



Irradiation grafting of natural rubber latex with maleic anhydride and its compatibilization of poly(lactic acid)/natural rubber blends

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ABSTRACT

Maleic anhydride (MA) is an interesting monomer to be grafted onto natural rubber (NR) due to its potential as a compatibilizer of hydrophobic rubbers and polymers with higher polarity. So far, radiation grafting of MA onto NR in latex state has not been reported. In this study, the grafting of NR with MA in latex state was investigated by exposing the latex to cobalt-60 gamma irradiation at a fixed MA content of 9% and a varied absorbed doses from 2 to 10 kGy. The FTIR spectrometer, ^1H NMR spectrometer and gel content analysis have confirmed successful grafting of MA onto NR after irradiation. The grafted NRs were then used to increase the compatibility and the impact property of PLA/NR blends. It was found that the highest impact strength of the blends was achieved when the grafting was carried out at the absorbed dose of 4 kGy.

1. Introduction

One of the important polymers produced from natural resource is NR, extracted from rubber trees (*Hevea brasiliensis*) in the latex form. NR latex contains water, proteins, lipids, phospholipids, carbohydrates and other organic and inorganic compounds (Sakdapipanich, 2010). NR has better mechanical properties than synthetic rubbers including higher elasticity, tensile strength and tear resistance (Kongparakul, 2013). This is because NR latex contains proteins and phospholipids around the surface of its particles. Besides, NR naturally has higher molecular weight than synthetic rubber. However, since NR is hydrophobic, its applications as blends with higher polarity compounds are limited. One of the polymers that NR could be incorporated is PLA because PLA is also another biopolymer. The blends of NR into PLA could reduce its inherent brittleness. Hence, many researches have been carried out to improve the compatibility of NR with PLA by using compatibilizers (Pattamaprom et al., 2016) or by grafting NR with other polymers (Puteh et al., 2015; Sabbagh, 2003; Mohammad et al., 2015). Examples of monomers used to graft with NR includes acrylates (Ragupathy et al., 2011), methyl methacrylate (Bakar and Fauzi, 2012), glycidyl methacrylate (Juntuek et al., 2010, 2012) and poly(vinyl acetate) (Chumeka, 2013), acrylonitrile (Angnanon et al., 2011), styrene (Dung et al., 2017), poly(dimethyl (acryloyloxymethyl)phosphonate) (Derouet et al., 2008), poly(dimethyl (methacryloyloxyethyl) phosphonate) (Derouet et al., 2008) and maleic anhydride (Somboonchai et al., 2013; Nakason et al., 2004, 2006; Wongthong

et al., 2013). The grafting was carried out either by reactive blending, in NR solution or in NR latex. The commonly used chemical initiator is benzoyl peroxide (BPO).

In the past, Nakason et al. (2004) studied the grafting of MA onto NR in toluene solution using benzoyl peroxide (BPO) as an initiator. Wongthong et al. (2013) also used benzoyl peroxide (BPO) as an initiator to graft MA onto deproteinized NR (DPNR) in latex state. The latter paper proposed that the grafting mechanism in this case was the free radical mechanism, where the reaction sites were at the double bonds of NR molecules. Nevertheless, prior to these works, Nakason et al. (2006) studied the reactive blending of NR with MA in an internal mixer without chemical initiator. They proposed two reaction pathways for grafting NR with MA as shown in Fig. 1, which were free-radical mechanisms (a and b) and Alder-ene reaction (c). For the free radical mechanisms, the free radicals were generated at the C=C bonds (a) and at the allylic carbon atoms (b), respectively. The Alder-ene reaction (c) is another possible grafting mechanism occurring by a substitutive addition reaction.

Later on, radiation method has been investigated for grafting and/or crosslinking of polymers. This technique is interesting in that it could reduce the amount of chemicals used in the reaction by leaving out chemical initiators. Makuuchi (1994) proposed that radiation excited polymer molecules and caused the formation of polymer radicals ($\text{R}\cdot$) in the same way as chemical initiators. These radicals then further reacted with monomers (M) and formed branches on polymer molecules. The bridging of growing polymer radicals also created crosslinked

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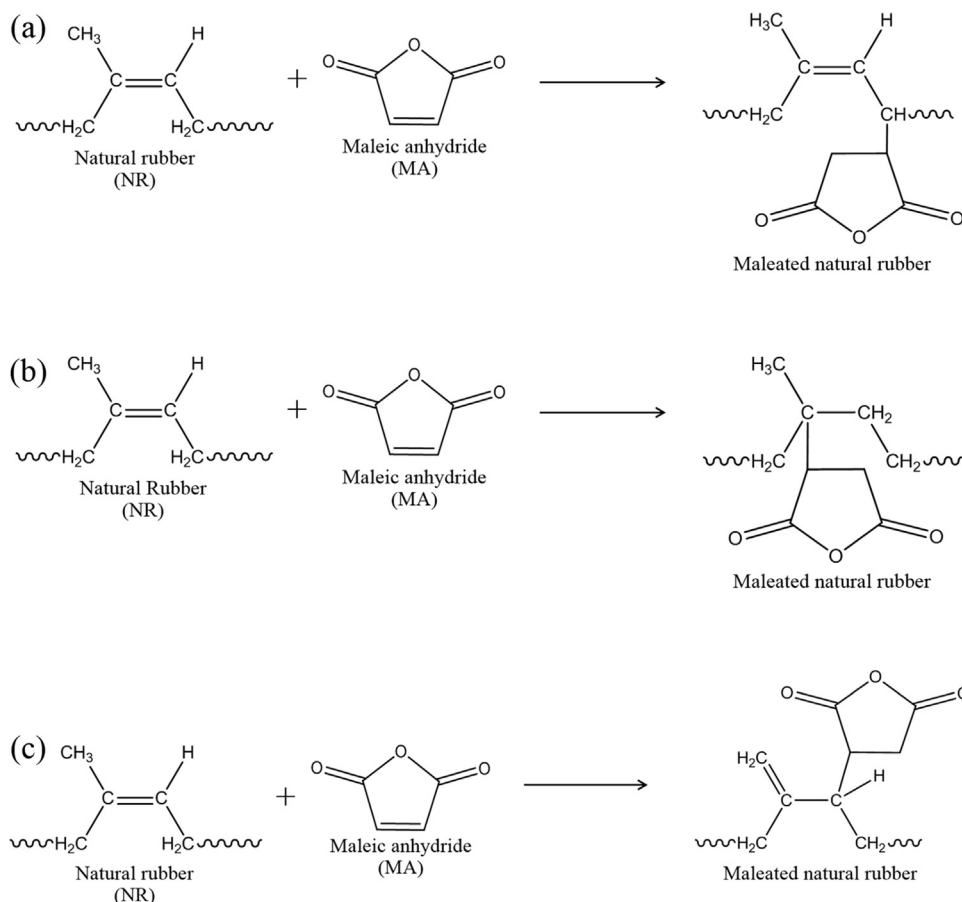


Fig. 1. Possible grafting mechanisms of MA onto NR by reactive melt blending, where (a) and (b) are free radical mechanisms and (c) is the Alder-ene reaction (Nakason et al., 2006).

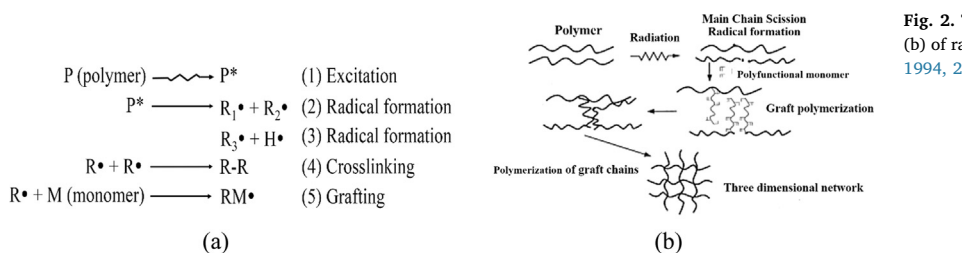


Fig. 2. The reaction mechanisms (a) and the schematic drawing (b) of radiation grafting and crosslinking of polymers (Makuuchi, 1994, 2003).

Table 1

The composition of MA-NR latex mixtures used for the radiation grafting experiments.

Ingredients	Quantities
HA-NRL (60% NR)	100 g
SDS	0.8 g
KOH	0.4 g
Na ₂ CO ₃	0.4 g
Deionized water	100 g
MA	9 wt% ^a

^a Percent by weight of dried rubber.

Table 2

The formulations of the PLA/NR blends used in this study.

Sample	PLA (wt%)	NR (wt%)	NR-g-MA (wt%)
PLA	100	–	–
PLA/NR/NRL ^a	90	5	5
PLA/NR/NR-g-MA ^b	90	5	5

^a NRL is the ungrafted natural rubber coagulated from the HA-NRL.

^b Five NR-g-MA samples used in this work were synthesized at the absorbed doses of 2, 4, 6, 8, and 10 kGy, respectively.

structures. The described mechanisms for grafting and crosslinking by radiation method are illustrated in Fig. 2. Radiation grafting of NR latex has been reported using some monomers (Dafader et al., 2006; Iskandar, 2011). Dafader et.al (2005) studied on grafting of acrylic monomers onto natural rubber by using a Co-60 gamma irradiation at the absorbed doses of 2–8 kGy and found that the optimal absorbed dose that provided the highest mechanical properties was 4 kGy. This is consistent with another work by Iskandar (2011), which reported that

the maximum tensile strength of the film produced by irradiated starch-rubber latex was achieved at 5 kGy. Therefore, in this work, the grafting of MA onto NR in latex state was investigated at the absorbed doses ranging from 2 to 10 kGy and the synthesized MA-grafted NR films were also investigated for their potential as compatibilizers for PLA/NR blends.

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