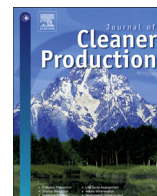




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Evaluating the environmental impacts of graft copolymer prepared by conventional emulsion polymerization, electron beam irradiation, and gamma ray irradiation through life cycle assessment

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ABSTRACT

Graft copolymer of natural rubber (NR) and styrene to used as a blend with styrene-butadiene for green tire can be produced via three techniques: emulsion polymerization (EP), electron beam irradiation (EB), and gamma ray irradiation (GR). Products from all techniques had similar cross-linking properties; leading to similar mechanical properties. Therefore, producing the graft copolymer using less resources is the best choice as it is a cleaner production. This paper compared the environmental impact assessment of these three production processes. SimaPro version 7.3 software using Eco-indicator 99 method for environmental impact assessment was used throughout this study. The functional unit (FU) used was 100 g of grafted rubber product, with the considered system boundary starting from raw materials for the production of a specified product and continuing to the end of copolymerization processes (cradle-to-gate). The results indicated that, at the laboratory scale, the EB technique had the largest contribution of environmental impacts, followed by EP and GR techniques. However, operating at the maximum capacity led to an overall reduction of all impacts. The reduction was improved by 86.32% for EB and by 60.5% for GR. However, production by EB still contributed the highest overall environmental impact, while GR contributed the least. In decreasing order of significance, the main impacts of EB were resources (R), human health (HH), and ecosystem quality (EQ). The essential factors which caused these impacts were, in decreasing order of significance, the usages of electricity, NR, and styrene. Therefore, GR method was the choice for scaling up the green production process in both saving the electricity usage and for improvement in the environmental impacts.

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1. Introduction

Natural rubber (NR) is a product from para rubber, botanically known as *Hevea brasiliensis* (Family: Haveaceae, Genus: *Hevea*, Species: *Brasiliensis*) (Dayaratne and Gunawardana, 2015). This tree is one of the major economic tree crops in tropical areas throughout the world, especially in southern China, Thailand, Vietnam, and

Cambodia (Kumagai et al., 2015). Around the 1910s, NR was introduced to Thailand (Jawjit et al., 2010). Since 2003, Thailand has been ranked as one of the four largest producers of NR in the world with a production of about 35% of the latex produced worldwide (Jawjit et al., 2010, 2015; Songsing et al., 2013). In 2008, Thailand's NR upstream productivity was 1698 kg/ha, while that of Malaysia was 1430 kg/ha, that of India was 1930 kg/ha, and that of Indonesia was 935 kg/ha (Marimin et al., 2014). In 2011, the fresh latex production in Thailand was about 3.4 Mt with an average yield of fresh latex around 1600 kg/ha (Jawjit et al., 2015). Thailand has exported NR to the international market in the form of block rubber, ribbed smoked sheet rubber (RSS), concentrated latex, and rubber

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products, which together constitute 3.12 Mt or 86% of total production. This increases the national income by approximately 3.3 billion THB. In 2011, Thailand exported about 880,000 t of concentrated latex, with a value of 77,000 million THB (Jawjit et al., 2015). The domestic production was 0.5 Mt or 14% of total production. This increased national income by approximately 2.6 billion THB. In high of all this, NR industry in Thailand should be considered to be of economic and social importance: it has significant production value, revenue from exports, and employment. Due to the current NR crisis, the market value of NR has been continuously decreasing while the production yield remains stable; hence this study focused on the improvement of NR to increase market value. NR has high elasticity, high resilience, a highly cohesive character, adhesive strength, and high tear strength even without reinforcing filler (Chern, 2006; Derouet et al., 2009; Hassan et al., 2013; Matos et al., 2014). Its structure is similar to that of butadiene. Although butadiene has properties of flexibility, abrasion resistance, and low heat absorption, there is sufficient evidence that butadiene causes cancer of the hematolymphatic organs in humans. Such a conclusion was based on the studies of the increasing risk of leukemia in epidemiologic study in among styrene-butadiene rubber (SBR) industry workers and on the increasing risk of leukemia and lymphoma in butadiene monomer industry workers (Robert Baan et al., 2009; Sathiakumar et al., 2015). Hence, NR is a good alternative to substitute butadiene or to use as a blend with SBR for green tire products.

To produce copolymer of NR and styrene, graft copolymerization is a well-known technique to impart a new property or enhance the existing properties in the polymer with minimum loss of the original properties. The nature of the changes in the properties depends on types of monomer being grafted, percentage of grafting, method of grafting, and distribution of the grafted chain throughout the polymer (Khan et al., 2002). NR has typically been modified or grafted by various types of polymers in order to alter and improve its properties. L. Thiraphattaraphun et al. (2001) studied the graft copolymers of NR and PMMA by a melt-mixing system. The tensile strength, tear strength, and hardness of the product increased with an increase in PMMA content. Much research has reported on the grafting of vinyl monomers such as methyl methacrylate (MMA) and styrene (ST) onto NR latex (Kongparakul et al., 2009). Pukkate et al. (2007) reported the graft copolymerization of ST onto deproteinized NR to form a nano-matrix structure. The ST conversion and grafting efficiency were found to be dependent on the ST/rubber ratio.

There are many methods nowadays to produce a copolymer of styrene and rubber: conventional emulsion copolymerization (EP), electron beam irradiation (EB), and gamma ray irradiation (GR) being examples of such methods which were used in this study. Each method has its own advantages and disadvantages. EP technique uses water as a medium in the reaction. In this case, an initiator is dissolves in water instead of monomers. Emulsifier is then assembled into micelles, and monomers enter into micelles and spread throughout the water. Throughout this process, polymerization reaction occurs when initiators enter into micelles. The advantages of this method are the ability to prepare polymer particles with a size range between 0.05 and 5 μm and with low-cost production. Based in part on such advantage, this method is commonly used in industries such as the EP of ST and MMA (Shen et al., 2015). There have been many studies of graft copolymerization of PS-g-NR by EP in terms of influences of reaction conditions on monomer conversion, grafting efficiency, graft yield, and mechanical properties (Jaimuang et al., 2015; Suksawad et al., 2011). EB technique uses the electron beam produced from an electron gun to enhance polymerization efficiency. Electron beam curing is a fast, clean, and non-thermal continuous process that utilizes highly

energetic electrons at controlled doses to polymerize and crosslink materials (Vijayabaskar et al., 2008). It can affect the physical and chemical modification of polymeric materials (Noriman and Ismail, 2011) as well as improve productivity and mechanical properties, increase the rate of production, and reduce emission of pollutants and solvents (Shin et al., 2009; Yasin et al., 2005). EB technique has been used in the dynamic vulcanization of polymer blends leading to thermoplastic elastomer vulcanizates (Naskar, 2009) and has recently been used to produce polystyrene-graft natural rubber copolymer (PS-g-NR) (Khampod et al., 2015). GR technique, for its part, uses gamma rays from cobalt-60 (Co-60) to accelerate the polymerization process in a rubber system. The advantages of this method are the ability to produce copolymer in large quantities at low cost. Gamma radiation was found to affect the physical, mechanical, and thermal properties of the ethylene propylene diene monomer rubber (EPDM), high density polyethylene (HDPE), and ground tire rubber powder (GTR) composites. The results indicated that the GR led to a significant improvement in the properties of the blend compositions (Abou Zeid et al., 2008). In particular, El-Nemr and Khalil (2011) found that GR achieved the compatibility of recycled HDPE/ground rubber powder blends led to an improvement of mechanical properties. Zaharescu et al. (1999) reported that GR of EPDM/polypropylene (PP) at different ratios of utilizing waste PP increased gel content and improved mechanical properties. However, there has not yet any study on the environmental impact from the grafting production processes using the above-mentioned techniques.

The environmental impact as manifested in ozone depletion, photochemical smog, and ecotoxicity has been focused on recently in term of climate change, faster melting of polar ice, rapid sea level rise, etc. There are consequences of global warming crisis. The main reason for global warming is the emission of greenhouse gases (GHGs), which mainly consist of carbon dioxide (CO_2), methane (CH_4), nitrous oxide (N_2O), hydrofluorocarbon (HFCs), perfluorocarbons (PFCs), and sulphurhexafluoride (SF_6) (Dayaratne and Gunawardana, 2015). These emitted gases increase the earth's temperature (Ahmad and Hossain, 2015) and such a raise in temperature also affects to the livelihood of the people directly. Environmental researchers have studied strategies to provide better environmental management. One such environmental assessment tool is life cycle assessment (LCA), which has been used to evaluate the environmental impacts which occur due to the production process or the product itself. Beginning with raw material production, through manufacturing, packaging, and marketing processes, LCA includes the use, re-use and maintenance of the product, as well as disposal or waste management after use (de Vries et al., 2015; Nucci et al., 2014; Tukker, 2004). For example, the GHGs emission from the rubber production in Thailand per ton of concentrated latex, RSS, and block rubber (STR 20) released 0.54, 0.64, and 0.70 ton CO_2 -eq., respectively (Jawjit et al., 2010); the emission of total CO_2 from the rubber band manufacturing of production in Sri Lanka amounted to 1.16, 1.53, and 1.23 ton CO_2 -eq./ton of product in factories A, B, and C, respectively (Dayaratne and Gunawardana, 2015); also the overall land used changed from oil palm cultivation in the East of Thailand, yielding GHG savings amounting to 47,214 ton CO_2 -eq./year (Permpool et al., 2015). Moreover, Lee et al. (2012) used LCA to assess the environmental impacts of sewage effluent through three different disinfection processes — EB, ultraviolet (UV), and ozone systems. These LCA results showed that the lowest impact was from UV, followed by EB and ozone, while the electricity usage was the key issue of those disinfection processes. Bhuiyan et al. (2016) studied wastewater treatments for the reduction of environmental problems in the textile dyeing industry by using high-energy gamma radiation. The results indicated that the treated wastewater can be used as an

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