



Modification of thermal and rheological characteristics of bitumen by waste PET/GTR blends



Marzieh Habibi Karahrodi^{a,*}, Omid Moini Jazani^{a,*}, Seyed Mohammad Reza Paran^b, Krzysztof Formela^{c,*}, Mohammad Reza Saeb^d

^a Department of Chemical Engineering, College of Engineering, University of Isfahan, Isfahan, Iran

^b Department of Polymer Processing, Iran Polymer and Petrochemical Institute, P.O. Box 14965/115, Tehran, Iran

^c Department of Polymer Technology, Faculty of Chemistry, Gdansk University of Technology, Gdansk, Poland

^d Department of Resin and Additives, Institute for Color Science and Technology, P.O. Box: 16765-654, Teheran, Iran

HIGHLIGHTS

- Waste PET/GTR blends obtained via melt-compounding were used as bitumen modifiers.
- Performance properties of modified bitumen depends strongly from composition and content of wPET/GTR blends.
- wPET/GTR blends improved rheological properties and thermal stability of bitumen.

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ABSTRACT

Waste poly(ethylene terephthalate) (wPET)/ground tire rubber (GTR) blends with variable compositions were applied as low-cost modifiers into bitumen and their performance has been evaluated via morphological, thermal and rheological analyses. Depending on the weight ratio of wPET/GTR, dispersion of single/composite domains of waste polymers in the bitumen has promoted the interaction between bitumen and blend components. Fourier transform infrared spectroscopy confirmed an adequate physical interaction between bitumen and wPET/GTR blend, while morphological analyses suggested deterioration of interaction upon increase of GTR content, which can be ascribed to the cross-linked structure of GTR. Increasing wPET/GTR content led to an improved rheological properties and thermal stability of the bitumen, especially for wPET/GTR blends composed mainly of wPET due to its thermoplastic character. The present work indicates that rheological and thermal properties of bitumen can be satisfactorily modified varying composition and content of wPET/GTR in the bitumen matrix.

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1. Introduction

Bitumen, by-product of crude oil fractionation, increasingly finds widespread use in road pavements, sealants, binders, waterproof coatings or other civil/engineering applications [1]. The estimated data shows that current world production of bitumen is 113 Mton per year and is expected to reach 135 Mton in 2020, exceeding by a 2.7% annual growth rate from 2014 to 2020 [2]. Bitumen is colloidal dispersion of asphaltene into maltene matrix, constituted by saturates, aromatics, resins and asphaltenes [3]. The complex chemical composition of bitumen and its variability is related to the source/type of crude oil and the manufacturing process, which

also affects internal structure of bitumen [4]. Consequently, the performance properties of bitumen are normally very sensitive to manufacturing conditions. One of the most commonly method used to resolve this burning issue is physical or chemical modification of bitumen by polymers [5]. Generally, introduction of polymers improves the stiffness of bitumen at high temperatures, its resistance against moisture and cracking at low temperatures, which prolongs its fatigue life. This has encouraged researchers and engineers to search for new effective and pro-ecologic methods of bitumen modification, thereby allowing for tailoring performance properties of bitumen [6–8].

At the turn of the last few years, diverse polymers including plastomers (e.g. poly(ethylene) (PE) [9], poly(propylene) (PP) [10], poly(ethylene-vinyl acetate) (EVA) [11], poly(styrene-acrylonitrile) (SAN) [12], poly(ethylene terephthalate) (PET) [13]), elastomers (e.g. natural rubber (NR) [14], styrene-butadiene

* Corresponding authors.

E-mail addresses: o.moini@eng.ui.ac.ir (O.M. Jazani), krzysztof.formela@pg.gda.pl (K. Formela).

rubber (SBR) [15], ethylene-propylene-diene rubber [16], etc.) and thermoplastic elastomers (e.g. poly(styrene-butadienestyrene) (SBS) [17], poly(styrene-ethylene/butylene-styrene) (SEBS) [18] and poly(styrene-isoprene-styrene) (SIS) [19]) were applied as bitumen modifiers. However, due to environmental and economic factors, interesting alternative is ground tire rubber (GTR) obtained during mechanical disintegration of waste tires. Estimated data shows that more than 17 Mton used car tires are discarded every year worldwide [20]. Made from high-quality of rubbers and reinforcing fillers (carbon black, silica), used tires and other waste rubber goods can be used as valuable source of secondary raw materials during preparation of different kinds of polymer blends/composites [21,22].

Building industry, especially in modification of cements or road bitumens, is among applications of GTR at an industrial scale. Comprehensive literature reviews of the research on the modification of bitumen with GTR were recently presented in works [23,24]. The results of the research performed over the last few decades by independent research groups confirm that addition of GTR as modifier to bitumen enhances elasticity and fatigue resistance of road pavements, which prolongs life fatigue of bitumen and reduces maintenance cost compared to unmodified road pavements. Furthermore, use of bitumen modified with GTR reduces the noise emission generated during car transport and improves the comfort and safety of driving through shortening of breaking distance. On the other hand, factors limiting application of bitumens modified with GTR are high production costs resulting from high-energy consumption of the process and modification or adjustment of commonly used apparatus. Moreover, cross-linked structure of waste rubber brings some disadvantages like low stability (related to sedimentation of GTR particles at elevated temperature) and high viscosity of GTR-modified bitumen, and therefore, limits storage time and further applications of the resulting compounds.

A cost-efficient method to increase homogeneity and stability of GTR-modified bitumen is using additives (e.g. curing additives, waxes, plasticizers, thermoplastics polymers, etc.), which may improve solubility and dispersion of GTR in bitumen [25–27]. Use of a preliminary reclaiming/devulcanization or oxidation of GTR would be another strategy for this purpose [28–30]. Consequently, a combined method to improve performance properties and stability of the modified bitumen seems to be promising for further research in this field.

Yan et al. [31] studied the effect of GTR and recycled low-density poly(ethylene) (LDPE), individual and combined, on the rheological properties of bitumen. The authors emphasized that rheological properties of modified bitumens are strongly dependent on the recycled LDPE or GTR content. It was also reported that an enhanced mechanical properties can be achieved using a combination of both aforementioned modifiers. Zhang and Hu [32] used PE, GTR and dioctyl phthalate as modifiers of bitumen. The obtained results led to conclusion that PE is responsible for achieving high-temperature performance of bitumen, while addition of dioctyl phthalate as a plasticizer improves its low-temperature properties. Fang et al. [33] used GTR and EVA as bitumen modifiers. The results indicated that the used modifiers significantly enhance the performance properties and stability of bitumen.

Recently, Wang et al. [34,35] proposed use of PE and reclaimed rubber blends as modifiers of bitumen. They proved that blending of thermoplastics with reclaimed rubber *via* extrusion improves the storage stability of PE/reclaimed rubber – modified bitumen, which was related to improved dispersion of modifiers into bitumen matrix. Similar observations were reported by Xiao-qing et al. [36] when studying the effect of mechanochemically reclaimed GTR and SBS blends on properties of modified bitumen. The authors proved that reclaimed GTR/SBS blends had a better

interfacial adhesion with bitumen matrix than the SBS alone. Our recent studies [37] demonstrate that composition of thermoplastics/GTR blends and interfacial interactions between the used components have a crucial impact on the final properties of bitumen. This means that conventional, thermal and rheological properties of modified bitumen can be improved and easily modified by changing composition of thermoplastics/GTR blends. However, as discussed above, reports in this field are relatively new and still very limited [38].

In the present work, waste poly(ethylene terephthalate) (wPET)/ground tire rubber (GTR) blends with different composition were applied at variable content as low-cost bitumen modifiers to improve processing and performance properties of bitumen. Thermoplastic modifiers based on wPET/GTR blends were obtained *via* melt-compounding using a co-rotating twin screw extruder. This continuous process, commonly used in the industry, allows partial reclaiming of GTR [39–41] and promotes significantly interaction between thermoplastic polymer and bitumen matrix. Morphology (optical microscopy), chemical composition (Fourier transform infrared spectroscopy – FTIR), thermal properties (thermogravimetric analysis – TGA) and rheological properties (dynamic shear rheometer tests) of the modified bitumen were investigated and discussed.

2. Experimental

2.1. Materials

Road bitumen 60/70 was received from Jey Oil Refining Co. (Isfahan, Iran). Characteristics of the used bitumen are presented in Table 1.

The wPET was purchased from Isatiss Rubber Co. (Iran). The physical properties of the wPET are measured using standard test methods. It has a density of 1.08 g/cm³, glass transition temperature of 70 °C and melting point of 250 °C. GTR with particles size below 0.8 mm was received from Isatiss Rubber Co. (Iran). GTR was obtained during ambient grinding of the used tires (mix of passenger car tires and truck tires). All ingredients mentioned above were used as received.

2.2. Sample preparation

2.2.1. Step I: Preparation of wPET/GTR blends

The wPET was dried at 80 °C for 4 h in a vacuum oven before wPET/GTR blends preparation. The blends with various ratios of wPET/GTR were prepared, as in Table 2, through a direct melt compounding using a co-rotating twin screw extruder from Brabender Co. Model TSE 20 (L/D = 40, D = 20 mm) at 260 °C and 65 rpm. wPET and GTR were dosed directly into hopper with constant throughput 1.25 kg/h.

2.2.2. Step II. Modification of bitumen

Samples were prepared using a laboratory high shear mixer (Silverson Co. model L4R) at 3000 rpm. The bitumen was heated at 150 °C until complete melting was received to obtain low-viscosity bitumen ready for mixing with wPET/GTR blends. The

Table 1
Basic specifications of the used bitumen.

Property	Standard	Value
Density, g/cm ³	EN 15326	1.0145
Penetration at 25 °C, dmm	EN 1426	66
Softening point, °C	EN 1427	51
Flash point, °C	EN ISO 2592	≥232
Change of mass (after aging), wt%	EN 12607-1	0.2≤

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