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Liquefaction of ground tire rubber at low temperature

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ABSTRACT

Low-temperature liquefaction has been investigated as a novel method for recycling ground tire rubber (GTR) into liquid using an environmentally benign process. The liquefaction was carried out at different temperatures (140, 160 and 180 °C) over variable time ranges (2–24 h) by blending the GTR with aromatic oil in a range from 0 to 100 parts per hundred rubber (phr). The liquefied GTR was separated into sol (the soluble fraction of rubber which can be extracted with toluene) and gel fractions (the solid fraction obtained after extraction) to evaluate the reclaiming efficiency. It was discovered that the percentage of the sol fraction increased with time, swelling ratio and temperature. Liquefied rubber was obtained with a high sol fraction (68.34 wt%) at 140 °C. Simultaneously, separation of nano-sized carbon black from the rubber networks occurred. The separation of carbon black from the network is the result of significant damage to the cross-linked-network that occurs throughout the liquefaction process. During liquefaction, a competitive reaction between main chain scission and cross-link bond breakage takes place.

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1. Introduction

Vulcanized rubber was developed in the 19th century and has become one of the most widely used polymers across the world. It has been utilized in numerous industrial products, especially in tires (Xiang et al., 2015). Estimated data suggests that the growth in worldwide demand for tires, increasing approximately 4.3% per year, reached nearly 1 billion units in 2015 and 2.9 billion units in 2017 (Machin et al., 2017). Concurrently, the generation of the amount of waste tires has increased significantly, with approximately 17 million tons generated annually around the world (Czajczyńska et al., 2017; Luo and Feng, 2017). Thus, the increasingly rubber consumption pose a serious threat to the environment because of the limited number of efficient processes for the disposal of scrap material.

Waste rubber recycling is the preferred method for disposal of waste rubber and is supported both legislatively and economically. GTR has the potential to be used as an alternative to rubber (Saiwari et al., 2012) and rubberized asphalt for road construction (Yan et al., 2016). A number of studies have reported the use of GTR in rubber stocks, including natural rubber (NR), butadiene (BR), styrene/butadiene rubber (SBR), etc. The use of 20% reclaimed GTR has been shown to improve the mechanical performance of BR/GTR and SBR/GTR, for example, the model tire compound (BR/IR/SBR) has been prepared with less than 20 wt% GTR content

(Grigoryeva et al., 2004). Research concerning the incorporation of GTR in NR demonstrated that good tensile strength and abrasion resistance were maintained, especially when smaller particles within a 10 phr loading were used (Li et al., 2004). In order to avoid degrading the basic properties of the materials, it has been suggested that GTR should be subjected to devulcanization or reclamation to enhance the GTR loading content; otherwise, the amount of GTR incorporated into fresh rubber will not surpass 10 phr (Karger-Kocsis et al., 2013).

It has also been widely reported that rubberized asphalt significantly improves the mechanical properties of asphalt pavement, leading to high performance in terms of resistance to rutting, cracking, water and fatigue damage. In addition, rubberized asphalt pavement also assists in improving traffic safety and reducing traffic noise (Ameli et al., 2016; Lo Presti et al., 2012; Nazzal et al., 2016). As rubber chains have a chemically cross-linked-network structure, endowed by sulfur cross-linking and reinforced by carbon black, their devulcanization and swelling in asphalt is limited. The lack of interaction between the asphalt and rubber particles causes the rubber to act as only dispersed filler within the asphalt (Gawel et al., 2006; Ghavibazoo and Abdelrahman, 2013) rather than as an effective network-forming modifier. Furthermore, the density of rubber is greater than that of asphalt, which often leads to the poor stability of rubberized asphalt (Lo Presti, 2013). In order to improve the compatibility between asphalt and GTR, pre-devulcanized or reclaimed rubber can be used to prepare rubberized asphalt with better stability and less pollution (Yao et al., 2015).

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The reclamation of GTR involves the conversion of waste vulcanized rubber from the thermoset state to the plastic and viscous states, allowing for the rubber to be more easily processed and re-vulcanized. Therefore, the reclamation of waste rubber can be considered to a process in which the cross-linked network structure of vulcanized rubber is damaged or degraded due to the effects of heat, oxygen, mechanical force and reclaiming agents. Nowadays, the various methods used to reclaim rubber can be broadly classified into three groups: (1) chemical reclamation; (2) physical reclamation; and (3) biological reclamation (Saiwari et al., 2012; Shi et al., 2013; Verbruggen et al., 1999). The majority of previous studies have focused on reclaimed rubber consisting of approximately 20% sol fraction (Formela et al., 2014, 2016a). This type of reclaimed rubber has mostly been used in tires in the form of physical filling with carbon black bound in the rubber's cross-linked network. However, several research groups have started to investigate the use of reclaimed rubber that possesses a high sol fraction content. Such rubber can be applied as a plasticizer in tire rubber and as an extender in rubberized asphalt (highly liquefied rubber is defined as liquefied rubber with possessing over 50% sol fraction content). A co-rotating twin-screw extruder has been used to obtain liquefied rubber at 300 °C consisting of 73.5% sol fraction of 73.5% (Shi et al., 2014). Wu et al. conducted a liquefaction using a temperature-controlled hydraulic press and obtained rubber with over 80 wt% soluble fraction at a temperature of 280 °C (Wu et al., 2017, 2016).

Common processing conditions for rubber liquefaction rely on fairly harsh conditions, such as a high temperature (220~300 °C), a high pressure and a strong shearing force. High temperatures usually give rise to the generation of malodorous gases consisting of some components of the rubber or rubber additives volatilizing and acting as secondary pollutants. Volatile substances released during the liquefaction process include benzene, toluene, ethylbenzene, xylene and other benzene compounds (BTEX) and sulfides, which are harmful to people's health (Formela et al., 2016b; Martínez et al., 2014; Rajan et al., 2006). Accordingly, a low cost, highly efficient, easy to operate and environmentally benign GTR liquefaction process is desired.

Plasticizers such as aromatic oil and pine oil are added to GTR before devulcanization. For general purposes, these oils are used to improve the diffusion and dispersion of the reclaiming agents. Swelling of GTR with oil containing the chemicals facilitates the penetration of the reactive chemicals into the rubber matrix (Formela et al., 2016b). Additionally, it has been reported to accelerate the oxidation of the rubber and to prevent the formation of gel that acts as a radical acceptor (Rajan et al., 2006). It was found that the sol fraction content of reclaimed rubbers progressively increases with following increase in oil content (in range: 0–20 phr) (Xu et al., 2014). In this study, a greater quantity of oil has been added to the reaction system in order to achieve liquefaction of waste rubber. It is envisaged that the liquefaction of rubber at low temperatures and with high oil content can be achieved due to promotion of liquefaction caused by the pre-swelling of GTR by the oil. Thereafter, the highly liquefied rubber can be used as a plasticizer alternative for tire rubber and asphalt.

An additional part of this work focuses on the presence of carbon black in the liquefied rubber. Carbon black is added to rubber for reinforcing the three-dimensional cross-link rubber network because of the strong physical and chemical reaction between them. It is generally believed that the rubber that is present outside of the carbon black exists in three different states: primary bound rubber, loosely bound rubber, and the free rubber chains (Leblanc, 2002; Li et al., 2016a). Bound rubber is formed through interactions involving physical bonding via vander Waals forces, and polymer-polymer and carbon black-polymer chemical covalent bonding during the compounding process; it cannot be dis-

solved by the solvent that is used for rubber (Gabriel et al., 2016; Litvinov et al., 2011). The interactions between rubber and carbon black influence the separation of rubber from carbon black during the liquefaction of GTR.

2. Experimental

2.1. Materials

GTR (500–600 μm) was obtained by shredding and grinding scrap rubber at ambient temperature by Jiangsu Baoli International Investment Co., Ltd (Jiangyin, China). The GTR consists of 8.24 wt% soluble material, 41.46 wt% natural rubber (NR), 12.09 wt% synthetic rubber (SR), 31.59 wt% carbon black and 6.62 wt% inorganic filler. The aromatic oil used for the experiments was furfural extract oil of commercial grade and was supplied by Shanghai Gaoqiao Petrochemical Co., Ltd (Shanghai, China), and the percentage of aromatic hydrocarbon compounds was 65%.

2.2. Liquefying process of GTR

The GTR (m_G) and aromatic oil (m_a) were weighed and mixed at room temperature in certain proportions (1/1 signifies that the ratio of GTR to oil is 1:1, 2/1 is 2:1, 3/1 is 3:1, etc.). The blends were first stirred for approximately 3 min, then placed in an oven with hot air at constant temperature for liquefaction. The liquefaction of GTR was conducted at low temperature (140–180 °C), for a short period of time (2–24 h) and with a variable oil content (0–100 phr). Traditionally, the content of oil used in the devulcanization of GTR is less than 20 phr. To enhance the liquefaction effect and reduce the liquefaction temperature, a high oil content was selected. The experimental conditions for the samples are presented in Table 1.

2.3. Sol-gel analysis

Both the content of sol fraction and the gel fraction of the liquefied rubbers were determined in order to carry out a comprehensive investigation of the relationship between liquefaction conditions and the degradation efficiency of GTR. The blends of GTR and aromatic oil were enclosed in filter paper and weighed (m_0) before liquefaction. Then liquefied samples were subjected to Soxhlet extraction in toluene for 72 h to remove the majority of the soluble low molecular weight compounds and to separate the sol fraction from the products. The residues were dried in a vacuum oven at 60 °C to constant weight and weighed again (m_1), and were taken to be the gel fractions. It is worth noting that the aromatic oil (m_a) and soluble material (m_b) of the original GTR, liquefied rubber were both extracted with toluene, thus they were deducted when calculating the content. In addition, the mass of rubber (m_r) in GTR was not equal to that of m_G , this can be

Table 1
Samples and corresponding experimental conditions.

Sample	Oil content (phr)	Temperature (°C)	Time (h)
1/1	100	/	/
1/1-140-2	100	140	2
1/1-140-4	100	140	4
1/1-140-6	100	140	6
1/1-140-8	100	140	8
1/1-140-12	100	140	12
1/1-140-24	100	140	24
1/1-160-4	100	160	4
1/1-180-4	100	180	4
2/1-140-4	50	140	4
3/1-140-4	33.3	140	4

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