

Natural Rubber Derivatives for Adhesives Applications: A Review

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The purpose of this review is to describe the utilisation of natural rubber and its modified form in adhesive applications. Adhesive can be defined as material that capable of joining two or more surfaces permanently by the adhesion process. Recently, natural rubber has gaining great interest in the adhesive application in comparison to its counter parts synthetic rubber due to environmental reason as well as good mechanical and thermal properties. In this review, the adhesive properties along with mechanical and thermal properties of the natural rubber are reviewed. The recent advances in the modified natural rubber such as epoxidized natural rubber, liquid natural rubber, telechelic natural rubber and other modified natural rubber are also included and highlighted.

1. Introduction

Apart from tyre and coating industries, adhesives are examples of the major industries that utilising rubber as their raw material. The flexible properties make rubbers as a good candidate for such application. To date, synthetic rubbers are still the main players in the current rubber industry especially for adhesive applications. Various synthetic rubbers for example butyl rubber, nitrile rubber and polyisobutylene etc are were widely used in such application (Poh and Imran, 2011). The uses of these synthetic rubbers are however causing the environmental problem as well as harm to human body from exposing and inhaling fumes emitted from the rubbers (Nor and Ebdon, 1998). Due to the disadvantageous of using synthetic rubber, natural rubber (NR) has gained significant interest to the industry especially adhesives applications due to their environmental friendly, easy to handle and non-toxic properties. NR is also known to inherit excellent elasticity, great adhesion properties and ability to crystallize under stretching (Hashim and Ong, 2017). Generally, NR cannot be used alone due to lack of adhesion properties and NR is usually blended with tackifier resins such as aliphatic or aromatic hydrocarbon, polyterpenes, and rosin derivatives to improve the wettability and bonding to the adherents.

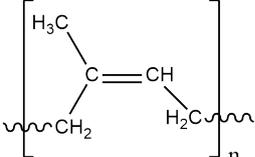
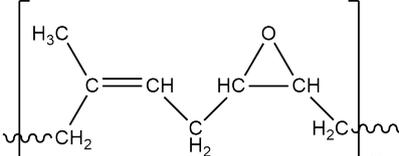
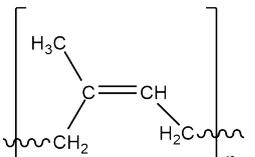
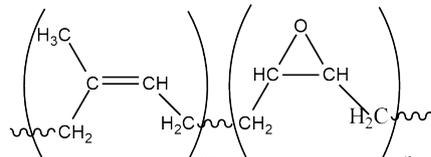
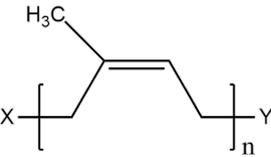
The wettability between the material and adherents is one of the most important factor for NR to be used as the adhesives as it causes great influence on the adhesion and mechanical properties (Poh, 2011). Modification of NR in order to improve the wettability between NR and the adherents is necessary due to the presence of unsaturated bond making NR inherits low polarity, poor ageing properties, poor oil and weather resistance and low abrasion (Hashim and Ong, 2017). Besides, high molecular weight and viscosity of natural rubber decrease the wettability of NR to adherents and reduce the adhesion properties. Enhancement of the properties and performance of NR can be achieved through grafting with polar materials, epoxidation of NR, depolymerisation to lower molecular weight of NR and addition of reinforcing fillers (Hashim and Ong, 2017). This modification of NR into more reactive material making it suitable for various applications particularly for adhesives application.

The present study aims to shed light on the utilisation of NR and its modified form in adhesives applications. Some of the important research studies relating to NR and its derivatives, including their synthesis methods, characterisation techniques, and recent research advancement, are briefly discussed.

2. Natural rubber for adhesives applications

NR is usually obtained by extracting from the Brazilian rubber tree namely *Hevea brasiliensis* that consist of long chains of monomer unit of cis-1,4-polyisoprene as shown in Table 1 (Hashim and Ong, 2017). Table 1 shows the structure of NR and its derivatives within the context of discussion. Besides rubber particles (30-40 %), NR also contains other components such as water (55-65 %), protein (2-3 %), sterol glycosides (0.1-0.5 %), resin (1.5-3.5 %), ash (0.5 -1.0 %) and carbohydrate (1.0 - 2.0 %) (Nor and Ebdon, 1998). In general, the dry rubber content of natural rubber latex is between 20 and 40 %. NR is known as a high molecular weight (M_w) polymer with molecular weight of up to 1×10^6 g/mol. In some cases, difficulty in mixing can happen where the intensive shearing process is needed to break down the molecular weight to a state where it is easier to blend with other additives (Ibrahim, Othman and Ismail, 2016).

Table 1: Chemical structure of NR and its derivatives

Type of NR derivatives	Structure of NR derivatives	Reference
Natural Rubber		(Hashim and Ong, 2017)
Epoxidised Natural Rubber		(Poh, 2011)
Liquid Natural Rubber		(Nor and Ebdon, 1998)
Liquid Epoxidised Natural Rubber		(Rooshenass, Yahya and Gan, 2018)
Telechelic Natural Rubber		(Nor and Ebdon, 1998)

Numerous methods for the fabrication of natural rubber-based adhesives have been reported. Traditionally, NR is masticated in a two-roll mill for 10 min at room temperature to cut the NR chains to a lower molecular weight rubber. The usage of low molecular weight rubber will enhance the dissolution process in the solvent. Masticated NR was then dissolved in toluene prior to slow the addition of resin under constant stirring and left at room condition of approximately 30 °C. Subsequently, resin was added into the prepared rubber solution to form natural rubber based adhesive (Poh and Putra, 2011). Similar method was reported by Poh and Saari for the preparation of adhesive based ENR 50 and magnesium oxide (2011). Another important modest method in the preparation of NR based adhesive was by directly adding NR (without mastication) and resin into the beaker followed by mechanical stirring at room temperature for 5 min at 100 rpm. Raja et al. (2013) demonstrated the mixing of NR and resin by stirring at room temperature using a mechanical stirrer for 5 min at 100 rpm. Hermiati, Fatriasari and Falah (2004) diluted NR in water to about 25 % total solid content in the presence of emulsifier and styrene monomer for better dilution of NR; followed by mechanical stirring at room temperature for various mixing time of up to 3 h. After that, the initiator and catalyst were added and the mixture was heated until the adhesive is formed. Other additives such as filler and antioxidant can be added to

the mixture to enhance the adhesion properties. The common fillers used for adhesive application are magnesium oxide, silica and organomodified halloysite. For curing with thermoset resins, curing agent was added and the mixture was stirred further for 15 min before the curing process (Tan et al., 2013). Another method was reported where the rubber and epoxy resin were mixed in a three-neck flask blanketed with nitrogen at 150 °C for few hours. Subsequently, filler and curing agent were added into the mixed solution of epoxy resin and rubber based on the proper weight ratio under stirring. Selection of curing agent can be differed according to the type of polymer resin (depending on the functional group presence in the resin). For example, liquid epoxy resin (diglycidyl ether of bisphenol) containing epoxide group in the structure can be used together with amine curing agent to form crosslinking between them (Zainudin et al., 2020). Usually, degassing process was performed to eliminate the air trapped in the mixed solution before the curing process (Lee, Wang and Chin, 1986).

Besides bulk NR, the preparation of adhesive using NR in the latex form was also extensively reported. Raja et al. (2013) reported the compounding of natural rubber latex (NRL)-based waterborne pressure-sensitive adhesives (PSA) at two resin contents (25 % and 50 %) with three aliphatic hydrocarbons water-based dispersion (varying softening point) which resulting in good adhesive properties at 50 % resin content. It was apparent that PSA containing 95 °C tackifier dispersion at 50 % resin content showed the optimum adhesive and cohesive characteristics on both stainless steel and low density polyethylene (LDPE) substrates. The analysis was conducted at different temperatures (5, 15, 25, 35 and 45 °C). Hermiati, Fatriasari and Falah (2004) investigated various factors that affect the synthesizing process of NRL and styrene blend. Apparently, factors such as heating time, catalyst and pre-stirring did not show significant influence to the strength of adhesives. Instead, by adding 10 % of phenol-formaldehyde (PF), it significantly improved the strength of adhesives. This observation was attributed to the cross-linking between reactive sites (aldehyde group) of PF and α -methyl or α -methylene group of polyisoprenes. The adhesion properties of NRL with nitrile-phenolic adhesive containing various concentration of toluene diisocyanate-nitrosophenol (TDI-NOP) adduct was reported (Achary, Gouri and Ramaswamy, 2001). It was found that the peel strength of the adhesive was enhanced with the increasing concentration of TDI-NOP adduct. This observation was attributed to the improvement of the interfacial adhesion between resin and rubber after the addition of TDI-NOP adduct.

The modification of NRL grafted with n-butyl acrylate (BA) and methyl methacrylate (MMA) were fabricated by Chumsamrong and Mondobiyai (2008). In this research, it was reported that the peel strength of NR grafted with BA and MMA possessed lower peels strength as compared to pure NRL. This observation was attributed to the polymer chain of grafted modified NRL did not sufficiently wet the substrate. The wetting ability of the polymer chain in this latex may have decreased due to short branching. However, adhesive with the monomer (BA: MMA) ratio of 80:20 acquired sufficient adhesion property for peel testing. NRL grafted with BA and MMA possessed higher oxidative stability than pure NRL (Chumsamrong and Mondobiyai, 2008). Thongnuanchan et al. (2014) reported the grafting of diacetone acrylamide (DAAM) onto NRL. It was observed that the adhesive properties such as tensile strength and lap shear strength were enhanced successfully. Proton nuclear magnetic resonance ($^1\text{H NMR}$) was utilised to determine the degree of grafting of poly (diacetone acrylamide) while Fourier-transform infrared spectroscopy (FTIR) was used to observe the keto-hydrazide crosslinking reaction of the ketone carbonyl of grafted PDAAM with adipic acid dihydrazide (ADH). It was noted that by increasing the tackifier resin (such as gum rosin) content up to 50 phr, the shear strength was increased (Thongnuanchan et al., 2014). Rezaifard et al. (1994) studied the properties of epoxy resin incorporated with poly (methyl methacrylate)-grafted natural rubber. It was observed that the poly (methyl methacrylate)-grafted natural rubber has induced a greater toughness in epoxy resin in adhesive and bulk form as compared to carboxyl-terminated butadiene-acrylonitrile (CTBN) rubbers. This is possibly due to the presence of polar group in poly (methyl methacrylate) that increased the compatibility between the epoxy resin and the rubber.

3. Natural rubber derivatives for adhesives applications

NR has undergone various modification to improve its performance in various applications. Some of the modifications are epoxidation of NR, grafting, depolymerisation and others. These modifications are important in enhancing the properties of NR in comparison to unmodified NR. The epoxidation process of natural rubber improved the heat and chemical resistance of the rubber due to the presence of epoxide group which increases the polarity of the rubber and at the same time reducing the amount of unsaturated double bonds. Grafting with polar material was also reported to improve some properties of NR such as mechanical properties and adhesive properties as the presence of polar groups increased the compatibility of NR with other materials such as resin or fillers. Depolymerisation of NR on the other hand, reduced the molecular weight of NR and convert NR into a lower molecular weight substance which also known as liquid natural rubber (LNR). LNR is easy to be processed without the need of high energy in comparison to high molecular weight NR and due to inexistence of reactive terminal group and the presence of high proportion of elastically

inactive chain ends, the LNR products did not offer good mechanical properties and the application of LNR is still limited in the industry (Nor and Ebdon, 1998). Further advances in modification of LNR by adding reactive group such as carboxyl and hydroxyl at terminal chain greatly improved the properties of NR as adhesive. It was reported that the adhesive properties such as shear strength and peel strength, thermal properties and chemical resistance were enhanced accordingly (Wayakron Phetphaisit et al., 2013).

3.1 Epoxidised natural rubber

Epoxidation of NR resulted in a more polar and reactive NR which known as epoxidized natural rubber (ENR) (Table 1). The presence of epoxide group in ENR enables the reaction with other polar material and enhanced the chemical interaction between them. Poh and Saari, (2011) investigated the adhesion properties of ENR50 incorporated with magnesium oxide. It was observed that by increasing magnesium oxide content, the viscosity of the adhesive was enhanced which led to the increment of peel strength and loop tack property up to maximum values of 30 parts per hundred parts of rubber. These observation could be due to the maximum wettability occurred at 30 parts per hundred parts of rubber. Khan and Poh (2012) prepared the ENR50-based adhesive using petro resin as tackifier. The peel and shear strength were enhanced with increasing molecular weight of ENR50 up to an optimum molecular weight of 4.2×10^4 g/mol of ENR50. Further increasing of the molecular weight led to decreasing peel and shear strength. The increment of peel and shear strength was attributed to the effect of wettability and mechanical strength of ENR50 itself. However, the drop of peel and shear strength after the optimum value of molecular weight was probably due to the higher viscosity as the entangled rubber chain does not flow effectively to produce good wettability on the substrate.

Khan and Poh (2010) investigated the effect of silica filler on the peel strength of natural-based adhesive with coumarone-indene resin as tackifying resin. It was observed that by increasing silica content, the peel strength increased up to 40 phr of silica content for both ENR50 and ENR25. This increment of peel strength up to 40 phr of silica content can be attributed the greater molecular interaction between silica and rubber molecules which enhanced the wettability of adhesive that led to better peel strength. Further increasing of silica content above 40 phr, the peel strength of adhesive decreases due to the increase in viscosity which restrict the chain mobility and reduce the peel strength of the adhesive. ENR based adhesives were prepared by two roll-mill using ENR25 with two crosslink system which are diamine/bisphenol A and anhydride/imidazole and resin PX1150 adhesive (terpene polymer) as tackifier resin (Yoksan, 2008). It was observed that the adhesive prepared from sulfur cured ENR25 showed higher tensile strength than the one from anhydride crosslinked ENR25 due to better crosslinking efficiency. Besides, the tensile strength and tensile modulus of sulfur cured ENR25 adhesive was improved with the incorporation of tackifier resin. On another note, organomodified halloysite (OHNT) was incorporated into ENR50 to optimise the properties of the rubber adhesives for potential application in flame-resistant adhesive (Tan et al., 2016). OHNT is known as efficient flame retardant due to its tube shape which can help in trapping the flammable volatiles. Thermal analysis revealed the maximum decomposition temperature of the adhesive was enhanced with the increasing amount of OHNT. The limiting oxygen index (LOI) value also increased from 21 to 72 % with the maximum addition of OHNT (5:1 weight ratio of OHNT:ENR50) and these adhesives can be classified as self-extinguished materials that may suitable for fire-resistant coating applications.

3.2 Liquid natural rubber and liquid epoxidised natural rubber

Depolymerisation of natural rubber resulted in the formation of LNR which is low molecular weight substance with similar microstructure as NR. The structure of LNR and liquid epoxidized natural rubber (LENR) is shown in Table 1. LNR and LENR can be produced via thermal, oxidative degradation, redox reaction, photolysis, mechanochemical peptization and photochemical oxidation (Baharulrazi et al., 2017). To date, the use of LNR and LENR in adhesive applications are not widely reported. In 2014, Suchat and Yingprasert reported the performance of natural rubber modified for eco-adhesive for wood application. Three latex types were examined, namely epoxidized natural rubber (ENR), liquid natural rubber (LNR), and natural rubber/polymethyl methacrylate (NR/PMMA) blend. The polar ENR based adhesives showed the highest adhesive properties with shear strength of 0.63 MPa followed by LNR and NR/PMMA with shear strength of 0.55 and 0.51 MPa. From this study, it is apparent that the presence of polar groups in ENR contributed significantly to the adhesive properties of NR in comparison to the effect of molecular weight.

LNR and LENR have been reported to be a good impact modifier or compatibilizer for polymer composites. Research by Saleh et al. (2014) utilised LNR as impact modifier for epoxy composites and it was found that tensile strain, fracture toughness were increasing while the tensile strength, strain and the glass transition temperature were decreasing with the addition of LNR. The enhancement of tensile strain is related to the plasticizing effect of LNR while the improvement of fracture toughness is due to the incorporation of LNR that enabled plastic deformation to occur in matrix. Similar finding was reported by Tan et al. (2013), using LNR and LENR as toughening agent for epoxy. It was found that by adding the rubbery phase into epoxy resin, the

toughness of the epoxy was enhanced. 3 wt% of LENR composite acquired the highest mechanical properties for both flexural and impact properties. While Muzakkar et al. (2015) studied the effect of incorporation of polyethylene grafted maleic anhydride (PE-g-MAH) as a coupling agent for melt blending of natural rubber, linear low density polyethylene (LLDPE) and LNR as compatibiliser forming thermoplastic natural rubber (TPNR) towards the adhesion and mechanical properties. The aluminium (Al) surface was pre-treated with 3-glycidoxy propyl trimethoxy silane (3-GPS) to enhance the mechanical properties of laminated composite. It was observed that by increasing amount of PE-g-MAH, the shear strength of the single lap joint Al-TPNR laminated composite was enhanced for the 3-GPS surface treated aluminium.

3.3 Telechelic liquid rubber

Telechelic liquid rubber can be defined as a low molecular weight rubber which consist of reactive terminal groups capable of being used in further chain extension and crosslinking (the structure is shown in Table 1). The addition of reactive group such as carboxyl and hydroxyl at terminal chain of liquid natural rubber is expected to increase the polarity and improve the chemical bonding with other polar material as compared to unmodified liquid natural rubber. Wayakron Phetphaisit et al. (2013) prepared the polyester polyurethane elastomer adhesive from hydroxyl liquid natural rubber (HTNR) and modified poly(ethylene terephthalate) with polymeric 4,4'-methylene bisphenyldiisocyanate (MDI). It was observed that the optimum wood adhesive shear strength and chemical resistance of the synthesized carboxyl terminated PET and modified rubber contained 50 % hydroxyl content occurred at NCO/OH molar ratio of 0.45:0.55:0.75. The production of carboxyl terminated liquid natural rubber (CTNR) was produced through photochemical reaction using maleic anhydride and masticated natural rubber (Dileep, Avirah and Joseph, 2003). It was found that CTNR based adhesive has better adhesive bonding between NR to NR and NBR to NBR compounds than the masticated NR based adhesive. By incorporating silica, it improved the peel strength of NBR compounds using CTNR based adhesives. Besides, the peel strength of adherents bonded by CTNR adhesive was enhanced with the addition of phenol-formaldehyde resin (PF) resin and 4,4', 4" triphenyl methane tri isocyanate (Desmodur-R) for bonding NR and NBR to mild steel. This observation was more prominent on the NBR adherents as compared to NR adherents due to the presence of possible ionic interactions involving the carboxyl group of CTNR, nitrile part of NBR and the isocyanate group of Desmodur-R. Radabutra et al. (2017) reported on the fabrication of modified telechelic natural rubber-based pressure-sensitive adhesive. The telechelic natural rubber containing hydroxyl and carboxyl groups grafted with maleic anhydride was characterised based on the thermal analysis and adhesive properties. It was observed that with the presence of grafted maleic anhydride, the glass transition temperature of adhesive becomes higher due to the presence of multiple hydrogen bonds while the adhesive properties increased due to enhanced wettability and formation multiple hydrogen bonds.

4. Conclusions

The main objective of this review is to cover a wide range of natural rubber and its modified form particularly for adhesives applications. Various properties such as adhesive, mechanical and thermal properties were highlighted and discussed in brief. The techniques for the preparation of these adhesive based materials were also reviewed. From this review, it can be concluded that the adhesion properties of modified natural rubber are superior compared to the unmodified NR which probably due to the presence of polar groups and addition of filler which improved the wettability and the overall properties of adhesives. The usage of telechelic liquid rubber such as HTNR and CTNR are still limited in contrast to NR, ENR and LNR due to higher price and challenges in terms of mass production. Telechelic liquid rubber which suitable for further chain extension and crosslinking have unlimited potential to be uncovered not only for adhesive applications but in other engineering fields particularly toward sustainable development in the scientific field.

Acknowledgments

The authors would like to thank Ministry of Higher Education (MOHE) and Universiti Teknologi Malaysia for the Fundamental Research Grant Scheme (Q.J130000.7851.4F998) and UTM Transdisciplinary Research Grant (Q. J130000.3551.06G39) for supporting this research.

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