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Chemical and microscopic investigation of co-pyrolysis of crumb tire rubber with waste cooking oil at mild temperature

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ABSTRACT

Approximate rubber/bitumen homogeneous system formed by desulfurization and degradation of crumb tire rubber in bitumen under high temperature is beneficial to enhance the storage stability of rubberized bitumen. However, the main problems during the processing of desulfurized and degraded rubberized bitumen are aging caused by volatilization of light components, and burning or explosion due to the direct utilization of low flash point bitumen. Therefore, waste cooking oil was proposed as a safer medium to desulfurize and degrade crumb rubber prior to production of rubberized bitumen. This study focused on the feasibility and effectiveness of the application of waste cooking oil in desulfurizing and degrading rubber particles through co-pyrolysis of them at mild temperature (240–280 °C). Chemical and microscopic analyses were performed to investigate the structural changes of vulcanized rubber. Results showed that solubility of rubber powder reached above 60 wt% after pyrolysis in waste cooking oil, which increased with higher temperatures and more of oil, while increased to a maximum at 2 h and then decreased with the extension of time. The rubber hydrocarbon content decreased greatly, and dramatic reduction of carbon, hydrogen and sulfur elements happened according to component and elemental analyses. The surface of pyrolysis product was even and smooth without obvious rubber particles. The grooves and cavities of rubber residues in scanning electron microscopy micrographs proved that shedding of degraded polymer molecules occurred. Fourier transform infrared spectra revealed that breakage of carbon-sulfur, carbon=carbon and sulfur=oxygen bonds took place during pyrolysis, with appearance of natural rubber characteristic peak.

1. Introduction

Over the past decades, the output of waste materials including end-of-life tires and waste cooking oil (WCO) has dramatically increased worldwide (Rahman et al., 2017; Singhackbarali et al., 2017). The improper disposal of a significant quantity of hazardous wastes may pose a severe threat to the environment (Charpe and Rathod, 2011), and to the human health mainly through air, water and food chain eventually (Panadare and Rathod, 2015). Besides, random discard of waste materials is an extreme waste of resources. Consequently, seeking for the potential ways to recycle wastes has become a vital and urgent issue from both energy-saving and environmental protection perspectives (Zanetti et al., 2015; Chen et al., 2018).

The application of crumb tire rubber (CTR) derived from grinding end-of-life tires as a bitumen modifier in road pavement has been shown to be a promising way to address waste management. Crumb tire rubber-modified asphalt (CTRMA) has superiority in resistance to rutting, intensive crack and fatigue, as well as noise reduction and cost saving, etc. (Yin et al., 2013; Feng and Hu, 2015; Xu et al., 2016). However, the poor storage stability of CTRMA affects the service performance of the pavement (Liang et al., 2015; Tang et al., 2016). Terminal Blend (TB) rubberized bitumen technology is considered as a potential method in improving storage stability of CTRMA due to the full digestion of rubber powder in bitumen, while maintaining considerable performance in pavement practice (Tang et al., 2016; Lin et al., 2017). In TB technology, cross-linked rubber molecules were desulfurized and degraded in neat bitumen as a solvent under 240–260 °C for more than 6 h. However, this method has a limit on the application of low flash point (<230 °C) bitumen because it is easy to burn or explode at processing site if the bitumen is under high temperature for a long time, and to cause aging of bitumen due to the volatilization of light components.

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Nomenclature

a	relative amount of the main chemical elements
A	absolute amount of the main chemical elements
C	carbon
CTR	crumb tire rubber
CTRMA	crumb tire rubber-modified asphalt
DTG	differential thermogravimetry
FTIR	Fourier transform infrared spectroscopy
GC–MS	gas chromatography-mass spectrometry
H	hydrogen
m_1	initial weight of the sample
m_2	weight of the sample residue
N	nitrogen
NR	natural rubber
O	oxygen
PRB	pyrolysis product of crumb tire rubber with neat bitumen
PRO	pyrolysis product of crumb tire rubber with waste cooking oil
r	proportion of crumb tire rubber in the binary mixture
S	sulfur
S_0	sol content of original crumb tire rubber
S_1	sol content of pyrolysis product
S_i	sol content of original crumb tire rubber or pyrolysis product
SEM	scanning electron microscopy
SR	synthetic rubber
TB	Terminal Blend
TGA	thermogravimetric analysis
WCO	waste cooking oil

According to the modification mechanism of TB rubberized bitumen, using solvents to desulfurize and degrade CTR is the best way because it is easy to control reaction process and to avoid environmental pollution in liquid environment (Khattak and Syme, 2009). Therefore, it is necessary to replace bitumen by a safer solvent prior to further modification of neat bitumen.

Yin et al. (2013) indicated that the short-term aging resistance of bitumen was effectively improved by addition of CTR impregnated in epoxidized soybean oil. The utilization of periodic acid as a solvent for oxidative degradation of tire rubber was described in the research of Sadaka et al. (2012). It can be found from their study that the choice of solvent should consider processing cost, conditions of experiments, efficiency and the controllability. In this sense, waste cooking oil may be a suitable solvent to desulfurize and degrade CTR owing to its low price, wide distribution, easy procurement and non-toxic. It is reported that partial replacement of bitumen by WCO would reduce carbon emission and pollution by bitumen and decrease the risk of breathing bitumen fume (Rahman et al., 2017), while maintain a good compatibility between WCO with neat bitumen. More importantly, it is safer to use WCO due to its higher flash point than neat bitumen (20 °C higher than neat bitumen used in this study). The application of this alternative idea could be a supplement of the disposal methods of these high-quality wastes. Nevertheless, to our knowledge, there are few reports concerned with the use of WCO in CTR desulfurization and degradation before the modification of bitumen.

Pyrolysis is the thermal degradation of the organic components (Williams, 2013), at typical running temperature (around 500–550 °C) (Chen et al., 2014) to break the long chain hydrocarbons into intermediate and small chain hydrocarbons (Prathiba et al., 2018). The aim of tire pyrolysis is obtain value-added pyrolysis oils, gases and char which can be used as fuels or the sources of chemical products (Tan et al., 2018). Pyrolysis can be classified into conventional pyrolysis and fast pyrolysis depending upon the heating rate, residence time and temperature (Zolezzi et al., 2004). However, neither type of pyrolysis is unsuitable for preparation of rubber modifier of bitumen because the chemical structure of rubbers would be totally destroyed through high-temperature pyrolysis. Pyrolysis at mild temperature allows production of degraded rubber with similar chemical structure to original rubber and high molecular weight, which contributes to improve high- and low-temperature properties of bitumen and to increase of the wastes dosage in bitumen (Wu et al., 2017). The pyrolysis of cross-linked rubber at 220–300 °C was studied by Wu et al. (2016) and Song et al. (2018) using twin-screw extruder and hydraulic press, respectively. Desulfurization and degradation of cross-linked rubber were realized under different levels in these methods. However, it is not easy to control the breakage degree of cross links due to the powerful mechanical force and short curing time.

Co-pyrolysis of crumb tire rubber with waste cooking oils at mild temperature (lower than the flash point of WCO, <300 °C) was proposed in this study. The common pyrolysis of various wastes is considered as an environmentally friendly way for transformation of hazardous wastes into valuable products. Co-pyrolysis of sawdust with waste glycerol was performed by Bartocci et al. (2017) to enhance the energy value of biomass, while co-pyrolysis of tires with coals was used to improve the production of liquid products (Mastral et al., 2001) or increase the char yield (Acevedo et al., 2013). The liquid fuels could be obtained from the co-pyrolysis of tires with oily wastes in the research of Önenç et al. (2012). In the future, co-pyrolysis product of CTR and WCO can be applied in modified bitumen as a novel modifier. The early aging of bitumen due to the volatilization of the light components in TB technology can be avoid through this way. Besides, the lower temperature (170–190 °C) and shorter time (1 h) which are consistent with traditional wet-process technique can be applied in the production of modified bitumen. However, there are few studies focused on this topic, and the evolution of CTR after treating by WCO has not been fully understood.

Accordingly, the main objective of this study is to develop alternative method for crumb rubber desulfurization and degradation considering wastes disposal, production safety and cost-saving. For this purpose, co-pyrolysis of crumb tire rubber with waste cooking oil was performed at various mild temperatures (in range: 240–280 °C) and times (in range: 0.5–4 h). Further, in order to obtain a better understanding of the influences of pyrolysis on chemical and microscopic properties of rubber, solubility test was used to evaluate the degradation degree of cross-linked rubber. Thermal and elemental analyses were used to explore the chemical composition of rubber. Fourier transform infrared spectroscopy was carried out to characterize the functional groups of raw materials and pyrolysis products. Scanning electron microscopy was conducted to describe the surface morphology of pyrolysis product.

2. Materials and methods

2.1. Materials

Crumb tire rubber (CTR) of 40 mesh particle size was obtained by grinding truck radial tire at ambient temperature. The CTR is