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Environmental effects and economic analysis of adhesives: a review of life cycle assessment (LCA) and techno-economic analysis (TEA)

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Environmental effects and economic analysis of adhesives: a review of life cycle assessment (LCA) and techno-economic analysis (TEA)

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Abstract. *In this paper, adhesives made either from fossil fuels or biological sources were examined. The purpose of this review was to investigate the development, processing, and production of bio-adhesives, especially key information about environmental and economic impacts of these types of adhesives. Specifically, the literature was reviewed in terms of life cycle assessment (LCA) and techno-economic analysis (TEA) in order to evaluate the environmental effects, economic performance and potential market acceptance. Several key parameters for life cycle analysis will be compared, such as resource consumption, ecosystem quality and human health; As for TEA, capital costs, operational costs, and unit costs will be explored, as well as the breakeven points. Underlying issues in LCA and TEA will be discussed, and we will examine areas needed for improvement for emerging biobased adhesives.*

Keywords. Adhesives, life cycle assessment, techno-economic analysis

Introduction

Adhesives have been discovered in Italy in the Neolithic age (Mazza et al., 2006). At that time, it was the simple birch-bark-tar adhesive used for hafted arrowheads. Ancient Romans, as well as Chinese, made bird lime from the juice of mistletoe as an adhesive to catch birds (Donkerwolcke et al., 1998). In 1791, adhesives were used for waterproofing because of its property as the natural rubber (Wetzel, 1962), but little improvements were made afterwards. Until the 20th century, phenol-formaldehyde resins have been introduced to produce structural adhesives (Hartshorn, 1986), such as hot-melt adhesives used in 1950s (Paul, 2003).

In ASTM D907, adhesion was defined as *the state in which two surfaces are held together by interphase forces*; adhesive is defined as *a substance capable of holding materials together by surface attachment* (ASTM, 2012). In a more general way, adhesive could be used interchangeably with cement, glue, resin, and paste. Most of the adhesives were used as the organic materials, both natural and synthetic adhesives.

Natural adhesives, primarily animal or vegetable origins, have been known for a long period. Even though there was a decreasing demand since mid-20th century, certain type of natural adhesives have been used with wood and paper products, such as labels and book bindings (Pike, 2015).

Synthetic adhesives are mostly based on synthetic resins and rubbers, thus, compared with natural adhesives, they are expensive to produce. However, synthetic adhesives could be produced in a constant supply and relatively uniform properties. Moreover, they can be modified in order to meet the best characteristics for the application (Pike, 2015).

The principles of adhesion were explained in several different ways. Below are the most common theories attempted to illustrate this phenomenon.

First, as well as the oldest, is the mechanical theory. Once upon a time, adhesion was considered only happened by flowing and filling pores on the substrates, when it hardened, the substrates were held together mechanically (Petrie, 2007). Adhesive penetrates cavities in the surface and clings through mechanical forces (Messrs et al., 1964). The mechanical interlocking is a common adhesion type, and it is also used in dental materials (Bayne et al., 1992). They are significant on macroscopic scale for fibrous material, such as paper, leather and wood, but not in microscopic scale. Moreover, the hybrid layer is formed when the monomers permeates throughout the collagen fibril matrix in etched dentin (Marshall et al., 2010).

Second theory is about adsorption. The adsorption theory mainly illustrates the chemical bond and the dispersion force between the adhesives and the material. The chemical bonding, numerous possibilities, such as covalent, ionic, metallic, and even chelation bonding could create it (Marshall et al., 2010); however, the dispersion force requires the two materials are in sufficiently close and intimate contact (Allen 1992). The core of adsorption theory is that van der Waals interactions should be sufficient for good adhesion if there is a good interfacial contact (Bateup, 1981).

Third, there is an electrostatic attraction between the two materials. Even though it is not regarded as the major theories, it is applied appropriately in various scenarios. The electrical double layer at the interface and the surface of the material has been used to explain this theory. These forces are typically dispersion forces and the forces from the interaction of dipoles. This theory proposed that between the adhesives and substrates, the phenomenon of adhesion is caused by the electrostatic charges of opposite sign (Anon., 2012).

Forth, for some specific adhesion of polymers, the diffusion reacts on the interface or segments, but it was not useful in adhesion between smooth and rigid materials (Allen, 1992). This theory is important only under conditions where macromolecular mobility and mutual solubility are favored to inter-diffusion across the interface (Bateup, 1981). This phenomenon is a two-stage process, and wetting is followed by diffusion. In order to make diffusion occur, the adhesives and adherent must be compatible in terms of miscibility, which could be another explanation of why the diffusion theory could only be explained in few scenarios (Petrie, 2007).

Due to various applications of adhesives, they can be classified into several different types.

Adhesives are broadly classified into two types based on their loading capabilities: structural and pressure-sensitive adhesives (Anon., n.d.). Structural adhesives refer to those relatively strong adhesive that are usually normally used under glass transition temperature, such as epoxies, and acrylic adhesives. Advantages of using structural adhesives include reduced weight, higher durability and less cost (Johnson, n.d.). As for pressure-sensitive adhesives (PSA), they are low modulus which could be deformed under small pressures, and how well the PSA will bond to the adherent is determined by the amount of pressure applied. It has the advantage of good shear strength and adherence (Anon., n.d.).

Typically, four main types: solvent-based adhesive, emulsion adhesive, hot melt adhesive, and reaction setting adhesive or reactive adhesive are organized based on the change of fluid to a rigid state during bonding, which is also known as the hardening process (Brewis, 1992). In terms of pressure-sensitive adhesive (PSA), the bond was formed when pressure is applied to the adherend. The degree of adhesion was influenced by the amount of pressure applied on the surface of the adhesives. Structural adhesives have been applied successfully in aerospace as well as automotives; Pressure-sensitive adhesives are designed mainly for permanent or removable applications, such as labels, tapes and automobile trim (Cullen, 1992).

The most common used adhesives were derived from phenol-formaldehyde (PF) and urea formaldehyde (UF) (Zhang et al., 2013), especially PF adehsive, which is widely used in exterior environment because of its high weather resistance and water-resistance (Cetin and Ozmen, 2002). In 2004, there was a reclassification of formuldehyde to Category Ⅰ – known human carcinogen, which driven the desire of finding the alternative of phenol-formaldehyde based adhesives (Mai et al., 2004).What’s more, concerns about indoor air quality also contributes in developing the adhesives alternatives.

There are increasing concerns over the materials of adhesives which are considered un-environmentally benign (McDevitt and Grigsby, 2014). The depletion of non-renewable resource and environmental issues arises in recent dacades. In order to decrease the public concerns, the use of renewable materials becomes the new trend of adhesives (Rajagopalan et al., 2012).

In terms of the biobased adhesives, it was defined in 2007: materials of natural, non-mineral or non-petroleum based origin that can be used either in their natural state or after small modification, capable of reproducing the behavior and performance of synthetic resins (Jones, 2007).

Adhesives from renewable materials could come from numerous sources: like enzyme based adhesives (Kharazipour et al., 1997; Felby et al., 2002), modified vegetable oils (Adekunle, 2007; Dunky et al., 2002), furans (Safe Work Australia, 2009), casein, lignin (Nimtz and Pizzi, 1983), soy protein adhesives, and other proteins (Heimingway and Kreibich, 1984).

Enzymes from wood rot fungi were proposed by several researchers as adhesives (Felby et al., 2002; Kharazipour et al., 1997).The principles of the research were using of two component systems and using of one component systems, which referred to activation of the lignin nested in wood. However, the cost of enzymes were high, and the performance without additional resin were not obvious.

The second material was modified vegetable oils. It has been reported by several groups (Adekunle, 2007; Dunky et al., 2002). Majority of the research showed that the use of adhesives was associated with the reactivity of the unsaturated bonds within the fatty acid of the triglyceride of the oil. Moreover, Adekunle (2007) presented the use of modified soy bean oil used as the composite binder.

The use of furans has been explored for several benefits (Safe Work Australia, 2009). On one hand, furfural could be produced through hydrolysis. Moreover, they are relative easy to condense with themselves or other chemicals. However, the process of deriving furfural may not be environmentally friendly since it is larged rely on acid hydrolysis.

Casein, a natural protein derived from cow's milk, has been used since 1960 (Langenberg et al., 2010). But the high price of casein may be the reason of small quantity in market used as adhesives.

Lignin, a highly disordered polymer, has been explored for decades. It serves as the primary role of binding cellulose chains and wood fiber walls. Also, it can be collected as by-product of pulp and paper manufacture. Usually, the spent sulfite liquor (SSL) is the main source for adhesives (Nimtz and Pizzi, 1983). As a paper adhesives, it is dark and also requires high temperatures for long cycle times. Moreover, due to the strong inter- and intra- molecular bonds, the departure of lignin could be linked with lignin itself, or other chemicals.

As for other materials, tannins were widely used. Historically, tannin was used to preserve hides or leather. The use of tannin could be traced from their reactivity and cross linking chemistry with formaldehyde. Condensed tannins are typically regarded as polyphenolic materials and are made of oligomeric structures (Heimingway and Kreibich, 1984).

The objectives of this paper were to review the evaluation of the environmental effect and economic infuence based on bio-adhesives, as well as the comparison of traditional adhesives and biobased adhesives.

Properties of adhesives

Adhesives are used as engineering materials, thus, their mechanical properties and chemistry properties are quite important. Fundamental forces that can be applied to an adhesive could be classified into three types: tensile force, shearing force, and cleavage force. Most adhesives bonds are designed to subject the adhesive to shear, rather than tension or cleavage forces; as for the cleavage force, it is also very critical because it measures the weakest property in most materials (Pocius, 2012).

Typical components of the adhesive are listed in Table 1 below (Li et al., 2008).

Table 1. The common components in adhesives.

Components	Physical properties	Functions
Polymer	High molecular weight	Strength; hot tack
Resin	Low molecular weight	Lowers viscosity
Diluent	Amorphous low molecular weight	Adjust glass transition temperature of system dilutes polymer entanglement network
Wax	Amorphous low molecular weight	Increases setting speed; Provides heat resistance;

In the area of construction, adhesive bonding is used as an effective connection technology, especially at low temperatures (5-10°C). Since connection needs to be independently of the actual environmental conditions and seasons, the adhesives used in most connections are cold-curing thermosets. High-temperatures curing or post-curing is typically not possible (Moussa et al., 2013). The mechanical properties at early-stage are depending on the curing temperature and curing time (Moussa et al., 2012). However, low-temperature curing may lead to an incompletely cured adhesive system.

In 2012, the newly-developed dry adhesives mimicking the gecko adhesive system attracted attentions (Jin et al., 2012). It offered anisotropic adhesion properties, which is when sheared in one direction, but it was non-adherent when sheared in the opposite direction. This property is one of the largest advantages of gecko adhesives compared with conventional pressure-sensitive adhesives, and it is attributed to the complex and curved structures (Jin et al., 2012). Even though the gecko adhesive system inspired adhesive industry, criteria under shear loading conditions still required development (Bartlett et al., 2012).

As for pressure-sensitive adhesives, thermal stability and optical transparency are important properties. However, the rubber-based PSAs cannot meet the requirement of thermal stability, and silicon-based PSAs are much more expensive than rubber-based PSAs. In 2011, Ahn et al. conducted an experiment focused on PSAs from soybean oils. Compared with traditional PSAs, it has excellent thermal stability, transparency, and peel strength (Ahn et al., 2011). Moreover, the results from soybean oil-based PSA showed great potential to replace petro-based PSAs, such as flexible electronics and medical device.

The performance of pressure sensitive adhesives (PSA) is typically evaluated by tack, peel strength, and shear strength. Tack refers to how well a PSA could bond to a substrate in short time and small contact force; peel strength means how strong a PSA can bond to a substrate by peeling it off the substrate; shear strength characterizes the capability to resist deformation under shear force (Qie and Dube, 2011). However, due to the gel network in solvent-based PSAs is continuous but discontinuous in latex-based PSAs, the solvent-based PSAs have much better performance and widely used under the scenarios that large shear strengths are needed (Tobing and Klein, 2001).

However, the use of latex-based PSAs is more environmentally friendly than that of solvent-based PSAs, researchers tended to improve the performance of latex-based PSAs by several methods, such as producing lattices with the functional groups or making PSA films using either only the latex or a mixture of the latex, most of which were focused on the optimization of the functional groups (Tobing et al., 2001). Qie et al. in 2011 conducted an experiment by decreasing the amount of small polymers in gel-free untreated PSA films, which led to significantly better performance. In addition, by increasing the molecular weight at the same time resulted in better performance.

In terms of the manufacturing properties, functional graded adhesives are being considered in bonded joints to reduce the peel stress concentrations located near adhering discontinuities, particularly by changing the loading conditions. In 2011, Stapleton et al. started an analytical study to explore the impact of the loading conditions on the effectiveness of grading and the alterations to the grading due to adhesive flow during manufacturing (Stapleton et al., 2012). They found that by reducing the grading of the modulus of the adhesives material, the peel stress concentration could be reduced; however, the actual application of such joints were still limited due to several concerns, such as by using functionally graded adhesives, the flow of the adhesive during manufacturing may change the shape of the grading and cause the grading to be ineffective (Stapleton et al., 2012).

As for wood adhesives, mechanical properties, such as tensile tests and modulus of elasticity (MOE), are typically conducted (Stoeckel et al., 2013). Among various group of adhesives, amino-based showed the highest stiffness (Modulus values ranged from 3.5 GPa to 7.5 GPa), while polyurethane adhesives represented the lowest end of the range with modulus values starting at only 0.1 GPa (Clauss et al., 2011). As for the effect of humidity and water, most wood adhesives were significantly softened by water contact or elevated humidity. Phenol-based adhesives exhibited a very distinct softening effect with a reduction of modulus between 20% and 100% compared to the dry state, but structural urea adhesives showed only moderate reduction in the range of 10-30% in wet state compared with the dry condition. After re-drying, most of the adhesives could regain their original performance (Konnerth et al., 2010).

The utilization of traditional adhesive materials are mostly composed of synthetic components, which are generally thrown away after the use, and it may not able to support a high load since it was difficult to remove while maintaining reversibility (Bartlett and Crosby, 2014). A renewable and reusable adhesive exhibited high force capacity and easy release characteristics was investigated in 2013. In that research, force capacity as high as 810 N over 100 cm^2 contact areas was examined and the results showed that it can enable high performing and reuse over many loading cycles (Bartlett and Crosby, 2014).

Two critical characteristics of adhesive materials are the loading capacity and the ability to be removed and reused (Pocius, 2002). Traditional pressure-sensitive adhesives has a widely use in commercial, however, they could either support a high load while difficult in removing or weak load but higher reversibility. Thus, a new adhesive material which could display high force capacity, easy release, and reusability over multiple cycles creates a new technological opportunity, which is to adopt renewable adhesives (Bartlett and Crosby, 2014).

Environmental effects

Even though the completely cured adhesives could be regarded as non-toxic and safe, the manufacturing process, such as machining and grinding could produce hazardous materials both to human beings and the environment.

Few primary factors: toxicity, flammability, hazardous incompatibility, and equipment safety, could be considered as the important ones in adhesive bonding procedures (Petrie, 2007). The environmental impacts of adhesives are not only the issues with human health, but also the issues within nearby community because of the release of volatiles and other waste. For instance, toluene, one of the solvents, is VOCs used as carrier fluids in conventional heat-melt adhesives, and this solvent is quite environmental damaging and may cause safety concerns (De Gray, 1998).

The following Table 2 exhibited the legally permissible exposure limits (PEL) for some chemicals mostly found in epoxy adhesive systems (California Department of Health Service, n.d.).

Table 2. The legally permissible exposure limits (PEL) in adhesive systems.

Chemical name	Common Abbreviation	California OSHA PEL, ppm
n-Butyl glycidyl ether	BGE	25
Isopropyl glycidyl ether	IGE	50
Phenyl glycidyl ether	PGE	1
Diethylenetriamine	DETA	1
Toluene	N/D	100
Xylene	N/D	100
Methyl ethyl ketone	MEK	200

When contacting with the chemicals, the skin even the eyes could be affected by the evaporation of the chemicals in adhesive system. If the resin is heated, the vapor and spray mists could irritate the lung; even though most organic resins are not acutely irritating, certain types are capable of causing skin sensitization. Moreover, solvents inhaled through the skin may affect the central nervous systems just like drinking alcohol.

The main components of adhesives and sealant materials may be hazardous, even affect the health to the contacts. For instance, the curing agents (aliphatic amines, cycloaliphatic amines, and so on) may cause irritation or damage to the skin, eyes, and lungs. Certain types of curing agents may cause damage to organs such as the liver and affect the blood's ability to carry oxygen.

Due to the volatile organic compound (VOC) emissions from the urea-formaldehyde (UF) resins, researches began to focus on the replacement of UF resin in the formaldehyde-based system (Wiglusz et al., 2002). However, the excellent bonding properties and relatively cheap price were the important advantages of formaldehyde-based resins. When adding PVAc to the natural tannin adhesives, the better bonding effect and a high level of wood penetration was showed compared with the commercial adhesives. The emission from the formaldehyde was lower by adding PVAs, and greatly reduced when UV curable urethane acrylate was coated (Kim, 2009).

In 2014, McDevitt and Grigsby compared the environment impact of bio- and petro- chemical adhesives used in fiberboard production. By using *Ecoindicator* 99, the authors found that as for the entire life cycle, the petro-chemical adhesives have a 22% higher environment impact than the bio-chemical adhesives. Other comparisons between petrochemical and biochemical adhesives were exhibited in Table 3, along with the Figure 1. But as the rapid growth in Australia, the authors believed that the methodologies issues would overcome and the differences between petro- and bio-chemical adhesives could be reduced (McDevitt and Grigsby, 2014).

Table 3. Comparison of petro- and bio- chemical adhesives (McDevitt & Grigsby, 2014).

Items	Comparison results
Functional Unit	One square meter of medium-density fiberboard
Fossil fuel changes	39% lower of bio-adhesives because of less use in crude oil in their production
Climate changes	9% lower of bio-adhesives because of less transportation emissions
Resource depletion	40% larger for the petro-based adhesives than the bio-based adhesives
Ecosystem quality impact	59% more in the bio-adhesives than the petrochemical adhesives

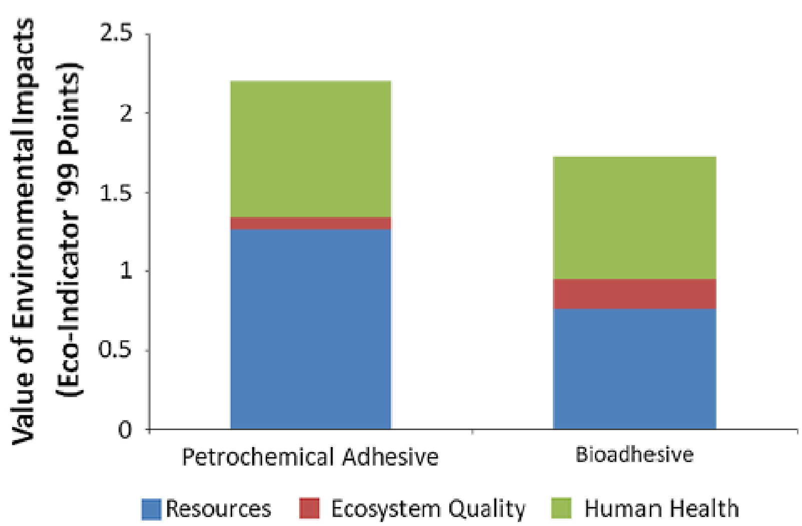

Figure 1. Life cycle environmental impacts comparison.

Due to the increasing restrictions on the release of volatile organic compounds (VOCs) to the atmosphere, there are decreasing trends from using organic solvents and dispersing media. As indicated in Pichelin et al. (1999), any species with a strong negative charge under alkaline condition is capable to render bonded panels of extremely low formaldehyde emission, in other words, the adhesives based on hexamine could reduce the impact of environment of formaldehyde. The results (Pichelin et al., 1999) showed that tannin-hexamine adhesive is a very environmentally-friendly product. As for lignin adhesives, in presence of pre-methylated lignin, adhesives pressed at high speed, and yielded exterior grade boards. However, it was still under tested at an industrial scale.

The electrically conductive adhesives (ECA) were considered as reliable materials for electronic components. Environmental life cycle assessment conducted showed that majority of the CO_2 emission came from a silver-epoxy based ECA compared with the tin-bismuth ECA. The researchers recommended that in order to save resources and decreases CO_2 emissions, the recycling of printed board assemblies using silver-epoxy based ECA was suggested (Andrae et al., 2008).

Conclusions

In this paper, the development of adhesives has been discussed and its related environmental issues have been investigated. Even though adhesives have been applied in industry for decades, few of the environmental influence and economic analysis has been done about adhesives. As more concerns over the VOC (volatile organic compounds) and other emissions, biobased adhesives have been explored. According to the review, the environmental effect from bioadhesives was much lower than petrobased adhesives. However, as the rapid development of bioadhesives market, more work needs to be done in both environmental and economic aspect.

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