

Use of coupling agents to stabilize asphalt–rubber–gravel composite to improve its mechanical properties

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ABSTRACT

The mechanical performance of rubber-modified asphalt roads depends, besides the intrinsic properties of the constituents (asphalt, rubber and gravel), on their interfaces. To improve the adhesion between constituents, two different coupling agents were required: one to link asphalt with the rubber particles to stabilize the composite creating an elastic network in the interior of the material to improve its elastic recovery, and other to increase the adhesion between the hydrophobic asphalt and the hydrophilic gravel. A phenolic resin was used to link the rubber particles with the asphalt, while a zwitterionic molecule (phosphatidyl-choline) was used to change the OH groups on the gravel surface by hydrocarbon chains to make it compatible with the asphalt.

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1. Introduction

New polymeric materials with novel properties can be obtained by mixing two or more immiscible polymers which possess, separately, the required properties for the new material. However to obtain high performance materials it is also required to have a good adhesion between phases [1–4]. From these composites, special attention deserves asphalt-based materials. As known, asphalt is one of the most produced organic polymeric materials in the world; it is an important low-cost thermo-plastic material widely used for construction. Asphalt-based materials are widely used in a variety of important applications like: roads binders, adhesives, sealants, and waterproof coatings among others [5]. As a building material, asphalt is exposed to a wide range of loads and weather conditions. However, this material has poor mechanical properties because it is hard and brittle in cold weather, soft and fluid in hot environment and has a low elastic recovery; the brittle character of the asphalt makes it easy to fracture when it is subjected to stresses. For these reasons asphalt is usually reinforced with different types of rubbers to improve its mechanical properties [6–8].

A rubber-containing molecular structure widely used to improve the mechanical properties of asphalt (specifically the elastic recovery) is styrene–butadiene block copolymers (SBR). The

strong incompatibility between polybutadiene and polystyrene produces, inside the asphalt, an elastic network that renders in a material with excellent elastic recovery and improved mechanical properties [9–11].

1.1. Asphalt–rubber coupling agent

An important rubber-based material used in vehicle industry is the tire rubber. Tires possess a complicated structure with a complex design and are made of different types of rubbers. The huge increase in the number of vehicles and aircraft means a huge number of tires that, once used, are discarded producing a huge contamination. Proper disposal of used tires is a technological challenge because non-pollution combustion processes are expensive. A partial solution to reduce the pollution produced by used tires and to improve the quality of the asphalt carpet in roads and highways is to modify the asphalt with rubber particles; these must be homogeneously dispersed and with good adhesion to asphalt [12–15].

Phenol–aldehyde (PA) resins are a family of thermoset compounds of high molecular weight where the phenol rings are linked each other by CH₂ groups (Fig. 1). These resins have been widely used as a binder in a large number of applications: paper, wood powder, husk rice, vulcanization, etc. When a phenol is reacted with an aldehyde, there are three positions in the phenol group susceptible to chemical attack by the aldehyde molecule producing a three functional molecule which can be used to

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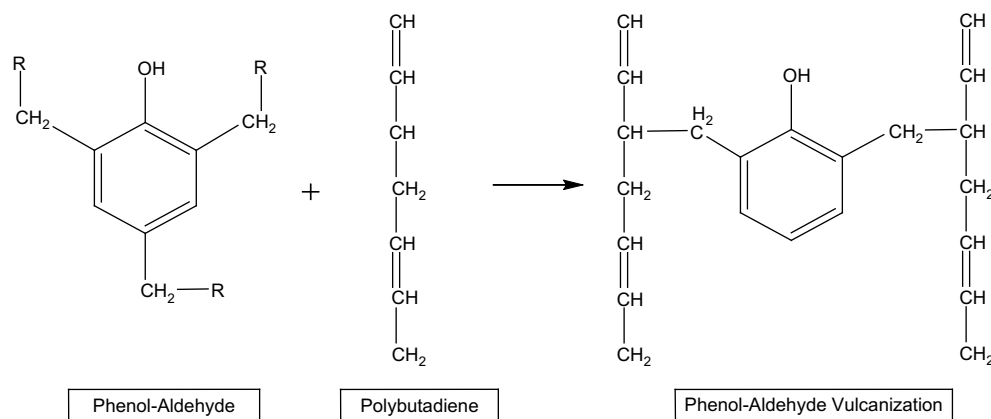


Fig. 1. Chemical reaction between PA resin and polybutadiene.

generate a highly crosslinked material. Due to the double bond of the polybutadiene molecule, the PA resin reacts chemically with the allyl hydrogen of the allyl group (C_3H_5) producing a highly crosslinked material due to the multifunctional character of the PA molecules. Additionally it is possible, by adjusting the R group (Fig. 1), to obtain materials with different properties (melting temperatures, reactivities, etc.) to react with the asphalt–rubber compound [16–18]. Due to the complex chemical structure of asphalt, no schematic representation is shown for the link of asphalt with rubber particles through the use of PA resin; however this is carried out similarly as the reaction between PA and the polybutadiene as shown in Fig. 1. The vulcanization reaction produced by the PA is similar in nature to the one carried out using sulfur, however due to the multifunctional character of PA, this is more effective [19,20]. The other possibility for the vulcanization reaction, with lower probability to occur, is carried out through the reaction of the PA with the double bond forming quinone groups.

1.2. Asphalt–gravel coupling agent

Gravel has been used in pavements from long ago with two main purposes: because is a hard material it protects the pavement against shear and compression stresses produced by the vehicles and, because is a ceramic, it protects pavement against degradation by UV radiation [21–23]. This is an important component in the pavement construction, and then it is necessary to assure a good adhesion between gravel and asphalt.

Gravel is obtained from igneous rocks by a grinding process to a desired particle size. This mineral has several phases and stoichiometry. It is typical to use basalt or granite with a Si:Al molar ratio of 3:1 and 4:1, respectively. These rock minerals are stable under the condition in which they were formed, i.e. at the temperature of molten lava; however they are unstable under the chemical and physical conditions at the earth's surface. These materials are slowly transformed by weathering (exposure to water, oxygen and CO_2) changing the initial unstable chemical structure in a more stable one where some of the chemical groups are hydrolyzed to form the groups $\equiv Si-OH$ and $\equiv Al-OH$. The complicated and varied structures of these rock minerals are due to different organizations of the basic silicon–oxygen tetrahedral and the aluminum–oxygen tetrahedral and octahedral structures [24].

Once the asphalt has been compacted, a uniform layer of gravel with specific granulometry is added; this is inlaid to the asphalt by pressure producing only a physical adhesion between phases. The adhesion between the asphalt and the gravel is difficult because the gravel is a hydrophilic inorganic material, while the

asphalt is a hydrophobic organic compound. Then it is required to change the hydrophilic character of the gravel surface into a hydrophobic one; this can be done by changing the OH groups on gravel by hydrocarbon chains. To do this the coupling agent must have in its structure a polar part with either ionic or zwitterionic groups to react with the OH groups of the gravel, and a hydrocarbon chain to provide the hydrophobic character to react appropriately with the asphalt. In this case the selected molecule was a zwitterionic compound with two fatty acids of long chain to provide the hydrophobic character, while the polar part consists of a strong dipole moment to react with the OH groups of the gravel. The phosphatidyl-choline (PC) is an inexpensive commercial available material of vegetal origin which possesses these characteristics [25,26]. The strong dipole moment of this molecule results from a positive charge from the nitrogen and the negative charge from the free oxygen linked to the phosphorous and with a distance between charges of around 7 Å. This large distance between charges increases the probability of link with the groups $\equiv Si-O^-$ or $\equiv Si-OHH^+$ (and the corresponding Al groups) present in the gravel surface.

The aim in this work is to use two different coupling agents to improve the mechanical properties of asphalt carpets: PA to link the rubber particles with the asphalt to produce an elastic network into the asphalt to improve its elastic recovery and mechanical properties; the other coupling agent (PC) to link the gravel with the rubber-modified asphalt to stabilize the composite.

2. Experimental

2.1. Samples preparation

The raw materials were characterized before the fabrication of the composites. The asphalt was a non-oxidized commercial asphalt AC-20 (Asphalt 6, PEMEX, México). The rubber particles were obtained by a non-cryogenic grinding process with an average particle size of 356 microns. The asphalt was molten at $(180 \pm 10)^\circ C$ and the rubber powder was added slowly and with mild agitation. Once the rubber particles were uniformly distributed into the asphalt, the PA powder was slowly added to obtain a homogeneous; then temperature was reduced.

Several samples were prepared with different compositions (Table 1). These samples were named as: Asphalt:PA:Rubber:PC:-Gravel. For example the sample A3:15:3:15:7F was prepared using asphalt 30% wt, crosslinking agent 0.15%, rubber particles 3%, coupling agent 0.15%, functionalized (F) gravel 70% wt.

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