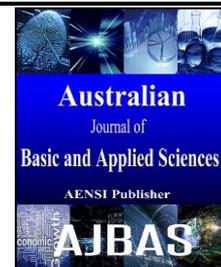




## AUSTRALIAN JOURNAL OF BASIC AND APPLIED SCIENCES

ISSN:1991-8178 EISSN: 2309-8414  
Journal home page: www.ajbasweb.com



### The formulation and characterization of hot melt adhesive from ethyl vinyl acetate for packaging industry

Mohammad Khairushany Abd Rashid, Mohd Aizudin Abd Aziz, Anwaruddin Hisyam, Rosli Mohd Yunus, Musfakri Musa, Zulkifly Jemaat

Faculty of Chemical Engineering & Natural Resources, Universiti Malaysia Pahang, 26300 Gambang, Pahang, Malaysia.

#### Address For Correspondence:

Mohammad Khairushany Abd Rashid, Faculty of Chemical Engineering & Natural Resources, Universiti Malaysia Pahang, 26300 Gambang, Pahang, Malaysia.

#### ARTICLE INFO

##### Article history:

Received 18 September 2016

Accepted 21 January 2017

Available online 26 January 2017

##### Keywords:

Hot Melt Adhesive, Ethyl Vinyl Acetate, formulation, packaging industry

#### ABSTRACT

In recent years, packaging industry prefers to use hot melt adhesive based on polyolefin due to the fact that polyolefin provides ease of processing, low off-taste, low smell and heat-seal ability. This research is intended to develop the formulation for Ethylene Vinyl Acetate (EVA)-based hot melt adhesive with the same properties of polyolefin-based hot melt adhesive (HMA) since EVA offers lower cost than polyolefin. In developing the EVA HMA, the EVA copolymers with various melt index were blended together with resin, wax and antioxidant in the molten state. The performance of the new formulations were compared with commercial polyolefin HMA. Its glass transition ( $T_g$ ) and melt viscosity were measured by using differential scanning calorimeter (DSC) and Brookfield Viscometer. The results indicated that three new formulations and commercial polyolefin had a glass transition temperature between  $-20\text{ }^\circ\text{C}$  and  $-25\text{ }^\circ\text{C}$ , which implied that the EVA copolymer blended with the resin was compatible. The peak observed on the heating curve corresponded to the melting point ( $T_m$ ). EVA-HMA 1 and EVA-HMA 3 were comparable to the commercial polyolefin based on identical broad peak. Furthermore, the melt viscosity of all EVA-HMA formulation and commercial polyolefin HMA decreased with an increasing temperature. Hot melt adhesive with lower viscosity would have more flexibility to flow out, indicating that it had better wetting properties when applied to the substrate. It was concluded that EVA-HMA 3 had the best performance for the packaging industry because of comparable thermal properties and viscosity at desired temperature with commercial polyolefin adhesive.

#### INTRODUCTION

Hot melt adhesives are used widely in our country because it ability to enable high speed manufacturing and their cost advantages (Jefrey;2015). The hot-melt adhesives have been in the market for a long time and they are used in a wide range of applications such as for bookbinding, packaging, footwear and wood industries (Y J Park; 2005). The most composition in manufacturing of hot melt adhesive basically it blended of four basic component : 1. Polymer ( Homopolymer or Copolymers) are used to provide the mechanical properties and also function as a backbone in the adhesive.2. Tackifiers are used to order to control the viscosity of the formulation and increase the adhesion to the surfaces. The hydrocarbon resins, which are used as tackifiers in HMAs, could be classified according to whether they contain primarily aromatic, aliphatic, or dicyclopentadiene (DCPD) monomers. These are also referred to as C9, C5, and C10, respectively, corresponding to the average number of carbon atoms per monomer molecule.

3. Wax that are low molecular weight are used because of to perform the crystallization rate in order to increase the setting speed and it also low cost. 4. Antioxidant are used in a small percentage other specific

#### Open Access Journal

Published BY AENSI Publication

© 2017 AENSI Publisher All rights reserved

This work is licensed under the Creative Commons Attribution International License (CC BY).

<http://creativecommons.org/licenses/by/4.0/>



Open Access

**To Cite This Article:** Mohammad Khairushany Abd Rashid, Mohd Aizudin Abd Aziz, Anwaruddin Hisyam, Rosli Mohd Yunus, Musfakri Musa, Zulkifly Jemaat., The formulation and characterization of hot melt adhesive from ethyl vinyl acetate for packaging industry. *Aust. J. Basic & Appl. Sci.*, 11(3): 182-186, 2017

objectives, i.e. shelf life and stability. A typical hindered phenol primary antioxidant is AO-1 (2,6-di-tert-butyl-para-cresol). AO-1 is widely being used as an antioxidant due to its lower cost. In addition, this antioxidant show higher activity, compatibility and resistance towards the formation of colored byproduct during compounding and application temperature of hot melt adhesives (Pizzi and Mittal, 2003) The properties of the hot-melt adhesives can be modified and customized to a specific end usage by adding these materials accordingly. The diluent system can help to lower the viscosity of the molten polymer and thus making it more convenient to be applied onto surfaces as well as to increase its wetting ability and adhesive strength. It can also provide molten tack and modify the physical properties of the hot-melt adhesives. However, the components need to be selected with care to ensure the compatibility of the whole system in order for the diluent system to be effective (Tse, 1988). The hot melt adhesive with lower viscosity will have more flexibility to flow out, which makes it easier for the adhesive to fully wet the substrate.

This provides an opportunity for the adhesive material molecules to orient themselves onto the substrate surface, hence forming a bond with the substrate. An adhesive with higher molten viscosity will be harder to flow out and completely wet the substrate, hence the adhesive material molecules will have less chance to orient themselves onto the substrate surface to form a bond. Ideally, at the application temperature, the viscosity of the molten adhesive should stay low long enough for the substrate to be brought into contact and then rise rapidly to set the band in minimum time (Macosko, 1977). The glass transition temperature ( $T_g$ ) is being used to determine the compatibility of the polymer blends. It has usually been associated with the onset of segmental mobility in the amorphous phase of an amorphous or semi crystalline polymer (Chen *et al* 2001). Open time is an important concept in the adhesive industry. Open time can be defined as the time lapse between applying the molten adhesive onto the substrate and the transformation of the molten adhesive to the solid state, at which point the hot melt adhesive loses its ability to wet the substrate (Cagle, 1973).

## MATERIALS AND METHOD

Three types of ethylene vinyl acetate were used in this study. Table 1 below shows some characteristics of the EVA copolymers, such as density and melting point.

**Table 1:** Properties of the EVA copolymers

Code	Density (g/cm <sup>3</sup> )	Melting Point (°C)
EVA- 1	0.95	63
EVA-2	0.95	66
EVA-3	0.95	68

Three types of resin were used in this study. Table 2 below shows the softening point and molecular weight of the tackifier.

**Table 2:** Properties of resin

Code	Softening Point (°C)	Molecular weight (g/mol)
Resin 1	105.0	670
Resin 2	124.0	660
Resin 3	103.0	800

The wax was used in this study to reduce the high viscosity of the polymer and resin to ensure efficient mixing. In order to reduce the thermal degradation of HMAs during melting process, 0.4 parts by weight of the antioxidant were used as a thermal stabilizer. So as to compare the performance of the EVA hot melt adhesive with the commercial adhesive, a type of polyolefin hot melt adhesive was used in this study.

### **Preparation of the EVA hot melt Adhesive:**

For the preparation of the EVA hot melt adhesive, the items needed were hot plate, stirrer, Pyrex glass beaker and glove as protection from the hot surface. At first, the ethyl vinyl acetate copolymer and antioxidant were simultaneously added in a Pyrex glass beaker and placed on the hot plate. The temperature was set at around 170°C - 180°C. The mixture was allowed to melt completely before the tackifier or resin was added. When the tackifier and EVA became masticated and formed a homogeneous melt, the wax was slowly added until completion. Lastly, the EVA hot melt adhesive was poured on a silicon paper and allowed to solidify under ambient temperature. The EVA copolymer, tackifier and wax were blended with various formulations.

### **Melt viscosity:**

A Brookfield Viscometer Model DV-II+ with a Brookfield Thermosel System was used to measure melt viscosity, which allowed the measurements under controlled high temperature. Viscosity was obtained at temperatures of 120°C, 130°C, 140°C, 150°C and 160°C. A SC4-21 spindle at a rotational speed of 3 RPM was used to measure a viscosity range of 200–5,000,000 cP

### Glass Transition Temperature:

The Differential Scanning Calorimetry (DSC) was used to assess the thermal characteristics of the adhesives. A TA Q200 DSC using standard aluminum pans and lids were used to perform the experiments. The cell environment was purged with nitrogen. The first samples at room temperature were cooled to  $-70^{\circ}\text{C}$ , and then heated to  $200^{\circ}\text{C}$  at  $10^{\circ}\text{C}/\text{min}$ . They were kept at this temperature for 2 min. Then, the samples were cooled to  $-70^{\circ}\text{C}$  and scanned to  $200^{\circ}\text{C}$  at  $10^{\circ}\text{C}/\text{min}$ . The first heating treatment removed the material's processing history; recrystallization and melting temperatures were evaluated from the heat flow signals observed during the cooling and second heating cycle respectively. The glass transition temperature ( $T_g$ ) values were identified from the DSC curves obtained in the second heating run. (Lee *et al.*, 2010).

## RESULTS AND DISCUSSION

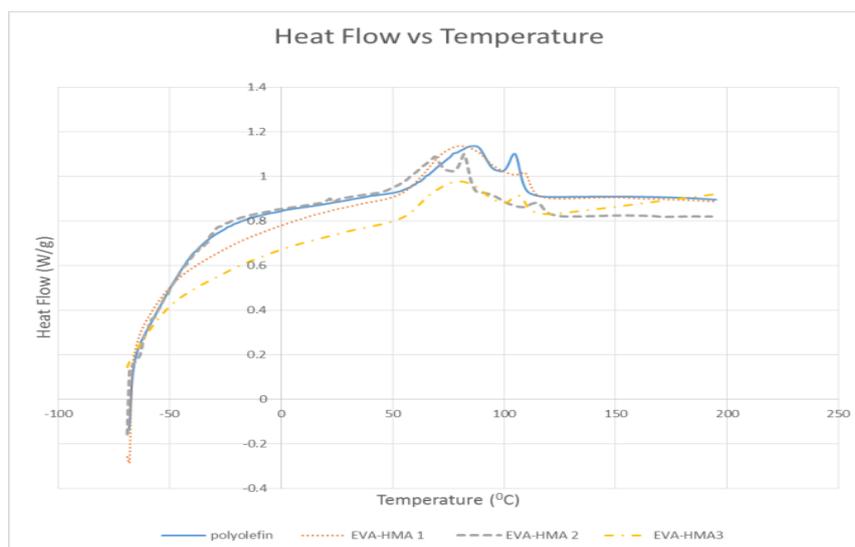
### Formulation of the EVA Hot Melt Adhesive

**Table 3:** Formulation of the EVA hot melt adhesive (weight percent)

Code	EVA-HMA 1	EVA-HMA 2	EVA-HMA 3
EVA 1	25	30.4	25.4
EVA 2	14.6	11	11
EVA 3	-	-	5.0
Resin 1	15.0	30.0	30.0
Resin 2	15.0	8.1	8.1
Resin 3	15.0	2.1	2.1
Wax	15.0	18.0	18
Antioxidant	0.4	0.4	0.4

### Differential scanning calorimeter:

The glass transition temperature usually take the middle of the incline line. It has usually been associated with the onset of segmental mobility in the amorphous phase of an amorphous or semi crystalline polymer (Chen *et. al.*, 2001).



**Fig. 1:** DSC Curve

**Table 4:** Melting Temperature

Adhesive	Melting Temperature 1, $T_m$ ( $^{\circ}\text{C}$ )	Melting Temperature 2, $T_m$ ( $^{\circ}\text{C}$ )
Polyolefin (commercial)	87	105
EVA-HMA 1	81	109
EVA-HMA 2	70	82
EVA-HMA 3	80	106

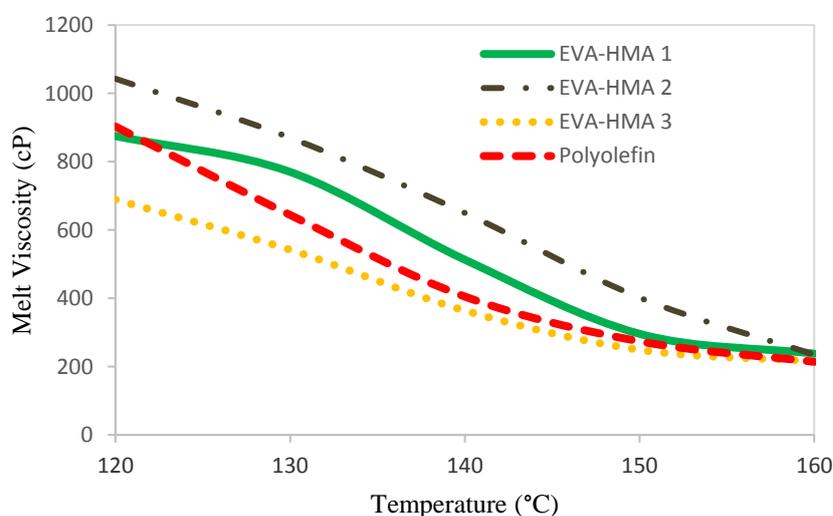
The thermal properties of EVA adhesive and polyolefin (commercial) were determined using modulated DSC. The curve in Figure 1 shows glass transition temperature,  $T_g$  and melting point,  $T_m$ . From DSC curve, the result indicated that three new formulations and commercial polyolefin has a glass transition temperature between  $-30^{\circ}\text{C}$  and  $-35^{\circ}\text{C}$ .

The glass transition temperature usually takes the middle of the incline line (Makromolekülen, 2015). The glass transition temperature was related with resin used to blend the EVA adhesive. According Park and Kim in

journal hot melt adhesive properties of EVA/aromatic resin blend showed the result of glass transition temperature is between  $-25^{\circ}\text{C}$  and  $-30^{\circ}\text{C}$ . Implying, the result  $T_g$  of the new formulation was acceptable and the blend of four materials was compatible. The peak observed on the heating curve corresponded to the melting point,  $T_m$ . It can be seen, all the four adhesive had different thermal behavior from each other. From the curve, all the adhesive had two peak points. Melting point 1 was used as the result.

In general, melting point of EVA adhesive was affected by vinyl acetate content of EVA copolymers, but less affected by melting index (MI) of EVA copolymers (Arsac *et al*, 1999). EVA-HMA 2 gave sharp peak indicating that the thermal event occurred in a short temperature span while EVA-HMA 1, EVA-HMA 3 and commercial polyolefin adhesive gave similar broad and shallow peak. It indicates that the thermal occurred in a long temperature span. The width and height of the peak observed from heat flow curve seemed to reflect the open time range suggested for each adhesive supplier (Le, 2011). Open time can be defined as the time lapse between applying the molten adhesive onto the substrate and the transformation to the solid state, at which point the hot melt adhesive loses its ability to wet the substrate (Cagle, 1973). EVA-HMA 2 deemed to have short open time because it has sharp peak. EVA-HMA 1 and EVA-HMA 3 is comparable with commercial adhesive base on the peak, it is supposed to have a long open time. It means the EVA-HMA 1 and EVA-HMA 3 is suitable to do EVA hot melt adhesive for packaging industry based on the thermal properties.

### Viscosity:



**Fig. 2:** Melt viscosity vs. Temperature

For the manufacture of hot melt adhesive, the materials used need to be sufficiently stable both during storage and processing as well as during their application. In particular, the most important of hot melt adhesive is thermal stability. Figure 2 shows the melt viscosity of three formulations of EVA hot melt adhesive and commercial polyolefin hot melt adhesive. A decrease of melt viscosity means thermal degradation of the adhesive. The increase of viscosity at the application temperature develop problems for the end user of the adhesive, resulting in unscheduled interruptions in production, because of plugged transfer lines and applicators (Colegrove *et al*, 1998). Usually, melt viscosity of EVA hot melt adhesive was affected by molecular weight of EVA copolymer, vinyl acetate and concentration (Qian *et al*, 1997).

As can be seen in Figure 2, at the beginning of measurement, the viscosity is slightly decreasing and lastly became stabilized. The melt viscosity decrease with increasing temperature and softening point of the adhesive also decrease (Park and Kim, 2003). EVA-HMA 1 and EVA-HMA 2 shows a relatively high melt viscosity compared with commercial polyolefin hot melt adhesive. EVA-HMA 2 shows the highest melt viscosity because the weight percent of EVA copolymer use EVA-HMA 2 is highest compared other formulations. It proved that the molecular weight of EVA copolymer will affect the melt viscosity. A rubbery polymer provides the elastic component, while the low molecular weight of resin constitutes a viscous component (Gierenz and Karman, 2001). Thus, the melt viscosity decrease as the content of weight percent increase use in the formulation. From the table 3, EVA-HMA 1 have a lower range of weight percent of resin compared to EVA-HMA 3. It proved base of the graph, EVA-HMA 3 has low melt viscosity compared to EVA-HMA 1. Based on the graph, EVA-HMA 3 has lower melt viscosity compared to the commercial polyolefin hot melt adhesive at temperature  $120^{\circ}\text{C}$  until temperature  $140^{\circ}\text{C}$  and similar melt viscosity at temperature  $140^{\circ}\text{C}$  -  $160^{\circ}\text{C}$ . The melt viscosity depends on the application.

Hot melt adhesive with lower viscosity will have more flexibility to flow out, which makes it easier for the

adhesive to fully wet the substrate. This provides an opportunity for the adhesive material molecules orient themselves onto the substrate surface, hence forming a bond with the substrate. Adhesive with higher molten viscosity will have a harder time to flow out and completely wet the substrate, hence the adhesive material molecules will have less chance to orient themselves onto the substrate surface to form a bond. Ideally, at application temperature, the viscosity for molten adhesive should stay low long enough for the substrate to be brought into contact and then rise rapidly to set the bond in minimum time (Macosko, 1977). Hence, EVA-HMA 3 can use as hot melt adhesive for the packaging due to melt viscosity.

### Conclusion:

The performance of three formulations of EVA hot melt adhesive and commercial polyolefin hot melt adhesives were characterized by DSC and Viscometer. All the three formulations of EVA hot melt adhesive have same glass transition temperature with commercial polyolefin. It proves the four materials used to produce EVA hot melt adhesive is compatible. EVA-HMA 1 and EVA-HMA 3 have a similar broad peak with commercial polyolefin, it indicates the hot melt adhesive has a long open time. The melt viscosity of all hot melt adhesive decrease with increasing temperature. The melt viscosity Formulation 3 is matched with the melt viscosity of commercial polyolefin at temperature 140 °C to 160 °C. Thus, from the three formulations, Formulation 3 have the best performance to be made the hot melt adhesive for the packaging industry because it's comparable for thermal properties and viscosity at a certain temperature. Further research needs to be conducted on EVA-HMA 3 for other properties to confirm its viability in replacing the commercial polyolefin based hot melt adhesive in packaging industry.

### ACKNOWLEDGEMENT

PLS INCLUDE ACKNOWLEDGEMENT

### REFERENCES

- Arsac, A., C. Carrot and J. Guillet, 1999. Rheological characterization of ethylene vinyl acetate copolymer, *Journal of Applied Polymers Science*, 74.
- Chen, X., H. Zhong, L. Jia, J. Ning, R. Tang, J. Qiao and Z. Zhang, 2001. Polyamides derived from piperazine and used for hot-melt adhesives: synthesis and properties. *International Journal of Adhesion & Adhesives*. 22: 75-79.
- Cagle, C.V., 1973. *Handbook of Adhesive Bonding*, McGraw-Hill, United States.
- Colegrove, L.F., W. Clay and R.D. Genova, 1998. Hot melt symposium Tappi, 1
- Gierenz, G and W. Karmann, 2001. *Adhesives and Adhesive Tapes*. Weinheim: Wiley-VCH.
- Jeffrey P. Kalish, Suriyakala Ramalingam, 2015. An analysis of the role of wax in hot melt adhesives *International Journal of Adhesion & Adhesives*. pp: 63-68.
- Le, G., 2011. The Effect Of Material Property And Open Time On The Performance Of Commercial Hot-Melt Adhesives, 24.
- Lee, J.J., Y.H. Song, S.K. Lim, D.S. Park, I.K. Sung and I.J. Chin, 2010. Effect of Composition of EVA-based Hot-Melt Adhesives on Adhesive Strength. *Journal of Adhesion and Interface*. 11: 157-160
- Li, W., L. Bouzidi and S.S. Narine, 2008. Current Research and Development Status and Prospect of Hot-Melt Adhesives, *Industrial & Engineering Chemistry Research*, 47: 7524-7532.
- Macosko, C., 1977. Adhesives rheology, *Adhesive Age*., pp: 35-37.
- Makromolekülen, P., 2015. *Humboldt-Universität Zu Berlin*. Retrieved from Differential scanning calorimetry investigation of polymers: [http://webcache.googleusercontent.com/search?q=cache:QKDwS9tDdfkJ:polymerscience.physik.hu-berlin.de/anleitg/dsc12\\_hu.pdf+&cd=3&hl=en&ct=clnk](http://webcache.googleusercontent.com/search?q=cache:QKDwS9tDdfkJ:polymerscience.physik.hu-berlin.de/anleitg/dsc12_hu.pdf+&cd=3&hl=en&ct=clnk)
- Qian, J.W., G.R. Qi and R.S. Cheng, 1997. Association of ethylene-vinyl acetate copolymer in dilute solutions—I. Solvent, concentration and annealing temperature effect. *European Polymer Journal*, 33: 1263.
- Tse, M.F., 1998. Hot Melt Adhesive Model: Interfacial Adhesion and Polymer/Tackifier/Wax Interactions. *The Journal of Adhesion*. 66: 61-88
- Park, Y.J., Hyo-Sook Joo, Hyun-Joong Kim and Young-Kyu Lee, 2005. Adhesion and rheological properties of EVA-based hot melt adhesives. *International Journal of Adhesion & Adhesives*. pp: 571-576.
- Pizzi, A. and K. Mittal, 2003. *Handbook of Adhesive Technology, Revised and Expanded*, 2nd Edition . New York: Marcel Dekker, Inc.