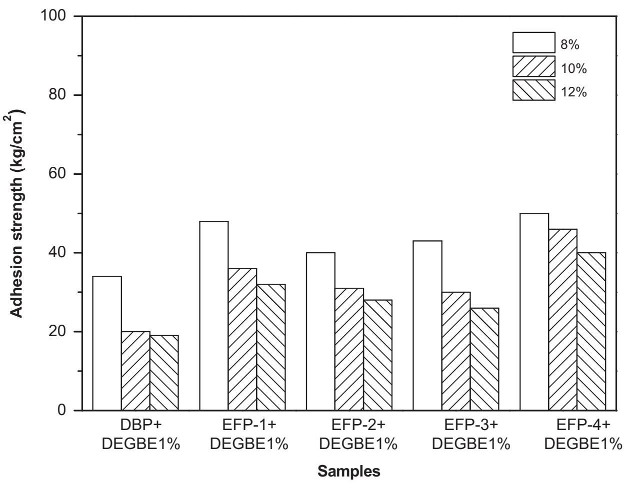

Figure 13. Comparison of the wet adhesion strength of PVAc emulsion adhesive using different plasticizer types and contents (and adding 1% DEGBE).

3.1.6. Storage stability

The viscosity of general wood adhesives is normally −5 to 20 mPa s, but such adhesives can be diluted, or a higher viscosity can be used to suit particular application machinery [7].

Figure 14 shows the storage stability of PVAc emulsion with each plasticizer. When combined with the eco-friendly plasticizer, the PVAc emulsion showed no change in viscosity when stored for a long period of time, and there was no layer separation detected or problems with the storage stability.

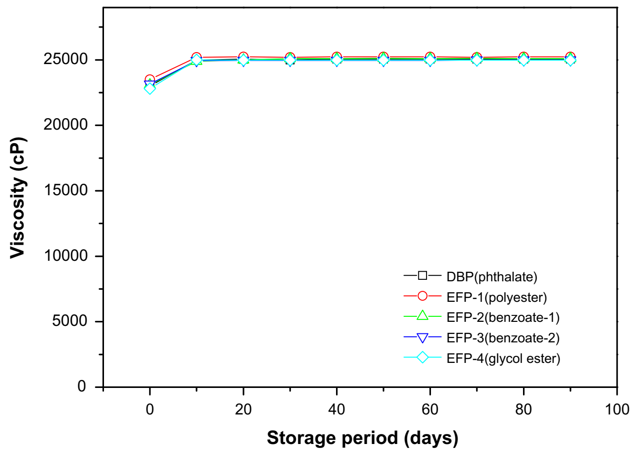

Figure 14. Comparison of the viscosity on storage of PVAc emulsions using the different plasticizers.

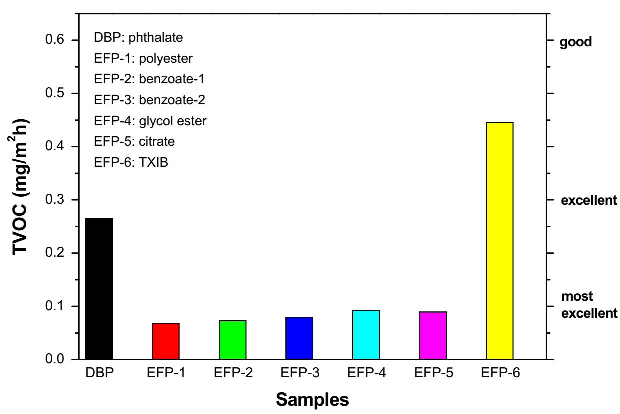

Figure 15. Comparison of TVOC emissions from PVAc emulsions containing different plasticizers.

3.2. Comparison of TVOC emission of the nonplasticizer type and the eco-friendly plasticizer

Emission testing is performed to assess the risk of exposure, and to determine the adverse effect on human health that result from exposure to hazardous pollutants in indoor environments [11].

Figure 15 shows the TVOC emissions. The TVOC emission of a PVAc emulsion that used an eco-friendly plasticizer was compared with that of an emulsion using DBP. The eco-friendly plasticizer type was confirmed to be an environment-friendly product when used at concentrations up to 12% for all products, with the exception of EFP-6 which had been excluded because of its low boiling point.

4. Conclusion

Several eco-friendly plasticizers were evaluated for their potential use in adhesives. A polymerization method using PVAc without DBP was examined to reduce the harm caused by the use of phthalates. In addition, the reduction of adhesion strength and the water resistance were assessed after an addition to reduce the whitening phenomenon observed at low temperatures.

When the eco-friendly plasticizers were used, some products had insufficient solids content, were costly, and showed very poor low temperature film capability. However, EFP-1 showed outstanding low temperature characteristics, even though the adhesion strength had deteriorated slightly.

EFP-2, 3, and 4 exhibited useful properties such as good adhesion and an eco-friendly nature, but showed poor low temperature characteristics compared to DBP. In particular, EFP-4 had a clear problem with its low temperature adhesion strength.

A small amount of glycol ester additive was used to overcome these shortcomings. When EFP-2 and EFP-3 were used, the low temperature characteristics improved, but this was not the case for EFP-4. On using the additive, the low temperature characteristics were not affected, but the adhesion strength decreased slightly compared to that in case of DBP, which is not considered to be a major problem.

In conclusion the eco-friendly plasticizers could potentially be used in industrial adhesives without increasing costs, if an additive is used to supplement this type of plasticizer. This eco-friendly water-based product is a potential replacement for phthalate, which is prohibited for use in the EU and other countries.

Acknowledgments

This study was supported by the research program of the Agricultural R&D Promotion Center. And also by the research program of Kookmin University in Korea.

References

[1] Pizzi A. Wood adhesives: chemistry and technology. New York (NY): Marcel Dekker; 1983.
[2] Skeist I. Handbook of adhesives. New York (NY): Van Nostrand Reinhold; 1990.
[3] Warson H. The applications of synthetic resin emulsions. London (UK): Ernest Benn Ltd; 1972.
[4] Cadogan D. Vinyl Technology. 1991;13:104.
[5] Matsumoto M, Hirata-Koizumi M, Ema M. Regulatory Toxicology Pharmacology. 2008;50:37.
[6] Kim S, Kim J, An J, Kim H. Indoor Air. 2007;17:404.
[7] Warson H, Finch C. Applications of synthetic resin latices: latices in diverse applications. New York (NY): Wiley; 2001.
[8] Rahman M, Brazel C. Progress in Polymer Science. 2004;29:1223.
[9] Wypych G. Handbook of plasticizers. Toronto (ON): ChemTec Publishing; 2004.
[10] Pizzi A, Mittal K. Handbook of adhesive technology. 2nd ed. New York (NY): Marcel Dekker; 2003.
[11] Anderson E, Albert R. Risk assessment and indoor air quality. Boca Raton (FL): CRC Press; 1999.