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Modification of bitumen by electron beam irradiated recycled low density polyethylene

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HIGHLIGHTS

- The modification of bitumen with electron irradiated recycled LDPE was investigated.
- Electron irradiation was applied on LDPE to provide a chemical interaction between LDPE and bitumen.
- The results revealed LDPE modification provides an improved rheological properties of bitumen.
- e-LDPE$_R$ modifier has a certain favorable effects on stiffening of bitumen.

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ABSTRACT

This paper describe the modification of bitumen with electron irradiated recycled low density polyethylene (e-LDPE$_R$). Electron beam irradiation was applied to recycled LDPE (LDPE$_R$) to provide formation of free radicals and some functional groups that may contribute to creation of strong chemical bonds between polymer modifier and bitumen. Five binders having different e-LDPE$_R$ polymer content of 1%, 3%, 5%, 7% and 9% were prepared. The effects of the e-LDPE$_R$ on bitumen were investigated by means of morphological, chemical and physical testing program, including Fourier transform infrared (FTIR) spectroscopy, differential scanning calorimetry (DSC), thermogravimetric analysis (TGA) conventional tests, rolling thin film oven (RTFOT), rotational viscosity (RV), dynamic shear rheometer (DSR) tests.

Based on the results, it can be noted that e-LDPE$_R$ modification provides an improved fundamental rheological properties of bitumen.

1. Introduction

Bitumen is a material used in many engineering applications especially in flexible highway pavements. Due to its viscoelastic and thermoplastic properties, bitumen behaves like an elastic solid at low temperatures or under rapid loading and like a viscous fluid at high temperatures or under slow loading [1,2]. Polymer modification is a common method to provide enhanced performance of bitumen by decreasing its temperature susceptibility. Styrene butadiene styrene (SBS) and ethylene vinyl acetate (EVA) are the polymers mainly used in bitumen modification [3,4]. In addition to SBS and EVA, various polymers are employed in bituminous materials. Polyolefins; such as low density polyethylene (LDPE), high density polyethylene (HDPE) and polypropylene (PP) have been used as modifier as well [5–7].

However, most of the polymers currently used as additive in bitumen are not economically attractive. Thus, employing recycled materials in flexible highway instead of virgin ones has become a fundamental case for researchers especially in terms of cost efficiency and environmental awareness. To date, many studies have been made to investigate using of recycled polymers in bitumen and generally improved mechanical properties of bitumen were determined [8–10]. However, one of the main problems for modified bitumen is lack of stability due to phase separation in storage condition. Exposing recycled polymers to irradiation can be used to overcome this problem by creating new bonds and functional groups in polymer chain that will further provide chemical bonding of the latter with bitumen preventing phase separation [11].

In this study, the rheological properties of electron beam irradiated recycled LDPE (e-LDPE$_R$) modified bitumen were investigated. Electron beam irradiation was applied to recycled LDPE (LDPE$_R$) to provide formation of free radicals and some functional groups that may contribute to creation of strong chemical bonds between polymer modifier and bitumen. In order to observe the changes in chemical structure of the LDPE after electron beam irradiation, Fourier transform infrared spectroscopy (FTIR) was employed.

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The thermal behavior of $LDPE_R$, e-$LDPE_R$, bitumen and e-$LDPE_R$ modified bitumen samples were studied by DSC method. The corresponding thermal characteristics were investigated by means of TGA. The quality of the dispersion of e-$LDPE_R$ modifier in the bitumen was investigated with fluorescent microscopy. Short aging processes were done with rolling thin oven test (RTFOT). Rotational viscometer (RV) was used to determine viscosity of the base and e-$LDPE_R$ modified bitumen. Rheological parameters, as well as frequency dependence behavior of the original and aged binders were determined with dynamic shear rheometer (DSR).

2. Experimental

2.1. Materials

In this study, the bitumen having 160/220 penetration grade was used as a binder throughout testing program. The $LDPE_R$ was manufactured from post-costumer greenhouse films, which were exposed to sunlight and other external environmental factors in Sicily, Italy at least for 1 year. During the fabrication of the $LDPE_R$, the waste films were all washed, dried and cut to piece by an industrial scale and finally extruded. The physical properties of the $LDPE_R$ were as follows: Melting point (T_m) = 109 °C, modulus of elasticity (E) = 180 MPa, tensile strength (TS) = 16 MPa, Elongation at break (EB) = 500%, melt flow index (MFI) = 190/2.16 = 0.29 g/10 min.

2.2. Electron beam irradiation

Electron beam irradiation is a process using the high energy accelerated electrons to treat a material for a variety purposes such as cross-linking or destruction of polymers, food sterilization, and surface coatings curing [12,13]. Thermal mechanical and chemical improvement of polymers can be achieved with electron irradiation [14]. In this paper, electron irradiation was applied to the surface of the $LDPE_R$ to form free radicals to provide a chemical interaction between $LDPE_R$ polymer and bitumen.

In this work electron irradiation process was carried out through thin aluminum foil windows in the cryostats of EG-2.5 Van-de-Graf accelerator. The spectra were measured in the same cryostats without intermediate heating. The post-irradiation process was monitored to see annealing on the samples at any temperature up to 300 K.

$LDPE_R$ polymer particles were exposed to irradiation (from two uniform surfaces) of the dose of 20 kGy in a case size of 100 × 100 × 40 mm. The dosage of radiation was controlled with chemical dosimeter after irradiation process.

2.3. Preparation of samples

After the electron beam irradiation, e-$LDPE_R$ modifier obtained in a form of pellet was milled with a grinder and the particles were sieved with a sieve of No 50 to work with smaller and homogeneous pieces. Five different binders were prepared with different polymer content ranging from 1% to 9%. High shear mixer was used for the preparation of the samples. 160/220 penetration grade bitumen was heated for 90 min at 163 °C, and then poured into the temperature-controlled flask and mixer adjusted to 500 rpm. Subsequently the e-$LDPE_R$ was added to bitumen by portions in at 15 min intervals, and then the mixing rate was increased to 1300 rpm and mixing was continued for the other 150 min. Temperature of the mixture was controlled with an air thermometer every 15 min. After the end of the mixing process, the samples were removed from the flask and divided into small containers, covered with aluminum foil and stored for testing program. Three different samples were prepared for each test performing on base and modified bitumens. Any statistical approach to compare these three different test results was not required in this case due to low standard deviation. Hence, the mean of results obtained from the samples given as the test result in this paper. The binders used in this study were coded as below;

base bitumen – "B";
base bitumen + 1% e-$LDPE_R$ – "B-1-e-$LDPE_R$";
base bitumen + 3% e-$LDPE_R$ – "B-3-e-$LDPE_R$";
base bitumen + 5% e-$LDPE_R$ – "B-5-e-$LDPE_R$";
base bitumen + 7% e-$LDPE_R$ – "B-7-e-$LDPE_R$";
base bitumen + 9% e-$LDPE_R$ – "B-9-e-$LDPE_R$";

2.4. Testing program

2.4.1. Fourier transform infrared spectroscopy

Fourier transform infrared (FTIR) spectroscopy was used to characterize various functional groups in polymer modifier and compositions. FTIR spectra were recorded with a Bruker Tensor 27 DTGS spectrometer between 4000 and 450 cm^{-1} using the Attenuated Total Reflection (ATR) mode. For each spectrum, 32 consecutive scans with a resolution of 4 cm^{-1} were averaged.

2.4.2. Differential scanning calorimetry (DSC)

Differential scanning calorimetry (DSC) measurements were carried out using a TA Instruments DSC Q2000 calorimeter under a nitrogen atmosphere. Samples with masses of ~10 mg were first heated from −60 °C to 160 °C with a programmed heating rate of 20 °C/min, and subsequently cooled back. A second scan was performed under the same heating conditions. The T_g values were measured in the second run: to determine a T_g range, the lower ($T_{g\ onset}$) and upper ($T_{g\ end}$) limits were identified as the values associated with the intercepts of tangent to midpoint of the specific heat increment with "glassy" and "viscous" baselines, respectively. Melting temperature (T_m), corresponding to the maximum in fusion endotherm, was noted. Heat of fusion (H) was calculated from the area under the endothermic peak and the degree of crystallinity was calculated accepting a melting enthalpy value, H_m = 283 J/g, for the 100% crystalline polyethylene (PE).

2.4.3. Thermogravimetric analysis (TGA)

Thermogravimetric analysis, TGA, was performed on a TA Instruments TGA Q-50 thermo-balance under nitrogen atmosphere. Sample pellets with masses ranging from 10 to 20 mg were heated at 20 °C min^{-1} from 20 °C up to 650 °C.

2.4.4. Morphology of bitumen

The quality of the dispersion of e-$LDPE_R$ modifier in the bitumen was investigated with fluorescent microscopy, which was able to study the morphology of modified bitumens by using the principle in which polymers become swollen after absorbing some of the constituents of the original bitumen [15].

The method of sample preparation for fluorescent microscopy followed the regular procedure consisting of heating, homogenizing and cooling as well as a surface preparation over thin films process. The samples were examined under a Carl Zeiss Primo Star, magnifying up to 1000X, using with a 40 W halogen lamp and blue color filter (d = 45 × 1.5).

2.4.5. Conventional tests

Penetration (ASTM D5), softening point (ASTM D36), ductility (ASTM D 113) tests were carried out on both original and aged base and modified bitumens. After RTFOT, the retained penetrations, softening points and loss of weights of the binders were calculated to understand the effects of the e-$LDPE_R$ modifiers on aging. Penetration index was also calculated in order to investigate the temperature susceptibility of binders [16].

2.4.6. Rotational viscosity test

The rotational viscometer was employed to determine the viscosity of the base and modified bitumens. Unlike the capillary viscometers used with the viscosity-graded method, the rotational viscometer can evaluate viscosity of modified bitumen binders by measuring the torque necessary to maintain a constant rotational speed of (20 RPM) a cylindrical spindle submerged in a bitumen specimen held at a constant temperature [17,18]. The tests were performed at two different temperatures of 135 °C and 165 °C, with a Brookfield viscometer (DVRV-II Pro Extra) as described in AASHTO standard [19]. The mixing and compacting temperatures of the hot mix asphalts are also calculated for base and modified bitumens. The convenient ranges of viscosities corresponding to the mixing and compacting temperatures considered as 150-190 cP and 250-310 cP respectively, in this calculation.

2.4.7. Aging of bitumen

The short term aging processes of the e-$LDPE_R$ modified binders were performed with the rolling thin film oven test (RTFOT) [20]. The binders were aged at 163 °C for 75 min in the oven [21]. After the RTFOT, which simulates the changes in the physical properties of binder during the storage, penetration, softening point tests were applied, and the weight loss was calculated to understand the effects of the short term aging. The aged binders were also tested with DSR for the examination of their viscoelastic properties.

2.4.8. Dynamic mechanical test

The most commonly methods for the fundamental rheological characterization of bitumen are dynamic mechanical methods using oscillatory type testing, which are generally conducted within the region of linear viscoelastic response by using a dynamic shear rheometer (DSR) [22].

DSR is used for the purposes to measure the viscoelastic parameters such as complex modulus (G^*), phase angle (δ) and rutting parameter ($G^*/\sin\delta$) of asphalt binders at intermediate to upper pavement service temperatures. G^* is defined as the ratio of maximum (shear) stress to maximum strain, a measure of the total resistance to deformation when the bitumen is subjected to shear loading. The phase difference between stress and strain in an oscillatory test, named as δ, is a measure of the viscoelastic balance of the material behavior [23].

These parameters are used in Superpave specification providing an indication of the rutting resistance of bitumen immediately following to the construction. In addition to measuring complex modulus and phase angle at a single frequency, the dynamic shear rheometer can also be utilized for a range of frequencies to understand the frequency (shear rate) dependency of behavior of the binders [24]. Hence, DSR are used to invoke time-temperature superposition and to construct thermo-