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# The effect of gamma-irradiated recycled polyolefins as asphalt binder modifiers

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## ABSTRACT

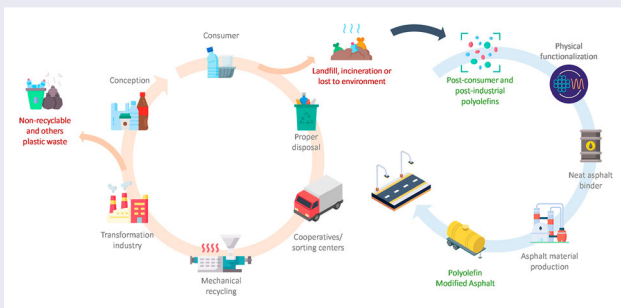
The current study evaluated the potential effects on the rheological properties of an asphalt binder modified with two recycled postconsumer polyolefins, polyethylene (PE) and polypropylene (PP), both with and without exposure to gamma-ray ( $\gamma$ -ray) irradiation, putting out a procedure that ensures benefits concerning the behavior and performance of asphalt pavements. The chosen analytical methods intend to investigate: the polyolefins' chemical and physicochemical properties; the asphalt binders' linear viscoelastic behaviour and damage characteristics; and the high-temperature storage stability. The results demonstrated: significant changes in the molecular structure of irradiated polyolefins, as well as the presence of contaminants in one of the recycled plastics; improvements in the rheological behaviour of the modified binders, showing better results performed after irradiated-polyolefins addition, with gains in rutting and fatigue cracking resistance; high separation tendency of polyolefins from asphalt binder matrix was observed, even for  $\gamma$ -ray exposure polymers.

## ARTICLE HISTORY

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## KEYWORDS

Recycled plastics; rheology; functionalisation; gamma-ray; polymers



## 1. Introduction

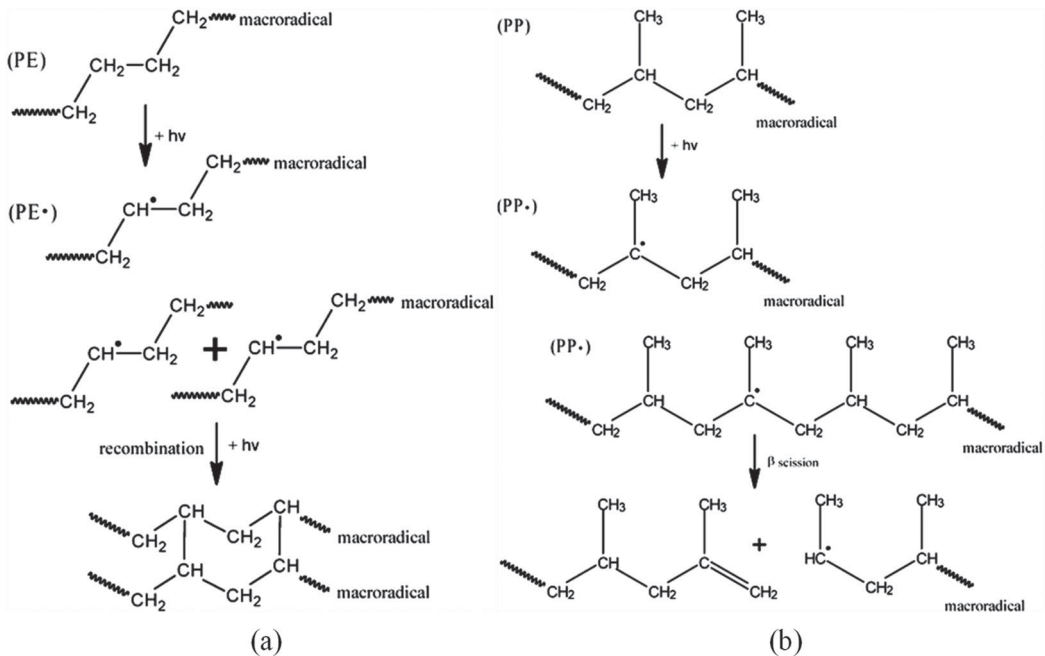
To minimise the exploitation of natural resources, the emission of polluting gases, and the costs associated with conservation, maintenance, restoration or even the construction of road pavement structures, it is now important to consider secondary and marginal materials in the infrastructure sector (Abukhettala, 2021). Previous research evaluated how different recyclable polyolefins, such as polyethylene (PE) and polypropylene (PP), can contribute to performance gain through asphalt binder

modification (wet process), promoting improvements in its rheological behaviour (Kakar et al., 2021; Mandal et al., 2015; Wahhab et al., 2017; Wang et al., 2023; Yeole et al., 2017).

However, the wet method of polymeric materials incorporation can result in unsatisfactory conditions, such as lack of homogeneity; lack of short- and long-term storage stability (poor compatibility between modifier and binder matrix); binder embrittlement and relaxation reduction (Otuoze et al., 2015; Willis et al., 2020; Wu & Montalvo, 2021). Thus, as an alternative to enhance the properties of the polymer-modified binder, physicochemical surface treatment approaches can be used, such as swelling, grafting, polymer coatings, and gamma radiation modification, generally called functionalisation technics (Kazemi & Fini, 2022).

### 1.1. Polyolefins and $\gamma$ -ray irradiation effects

Polymer functionalisation is defined as adding specific chemical groups into polymer molecules to conceive particular physical, chemical, and other properties, by changing the surface chemistry of the polyolefin (Horie et al., 2004). During PE  $\gamma$ -ray irradiation, its hardness and breaking strength increase with higher radiation doses, alongside changes in its other mechanical properties (Sabet & Soleimani, 2017; Suarez & De Biasi, 2003). These changes are primarily linked to radiation-induced crosslinking of the polyolefin, which can also lead to oxidative degradation depending on the environment (Sirin et al., 2022). This interaction of PE with the  $\gamma$ -ray irradiation process is illustrated in Figure 1(a), where the irradiation ( $+h\nu$ ) breaks C–H bonds, generating macroradicals along the polymer chain, which can recombine, forming cross-linked structures, possibly impacting PE mechanical properties. When PP interacts with  $\gamma$ -ray, under atmospheric conditions the polymer undergoes degradation through the  $\beta$ -scission mechanism, where the radical site destabilises the polymer chain, causing cleavage at the  $\beta$ -position, which leads to the formation of shorter polymer fragments, unsaturated bonds (double bonds) and additional radicals, process illustrated in Figure 1(b).



**Figure 1.** Mechanisms of exposure to  $\gamma$ -irradiation of (a) PE and (b) PP under atmospheric conditions. Source: Adapted from Sirin et al. (2022).

## 1.2. $\gamma$ -Ray functionalised polyolefins and their use in pavement materials

Studies investigated different functionalisation techniques and materials to enhance waste plastic properties and compatibility with asphalt mixture components. The results indicated improvements in high (rutting), intermediate (fatigue), and low (thermal cracking) temperature performance, compatibility, dispersion (homogeneity), aging resistance, moisture susceptibility, and storage stability of binder modified with treated polymers, such as styrene–butadiene–styrene (SBS), crumb-rubber (CB) and PE (Aldagari et al., 2022; Ibrahim et al., 2015; Schaefer et al., 2018; Usman et al., 2021). These improvements include increased strength, enhanced durability, and a higher viscoelastic limit temperature, ultimately leading to the development of more durable roadways with reduced maintenance requirements. The long-term benefits of increased durability and decreased maintenance frequency have the potential to offset the initial investment costs. Furthermore, the scalability of the irradiation technique makes it a promising candidate for large-scale economic viability. From an environmental sustainability perspective, this approach offers several advantages, such as the reduction of plastic waste, a lower carbon footprint, improved energy efficiency, and minimised chemical residues (resulting in fewer toxic chemicals within the binder).

The appropriate functionalisation of waste plastics can make them easier to incorporate in asphalt binders and provide a solution to improve their durability, which responses and physicochemical interactions are dependent on both the binder source and the type and amount of plastic (Kazemi & Fini, 2022). Although distinct functionalisation approaches have been evaluated, the current research will focus on gamma-ray ( $\gamma$ -ray) irradiation, which demonstrated advantageous outcomes when applied to building construction and infrastructure materials (Ibrahim et al., 2015; Khan et al., 2021; Schaefer et al., 2018; Usman et al., 2020, 2021), in addition to being efficient in terms of modification and rapid, when compared to other surface treatment approaches. Studies that investigated the effects of  $\gamma$ -ray irradiated polymers in asphalt binders are presented in Table 1, with the main findings regarding the effects of  $\gamma$ -ray irradiation, as well as the dose level applied, type, and content of polymer by weight (wt%) of binder, and blending process methods.

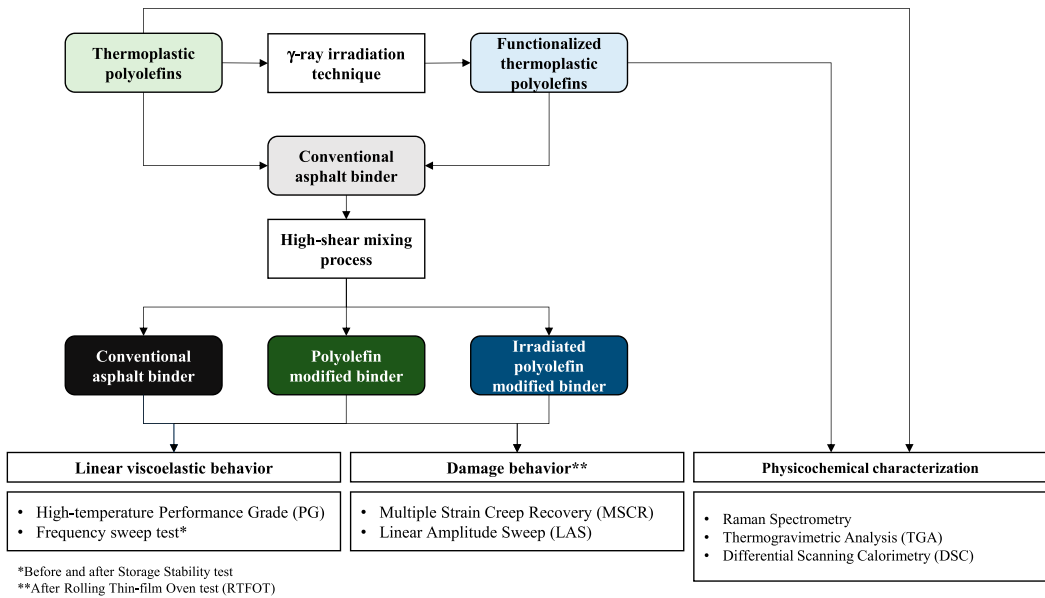
Polymers, such as CR, SBS, polyethylene terephthalate (PET)- and PE-based waste plastics, were used as modifiers. The researchers reported several gains in high- and low-temperature performance; storage stability, when combined with certain additives; anti-aging; and other properties after the wet incorporation method. Polymer structural modification after irradiation, accompanied by molecular crosslinking, radiation-induced polymerisation (grafting), or chain scission reactions, changes their mechanical properties, crystallinity characteristics and thermal transitions (Cota et al., 2007; Reyes et al., 2001; Sirin et al., 2022), which can be reflected in the final binder behaviour. Some drawbacks after the  $\gamma$ -ray functionalisation have been reported (Ahmedzade et al., 2014, 2017; Günay et al., 2022). Studies focusing on low-temperature performance noted that, although creep stiffness ( $S$ ) remained within Superpave limits (less than 300 MPa) for most modified binders, the decrease in the  $m$ -value – defined as the slope of the creep stiffness versus time curve – indicated a reduction in stress relaxation ability, thereby lowering the low-temperature performance. Some authors recommend using up to 3% of irradiated PE, for example, to avoid thermal cracking (Ahmedzade et al., 2017). Thus, the findings suggest that  $\gamma$ -ray irradiation has proven beneficial for increasing the amount of plastic in modified asphalt binders, with improvements in high-temperature performance. However, it has a slightly adverse effect on low-temperature performance grade (PG), making highly modified binders unsuitable for very cold climates due to an increased risk of thermal cracking.

## 2. Rationale and objective

A variety of waste polymers currently generated by industries and post-consumers shows the need for further investigation on how such addition affects the final behaviour of asphalt binders. A recent survey reported that recycled PE is the most used as asphalt binder modifiers and asphalt mixture

**Table 1.** Summary of studies that investigated polymer  $\gamma$ -ray irradiated effects on asphalt binders.

Reference	Dose level (KGy)	Polymer	Content (%)	Sample preparation	Main effects after functionalisation
Fu et al. (2007)	Not informed	Linear-like SBS containing 30 wt% of styrene (methacrylic acid grafted)	3, 4, 5, 6 and 8 wt% of binder	High-speed stirring for 40 min and low-speed stirring for 30 min @ 160 °C	Improvement in high-temperature performance Reduce temperature susceptibility Higher homogeneity, compatibility and storage stability
Ahmedzade et al. (2013)	20.0	LDPE composed of 65–70% of LDPE, 12–17% of LLDPE, and 12–15% of EVA <sup>3</sup> copolymer	1, 3, 5, 7 and 9 wt% of binder	Binder: 90 min @ 163 °C and 500 rpm Binder plus LDPE: added in portions in 15 min intervals for 150 min @ 163 °C and 1300 rpm	Reduces short-term aging Improvement in high-temperature performance Loss of low-temperature performance Optimum irradiated LDPE content of 5 wt% of binder
Ahmedzade et al. (2014)	20.0	HDPE	1, 3, 5, 7 and 9 wt% of binder	15 min @ 168 °C and 500 rpm 45 min @ 168 °C and 1300 rpm A rest period of 60 min @ 168 °C	Reduces short-term aging Improvement in high-temperature performance Loss of low-temperature performance
Zhu et al. (2014)	10, 20, 30, 40 and 50	Linear-like SBS containing 40 wt% of styrene	5 wt% of the binder	Not informed	Reticulation structures of aged SBS restored largely
Ibrahim et al. (2015)	100, 200 and 300	CR	5 and 10 wt% of binder	60 min @ 170 °C and 7000 rpm	Improvement in stability, resistance to aging and low-temperature ductility
Wang et al. (2018)	20.0	LDPE	1, 3, 5, 7 and 9% by volume of binder	Binder: 90 min @ 163 °C and 500 rpm Binder plus LDPE: added in portions in 15 min intervals for 150 min @ 163 °C and 1300 rpm	Improvement in high-temperature performance
Günay et al. (2022)	10	PP (maleic anhydride grafted)	1, 3, 5, 7 and 9 wt%	Added in portions of 15 min intervals @ 3200 rpm for 45 min	Improvement in high-temperature performance and fatigue resistance Slightly adverse effect on low-temperature performance Higher homogeneity, compatibility and storage stability



**Figure 2.** Experimental plan flowchart.

additives (Zhao et al., 2020). Moreover, PE and PP are the most consumed resins by the plastic transformation and recycling national and global industries for the production of new materials, reaching a consumption rate of 36.8% and 19.7% (Abiplast, 2023), respectively. However, in 2020, only 27.0% and 20.2% of the post-consumer PE and PP were properly recycled (Abiplast, 2021). Therefore, to prevent this waste from being released into the environment and to encourage the use of plastic waste, as an alternative material, for the production of modified asphalt binders, using innovative techniques, this study aimed to investigate the effects of  $\gamma$ -ray irradiation on different recycled polyolefins and its impact on the rheological properties of binders modified with post-consumer PE and PP, both with and without  $\gamma$ -ray irradiation treatment. Figure 2 presents the experimental flowchart of the research.

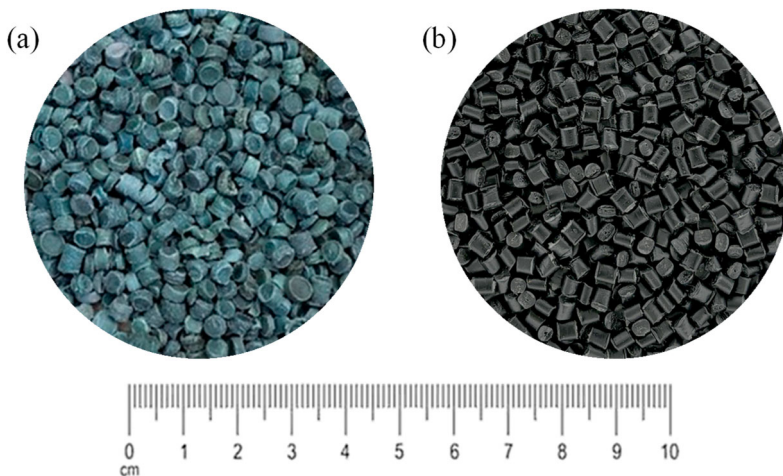
### 3. Materials and methods

#### 3.1. Asphalt binder and recycled polyolefins

A 50/70 penetration grade binder, whose characterisation results are presented in Table 2, was used as the base binder for the blending process. The post-consumer recycled PE and PP pellets, exhibited in Figure 3, were used as modifiers, provided by a local recycling industry, with a content of 3.0 wt% of the total binder. The percentage of polyolefins added to the binder can vary between 1% and 10%

**Table 2.** Neat binder 50/70 penetration grade characteristics.

Characteristics	Standard	Result	Limit
Penetration, 0.1 mm	ASTM D5 (2013)	55	50–70
Softening point, °C	ASTM D36 (2010)	50	> 46
Ductility, cm	ASTM D113 (2018)	> 150	> 60
Specific gravity	ASTM D70 (2021)	1.004	–
Viscosity, cp	ASTM D4402 (2012)		
@ 135°C		315	> 274
@ 150°C		159	> 112
@ 177°C		63	57–285



**Figure 3.** Pellets of post-consumer recycled (a) PE and (b) PP.

by weight of asphalt binder, the most common being between 3% and 5% (Brasileiro et al., 2019). The producers did not provide information regarding the polyolefins tested, PE and PP. The asphalt materials were denoted as 'B' for the neat binder, 'PE3' and 'PP3' for the non-irradiated, and 'PE3\_i' and 'PP3\_i' for the irradiated plastic-modified binders.

### 3.2. Functionalisation and Raman spectrum measurement

The polyolefins pellets were irradiated with a  $^{60}\text{Co}$  source at a dose level of 150 KGy (19 h at 7.84 KGy/h). Most polymers generally require  $\gamma$ -ray radiation doses ranging from 15 to 200 KGy (Naikwadi et al., 2022). Raman measurements were made using a HORIBA LabRAM HR Evolution system, using a 785 nm laser source with 25 mW power (25% of the full available power), and a long-distance 50x objective lens. To obtain a good signal-to-noise ratio, measurements were made with 6 s integration time and 15 accumulations.

### 3.3. Thermal analysis techniques

The recycled non-irradiated and irradiated polyolefins were submitted to both Thermogravimetry (TG) analysis and Differential Scanning Calorimetry (DSC). The first one is a thermal analysis technique in which the change in sample mass (loss or gain) is determined as a function of temperature and/or time, while the sample is subjected to a controlled temperature schedule (Canevarolo, 2004). The DSC measures the energy involved in thermal events (endothermic and exothermic reactions), in which the temperature difference between the substance and the reference material is measured as a function of temperature, while both are subjected to a controlled temperature schedule. One pair of samples per material, of 15 mg each, was tested at an initial temperature of 25°C, with an increment of 10°C/min, up to 1000°C. The DSC chamber was purged with nitrogen gas ( $\text{N}_2$ ) at a 20 ml/min rate.

### 3.4. Blending binder process

The grinding and blending process was performed according to preliminary studies (Kakar et al., 2021; Tušar et al., 2022), using a high-shear mixer (Silverson Laboratory, model L5M-A) at high temperature for a limited period, avoiding excessive plastic degradation and achieving a homogeneous distribution of the polyolefin in the asphaltic phase. To reduce the particle size and facilitate the blending with the binder, the recycled plastics were shredded in cold water, for 10 min at a speed of 5000 rpm, and then

wet sieved on a 230 mesh (or 0.063 mm), dried in an oven at 110°C. Once the binder blend temperature was reached, shredded particles were added and blended for 60 min at 180 °C and a 3500 rpm high shear speed.

### 3.5. Temperature sweep test

To define both continuous grading ( $T_c$ ) and maximum pavement design temperatures, or the high-temperature Performance Grade (PG), temperature sweep tests were conducted, based on ASTM D8239 (2021), using the dynamic shear rheometer (DSR), model Discovery HR-3, from TA Instruments. During the temperature sweep test, the dynamic shear modulus and the phase angle were determined by applying an oscillatory load, with a shear strain of 12.0%, angular frequency of 10 rad/s, using a 25 mm diameter parallel plate geometry, and a gap of 1 mm.

### 3.6. Frequency sweep test

The frequency sweep tests of unaged binders were conducted at a controlled strain of 0.1% at different temperatures, varying from 5 to 80°C, for a frequency range of 0.1–10 Hz, using the DSR. For temperatures below 40°C, the parallel plate geometry with an 8 mm diameter and 2 mm gap was used. At temperatures above 40°C, the parallel plate geometry with 25 mm in diameter and a gap of 1 mm was used. To obtain the master curves, a law describing the translation functions based on the time-temperature superposition principle, determined by Williams et al. (1955), was applied to capture changes in behaviour as a function of temperature and frequency. Based on the linear viscoelastic response of the unaged binders, rheological parameters were also considered to evaluate the fatigue cracking resistance of both modified and reference binders.

#### 3.6.1. Glover-Rowe (GR) parameter

The Glover-Rowe (GR) parameter was developed as an alternative method based on the rheological characterisation of binders to assess the long-term performance of asphalt pavements (Glover et al., 2005; Ruan et al., 2003). The parameter is currently calculated based on the rheological results of  $|G^*|$  and  $\delta$ , according to equation (1), obtained at a frequency of 0.005 rad/s, or 0.0008 Hz, and a temperature of 15°C.

$$GR = \frac{|G^*| \cdot (\cos\delta)^2}{\sin\delta} \quad (1)$$

The higher the degree of aging exhibited by the material, the higher the parameter value will be (Hao et al., 2017). Together with Black space diagrams, the trends and differences in the rheological behaviour of the modified binders, before and after irradiation treatment, were evaluated.

#### 3.6.2. Christensen-Anderson (CA) model

An alternative for characterising the mechanical behaviour of asphalt binder can be conducted by applying the semi-empirical Christensen-Anderson (CA) model. An important aspect of the model is related to its parameters, which have clear physical meanings regarding the nature of the master curves and the relaxation spectrum of asphalt binders (Christensen et al., 2017; Cicalon et al., 2019; Faxina & Klinsky, 2017; Fengler, 2018; Hao et al., 2017; Roja et al., 2020). The higher the crossover frequency ( $\omega_c$ ) value, the lower the material's stiffness. The rheological index ( $R$ ), also known as the  $R$ -value, acts as a measure of the temperature susceptibility or shear effect on the asphalt binder; for example, higher  $R$  values indicate lower thermal susceptibility.

In summary, obtaining these parameters allows for a better understanding of the rheological changes in asphalt binders. At a given reference temperature, the rheological index  $R$  increases, and both parameters crossover modulus ( $G_c$ ) and  $\omega_c$  decrease with aging. Additionally, it provides the advantage of determining the intrinsic physical characteristics of materials without the need for conventional tests, which require larger volumes of binder, such as the penetration test.

### 3.7. Multiple strain creep and recovery (MSCR)

For the determination of percent recovery ( $R\%$ ) and non-recoverable creep compliance ( $J_{nr}$ ) of neat and modified binders, samples were loaded at constant stress for 1 s, with a 9 s rest period duration. Twenty creep and recovery cycles were conducted at 0.1 kPa creep stress followed by ten creep and recovery cycles at 3.2 kPa, following ASTM D7405-20 (ASTM D7405, 2020). After short-term aging on the rolling thin-film test (RTFOT), samples were tested using a parallel-plate geometry with a 25 mm diameter and 1 mm gap at 70°C.

### 3.8. Linear amplitude sweep (LAS)

To evaluate the capacity of the neat and modified asphalt binders to resist cracking damage, the LAS test was performed after RTFOT, according to AASHTO TP 101-14 (2016), in which the material is submitted to cyclic shear loading at cumulative strain amplitudes, to enhance and speed the damage evolution. The asphalt binders were submitted to shear using a frequency sweep to determine their linear viscoelastic properties (no damage), and then the materials were tested with oscillatory load cycles at linearly increasing strain amplitudes up to 30% and a constant frequency (damage). The parallel-plate geometry with an 8 mm diameter and 2 mm gap was used, with a frequency of 10 Hz and temperature of 19°C. The viscoelastic continuum damage (VECD) approach was used to calculate both fatigue model parameters  $A$  and  $B$ , damage tolerance parameter ( $\alpha_f$ ) and the fatigue resistance ( $N_f$ ). The parameter  $A$  represents the material's resistance to cumulative damage,  $B$  illustrates its sensitivity to variations in applied shear stresses,  $D_f$  is the damage accumulation in the specimen at failure and  $N_f$  is the number of cycles to failure.

### 3.9. Storage stability test

A test was conducted to determine the polymer's tendency to separate from PMA (Standard Practice for Determining the Separation Tendency of Polymer from Polymer-Modified Asphalt, 2020), aiming to evaluate the high-temperature storage stability of the recycled polyolefins and binder matrix. For this, 50.0 g of each modified binder was submitted to a conditioned temperature of 163°C into a vertically held aluminium tube, for 48 h. Then the same tube was submitted to a  $-10^\circ\text{C}$  conditioning, in the same position, for at least 4 h. After this time, the sample was equally cut into three parts, and both top and bottom portions were submitted to frequency sweep tests for comparison.

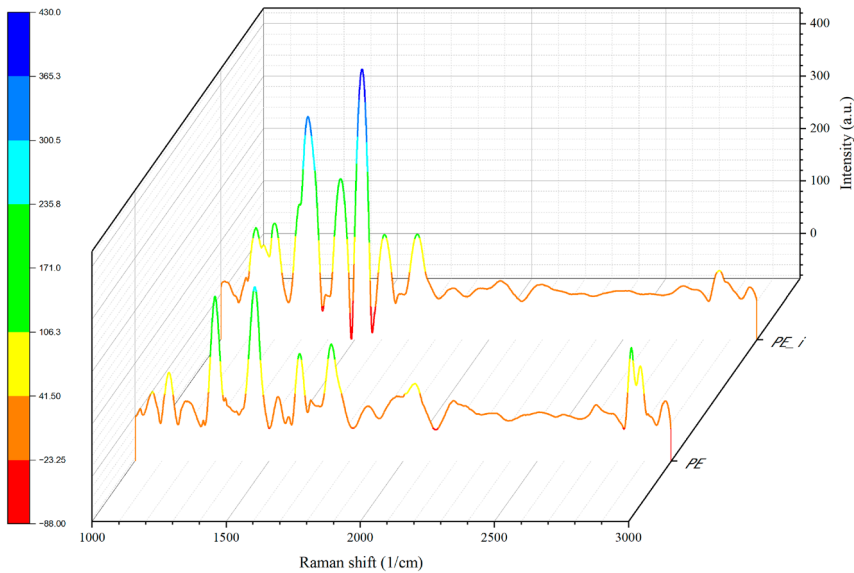
## 4. Results and discussions

### 4.1. Recycled polyolefin characterisation

#### 4.1.1. Raman measurements and interpretation

Homogeneity was verified of each tested sample by taking five different measurements on its surface. Since we obtained similar results, 20 different particles were measured, and an average was taken for the analysis. As seen in Figure 4, the PE is not pure, but a mixture of PE and PET, which is indicated by the aromatic ring vibrations (around  $1615\text{ cm}^{-1}$ ) and the carbonyl ( $\text{C}=\text{O}$ ) stretching vibration in PET (near  $1729\text{ cm}^{-1}$ ), and these peaks serve as markers for distinguishing molecular characteristics, such as crystallinity levels (Puchowicz & Cieslak, 2021). The averages of the irradiated and non-irradiated spectra were compared and are shown in Figure 4.

The changes were mostly related to the peak ratio before and after irradiation. Most of the peaks got proportionally smaller after irradiation, such as  $1064$  and  $1127\text{ cm}^{-1}$ , related to C–C stretching;  $1297\text{ cm}^{-1}$ , which is related to the polymer crystallinity through the C–H twisting, showing that the material became more amorphous with irradiation;  $1439\text{ cm}^{-1}$ , related to antisymmetric deformation;  $1729\text{ cm}^{-1}$ , related to  $\text{C}=\text{O}$  stretching; and both bands at  $2849$  and  $2882\text{ cm}^{-1}$ , related to C–H stretching. The  $1370$  and  $1451\text{ cm}^{-1}$  peaks,  $\text{CH}_3$  antisymmetric deformation, did not suffer any changes, and



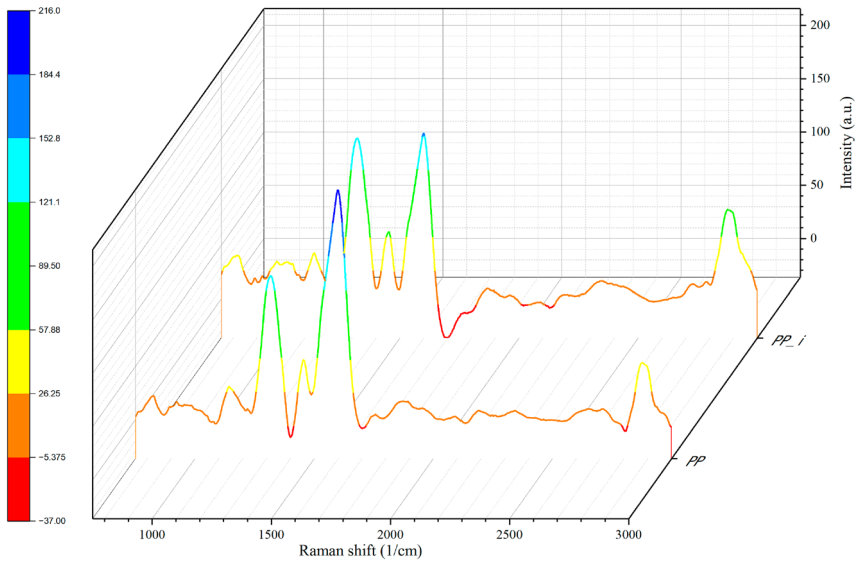
**Figure 4.** Raman spectra comparative of the non-irradiated and irradiated PE.

the  $1526\text{ cm}^{-1}$  band related to the nitrogen contamination increased proportionally. It was observed that the PE crystallinity diminished with irradiation, as shown by the decrease of  $1419$ ,  $1295$ ,  $1130$  and  $1080\text{ cm}^{-1}$  peaks, whereas the  $1450$  and  $1080\text{ cm}^{-1}$  amorphous bands increased or remained the same. The peak at  $1370\text{ cm}^{-1}$ , which presents crystalline and amorphous characteristics, also increased with irradiation (Visentin et al., 2006).

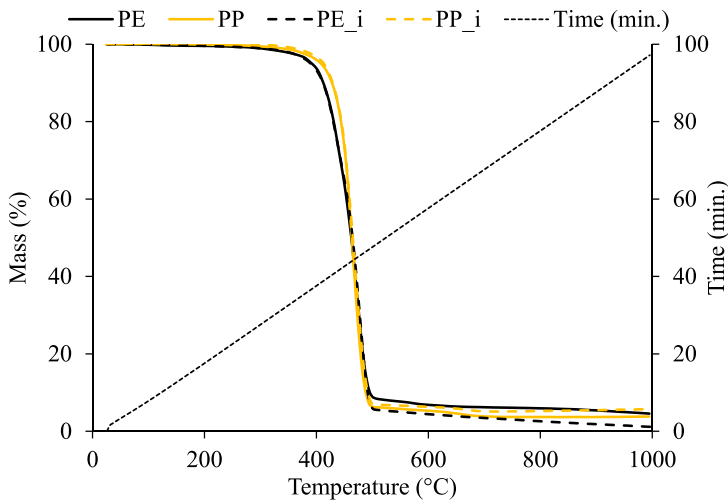
Raman spectra of the non-irradiated and irradiated recycled PP are presented in Figure 5. An increase of the peak at  $2885\text{ cm}^{-1}$  after irradiation is observed, referring to the overlapped band of C–H and O–H (hydroxyl) stretching vibrations (Abiona & Osinkolu, 2010). The peak intensity at  $1153\text{ cm}^{-1}$ , which slightly increased after irradiation treatment, reflects some amorphous phase of PP (Pirker et al., 2021). Other peak increments were detected around  $1329$  and  $1459\text{ cm}^{-1}$  wavelengths, representing  $\text{CH}_2$  twisting and  $\text{CH}_2$  bending, respectively. Both assignments provoke amorphous and crystalline features, leading to distinct mechanical responses since chain scission and cross-linking occur together. After the irradiation, the peaks at  $840$  and  $808\text{ cm}^{-1}$  increased, related to the  $\text{CH}_3$  rocking and C–C stretching, representing crystal structure formations (Furukawa et al., 2006). Physically, the literature points out that PP irradiation leads to more brittle material, lower elongation and elastic capability, and a tensile strength reduction, caused by the oxidative and degradation process (Abiona & Osinkolu, 2010; Pirker et al., 2021).

#### 4.1.2. Thermal analysis

The Thermogravimetric (TG) technique enables the identification of mass changes in materials induced by heating, facilitating the determination of the temperature range in which the decomposition begins, as well as monitoring the reactions' progress, such as dehydration, oxidation, combustion, etc. It is observed in Figure 6 a small degradation (around 0.5% of mass loss) for both polyolefins. Mass loss over 0.5% begins at  $220^\circ\text{C}$  for the recycled PE and recycled PE<sub>i</sub>, while recycled PP and PP<sub>i</sub> presented higher resistance to degradation as a function of increasing temperature, which starts around  $300^\circ\text{C}$ . For both cases, none of the tested materials were compromised by the blending process temperature of  $180^\circ\text{C}$ . PE and PP reached above 5.0% of mass loss at  $390^\circ\text{C}$  and  $410^\circ\text{C}$ , respectively, and more evident percentage differences occur at  $500^\circ\text{C}$ , indicating that PE<sub>i</sub> and PP are more susceptible to thermal degradation above this temperature.

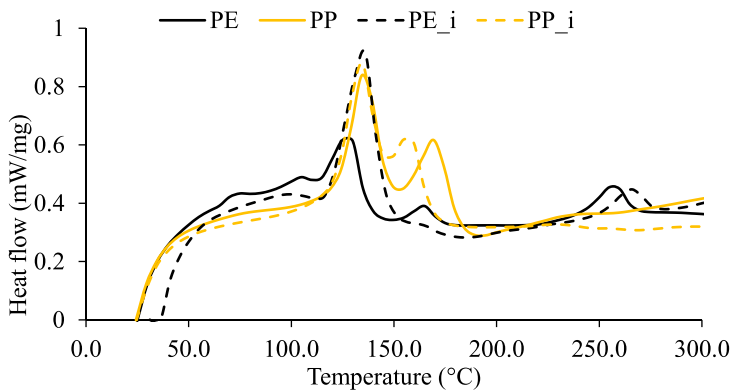


**Figure 5.** Raman spectra comparative of the non-irradiated and irradiated PP.



**Figure 6.** Mass loss of polyolefins before and after irradiation using TG analysis.

Figure 7 shows part of the heat flow curve of both PE obtained in DSC, whose peaks represent the enthalpy variation, or endothermic process (Canevarolo, 2004). This peak serves as an indication of the thermal region in which the material melts, and both recycled plastics present melting temperatures typical of pure polyethylene polymers, around 120 and 147°C (Pruitt, 2011). As expected, peaks at 254.9 and 266.1°C were observed, emphasising the presence of PET indicated by the Raman spectrometry, which melting point is around 265.0°C (Canevarolo, 2010). A peak at 164.9°C for the non-irradiated PE was also observed and may be related to the presence of PP polymer, a contaminant incorporated during the waste recycling process, whose usual melting point is around 165.0°C (Canevarolo, 2010). However, after irradiation, this peak is no longer apparent, a condition that may be related either to material degradation due to  $\gamma$ -ray irradiation or to variation in the recycled material itself, where the composition of the tested particle did not contain traces of PP. To ensure better dispersion and achieve



**Figure 7.** DSC heat flow curves of polyolefins before and after irradiation.

melting of polyolefin particles, it is essential to maintain binder processing temperatures higher than their melting points, which was expected for both plastics (lower than 180°C). However, it is important to note that impurities, such as PET, may cause blending difficulties.

Regarding heat flow intensities, PE<sub>i</sub> requires a higher amount of heat energy to melt, indicated by the heat flow intensity of 0.92 mW/mg, when compared to PE, with 0.61 mW/mg. Minor changes in heat flow intensity were observed comparing PP and PP<sub>i</sub>, as exhibited in Figure 7. The heat flow curve shows that, after irradiation, PP<sub>i</sub> required lower temperatures to reach its melting point for the same amount of heat energy, as evidenced by the peak shifting from 170°C to 160°C. According to Abiona and Osinkolu (2010), this melting temperature reduction can be explained by the changes in the crystal structure of PP, resulting in increased mobility of the polymer molecules as the  $\gamma$ -ray dose increases.

## 4.2. Asphalt binder characterisation

### 4.2.1. Linear viscoelastic behaviour

The dynamic oscillatory rheological tests on asphalt binders allow the characterisation of their linear viscoelastic behaviour, based on the response of the material when submitted to a dynamic shear load, reflected by the resistance to shear deformation. This relationship is expressed by the dynamic shear modulus,  $|G^*|$ , whose elastic and viscous components are designated as storage modulus,  $G'$ , and loss modulus,  $G''$ , respectively, and it is associated with the material stiffness. Since the response between the applied stress and the measured strain will not be immediate, because of the viscous component, this delay is represented by the phase angle, or  $\delta$ , where a value of 90° means a pure viscous behaviour, while 0°, a pure elastic behaviour.

In Figure 8, the upper continuous grading temperature ( $T_C$ ), determined in the linear viscoelastic region, is presented for each binder, based on the temperature sweep tests, and calculated according to ASTM D7643-22. The low temperature was not evaluated in this study. The minimum specification requirement of 1.00 kPa (ASTM D8239-21, 2021), for the permanent deformation resistance rheological parameter,  $|G^*|/\sin\delta$ , was utilised to define the higher PG of each tested binder, B, non-irradiated, and irradiated modified binder, which was, respectively: PG 70-XX, PG 76-XX and PG 82-XX, considering both recycled plastics. Thus, with the 3.0%wt addition of the recycled polyolefin and its irradiation, higher values of  $T_C$  were achieved, indicating less susceptibility of the modified binders for rutting.

Through the frequency sweep test at different temperatures, and the TTS principle, the  $|G^*|$  and  $\delta$  master curves were constructed, as presented in Figure 9. Comparing the  $|G^*|$  master curves, the PP3<sub>i</sub> demonstrated less susceptibility to the load frequency and/or the temperature variation. This material also presented higher stiffness modulus when compared to other binders, conditions that

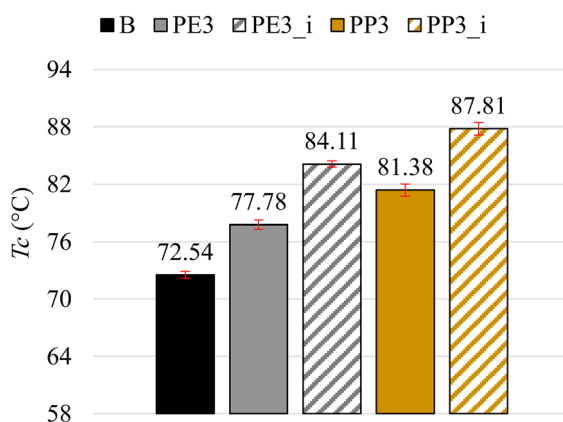


Figure 8. Upper continuous grading temperature ( $T_c$ ).

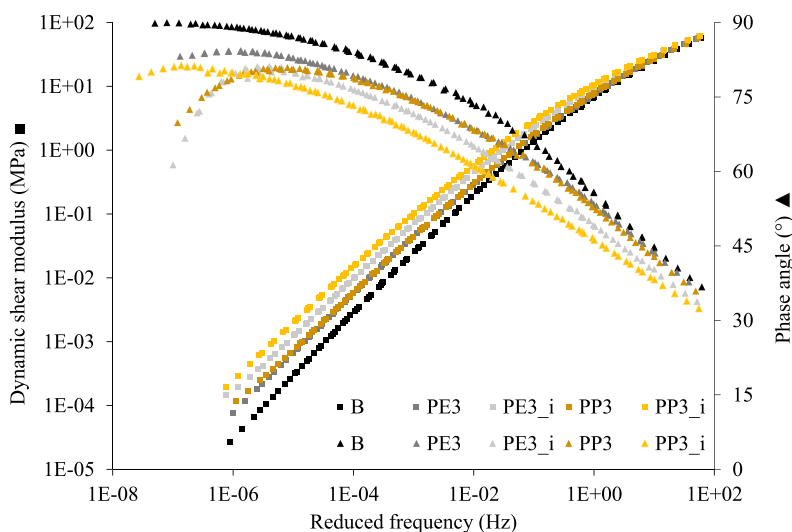
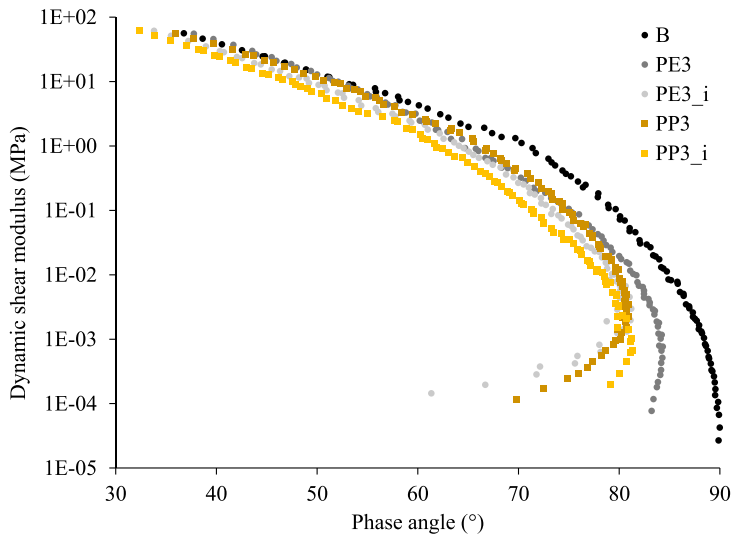


Figure 9. Dynamic shear modulus and phase angle master curves at 15°C.

accent at lower frequencies, or high temperatures. Regarding the material viscoelastic response, PE3\_i and PP3-modified binders demonstrated higher elastic behaviour, represented by the lowest  $\delta$  results in Figure 9, which tends to reduce at higher temperatures, or lower frequency values, when compared to the neat material. At intermediate and higher frequency ranges, stiffness differences tend to reduce among the binders, nevertheless the  $|G^*|$  for the PP3\_i remains higher. In this case, it is important to consider that binders with irradiated PE and PP may be more susceptible to fatigue cracking, due to the higher stiffness, within this frequency range (Rowe, 1993). Since stiffer and more elastic behaviour may influence their cracking resistance (Mensching et al., 2015), such conditions will be discussed further.

The Black space diagram (Figure 10), reinforces the higher elasticity of the modified binders, especially for the PE3\_i and PP\_i, in which the Black space diagram shifts in the high-temperature domain. Other two different trends can be observed when  $|G^*|$  decreases, in which the neat binder B achieved a pure viscous-like behaviour ( $\delta = 90^\circ$ ), typical for unmodified binders, while PE3 and PP3\_i have a slightly  $\delta$  reduction at lower stiffness state and tends to stabilise around 83°. Previous studies investigated the  $\gamma$ -ray irradiation influences on PE physicochemical characteristics, with different dose levels, time duration, and sources of the thermoplastic (Chen et al., 2014; Dipold et al.,



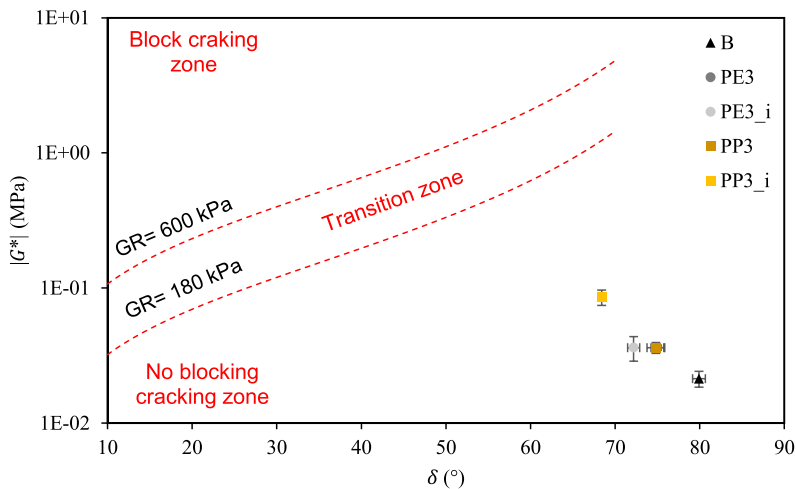
**Figure 10.** Black space diagram.

2022). The results showed that the hardness, melting point, and enthalpy of fusion increase with a higher dose level of up to 800 KGy, depending on the type of PE. The enhancement of crystalline structures leads to improved energy absorption through plastic yield and deformation and also promotes higher stiffness and toughness of the polymer (Dipold et al., 2022; Fan et al., 2009), a condition that may be linked to the change in behaviour of the binder modified with irradiated PE. On the other hand, PP irradiation leads to more brittle material, lower elongation and elastic capability, and a tensile strength reduction, caused by the oxidative and degradation process (Abiona & Osinkolu, 2010; Pirker et al., 2021), which can explain the reduction of PP3 binder elasticity after  $\gamma$ -ray treatment.

Particularly, most of the crystalline peaks presented in Raman spectrometry, which is related to a stiffer characteristic, were reduced after irradiation, yet, the semi-crystalline characteristic enhanced at  $1370\text{ cm}^{-1}$  wavelength, where PE and PET in the solid state have two conformations, planar zigzag, for the chain segments that form the crystalline phase, and ball, for the chain segments that form the amorphous phase (Visentin et al., 2006), which may be responsible for the gain in stiffness and elasticity.

Analyses were also conducted based on the rheological parameters of *GR* and the *CA* model, calculated from the results of frequency sweep tests of unaged binders, at different temperatures, using the DSR. With the acquisition of these parameters, the aim was to better understand the rheological changes in the non-modified and polyolefins modified binders. The *GR* results were plotted in the Black space diagram, as shown in Figure 11. Rowe (2016) proposed two threshold values for the *GR* parameter, resulting from the relationship between asphalt binder ductility and cracking in airport pavements. The value of 180 kPa represents the beginning of the damage zone or transition zone, and 600 kPa marks the onset of block cracking (Rowe, 2016).

The closer to the transition limit of the damage zone, indicated by the *GR* curve at 180 kPa, the higher the degree of material aging (Glover et al., 2005; Rowe et al., 2014). It is noted that all tested binders fall into the region with the least susceptibility to cracking. In comparison to PP3 and PP3\_i, the PE modified binders presented similar *GR* results before and after the irradiation, presenting a slightly gain of elasticity for PE3\_i. However, PP3\_i is positioned closer to the transition zone, which in this zone have a worse balance between stiffness and elasticity, increasing the likelihood of cracking under typical stresses, when compared to PP3.



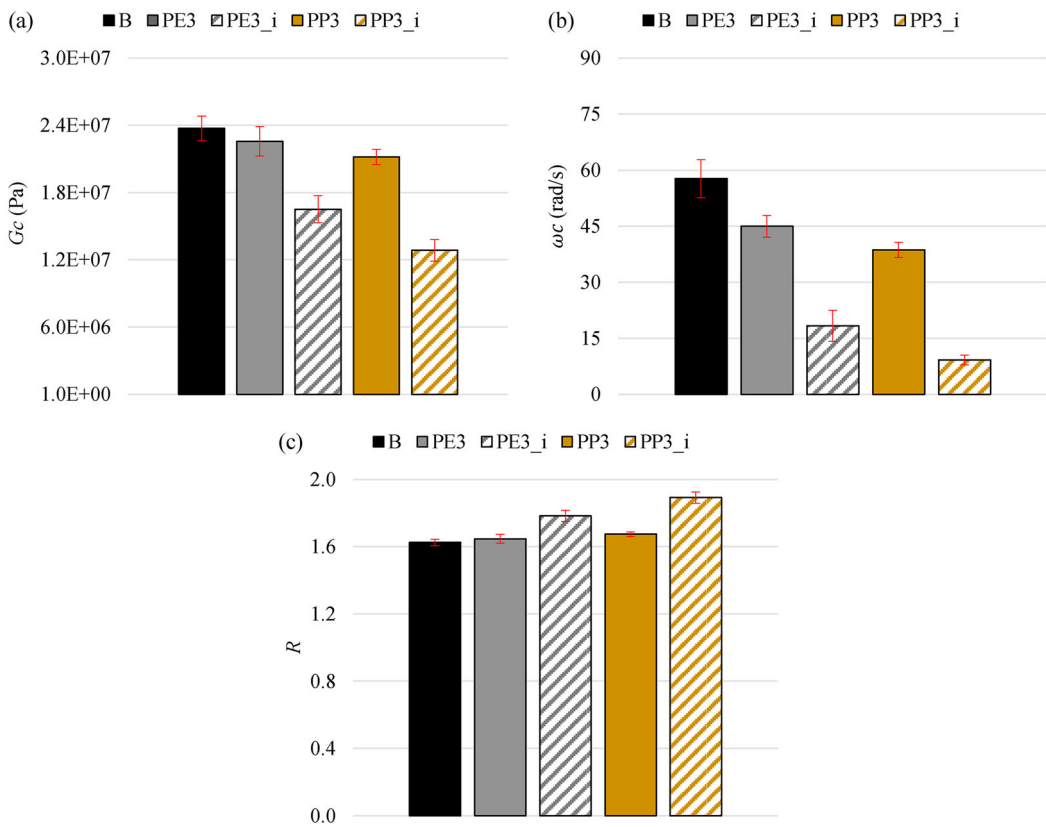
**Figure 11.** Results of the  $GR$  parameter plotted on the Black space diagram.

Figure 12 presented the  $CA$  model parameters obtained. In theory,  $R$ -value increase with aging, while the  $G_c$  and  $\omega_c$  decrease, at the reference temperature of 15°C (Christensen et al., 2017; Cucalon et al., 2019), moreover, this responses are directly associated to the asphalt binders' temperature susceptibility or shear effect (Christensen et al., 2017). The parameter results suggest that the irradiation treated materials provided lower thermal susceptibility and slower relaxation of the modified asphalt binders, indicated by the higher  $R$  values, in accordance to the  $|G^*|$  master curves, which might be more susceptible to fatigue damage at higher strain levels (Christensen et al., 2017; Elwardany et al., 2020).

#### 4.2.2. Damage characterisation

**4.2.2.1. Multiple stress creep and recovery (MSCR).** To evaluate the asphalt binder rutting resistance, both non-recoverable creep compliance ( $J_{nr}$ ) and percent recovery ( $R\%$ ) were determined at 70°C and are presented in Figure 13. Following the linear viscoelastic responses, polyolefin-modified binders presented less susceptibility to permanent deformation when compared to the neat binder, which  $J_{nr}$  values were higher for both 1.0 and 3.2 kPa stress levels. For a more severe shear stress load, the rutting resistance parameter decreased to half for both cases, when compared B with non-irradiated polyolefin, and non-irradiated with irradiated polyolefin. This reduction is directly associated with stiffness enhancement after the polymer incorporation, especially at higher temperatures, which is improved by the  $\gamma$ -ray treatment (Dipold et al., 2022; Fan et al., 2009). Recent studies observed the same trend for different contents and types of PE incorporated into asphalt binders (Chen et al., 2021; Delgado-Jojoa et al., 2018; Nuñez et al., 2014; Tušar et al., 2023; Wang et al., 2018; Zhou et al., 2021). Regarding the binder elasticity, all samples presented enhanced  $R\%$  after modification. The PE3\_i and PP3\_i exhibited the highest percentages, with 69.0% and 24.4% for 1.0 kPa, respectively, but also higher variability, as shown by the error bars, in Figure 13(c). The stress increase in stress, to 3.2 kPa, leads to a significant reduction in the material's elastic response, highlighting its susceptibility to the applied stress.

Figure 14 presents the relationship between  $R\%$  and  $J_{nr}$  at a creep stress of 3.2 kPa, with the elasticity response indicator standard line, defined in AASHTO TP70, representing the threshold between low and high elasticity behaviours. Results above the curve indicate a modified binder with a noticeable elastomeric characteristic, while values under the standard line represent unmodified materials and/or low-elasticity polymer-modified binders. As expected, even with the reduction of the  $J_{nr}$  and a slight recovery increase after  $\gamma$ -ray irradiation, all polyolefin-modified binders fail to achieve the shift to the



**Figure 12.** Results of CA model parameters (a)  $G_c$ , (b)  $\omega_c$  and (c)  $R$ -value.

graph's upper-left corner (higher recovery and lower permanent deformation). The lack of elasticity is related to the nature of thermoplastic elastomers, which are typically polyolefinic homopolymers or saturated polyolefin-based copolymers. The materials exhibit a high degree of crystallinity, increasing their stiffness, but imparting a more 'plastic' rather than 'elastic' character (Polacco et al., 2015).

**4.2.2.2. Linear amplitude sweep (LAS) test.** The cracking resistance of the neat and recycled plastic-modified binders was evaluated according to the LAS test, at 19°C, and the data analysis and interpretation were done using the viscoelastic continuum damage (VECD) approach. In Figure 15(a), effective shear stress–strain curves are presented as indicative of the damage progression and the failure of the materials. PE3\_i and PP3\_i binders presented a higher peak shear stress when compared to both neat and non-irradiated modified polymers, which is related to the materials' high stiffness. Also, the maximum shear stress is reached at a lower shear strain percentage for the PE3\_i, showing a more brittle-type behaviour for the irradiated PE modification, as previously observed in the literature (Ferreto et al., 2012, 2014; Suarez & De Biasi, 2003). A slight increase of PE3 and PP3-modified binder peak shear stress is observed, when compared to the neat binder, so polyolefin crystalline structures lead to improved energy absorption for both plastic-modified binders. Figure 15(b) presents the damage characteristic curves for each evaluated binder, where  $C$  is the normalised material integrity parameter and  $S$  is the damage. All tested binders presented approximated damage curves, with a slight left shift, or damage decrease, for modified binders, lowering for the  $\gamma$ -ray treated polyolefins.

Results of the fatigue model parameters,  $A$  and  $B$ , are presented in Table 3. The  $A$  value represents the material's resistance to cumulative damage, while the  $B$  parameter denotes its sensitivity

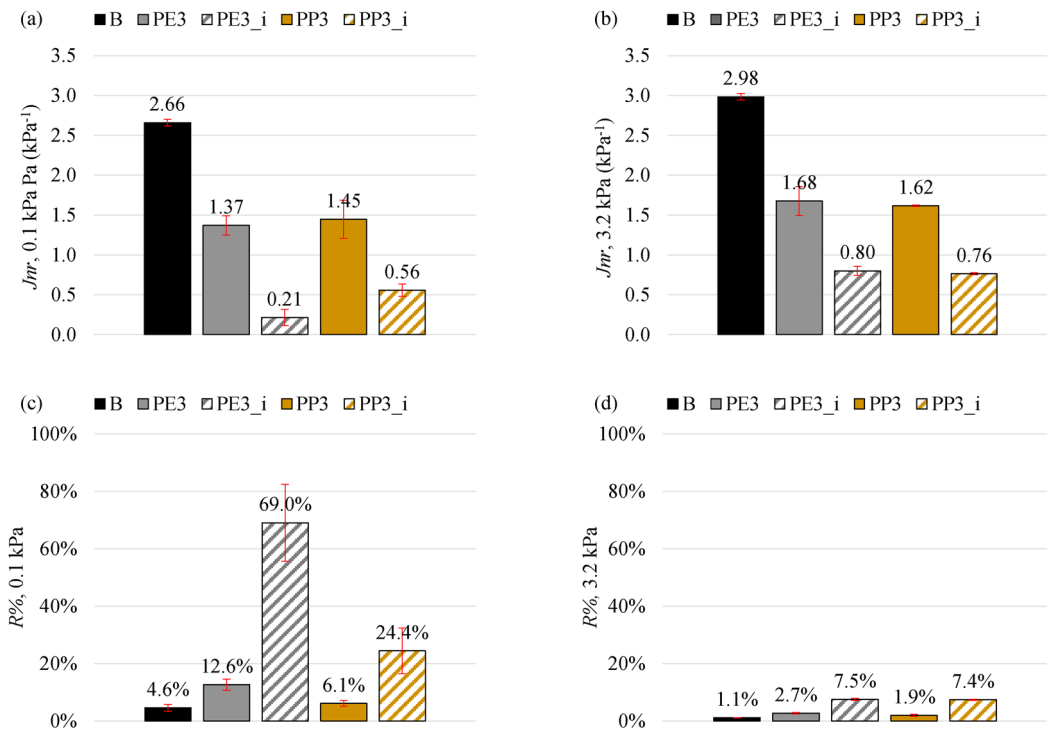


Figure 13. Results of (a)  $J_{nr}$  at 0.1 kPa and (b) 3.2 kPa, and (c)  $R\%$  at 0.1 kPa and (d) 3.2 kPa.

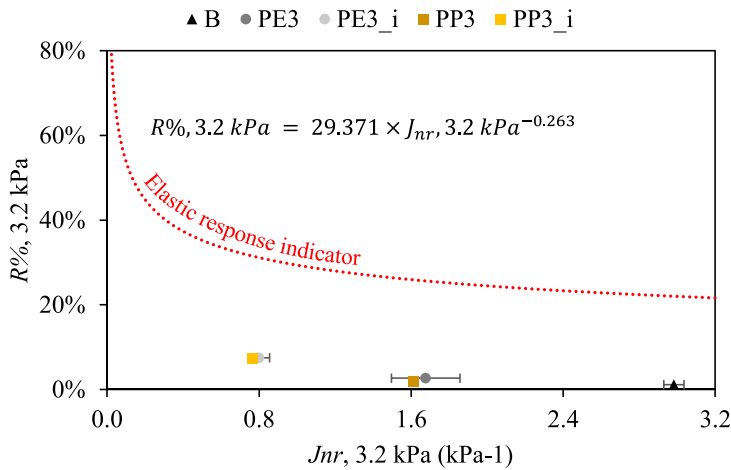


Figure 14. Relationship between  $R\%$  and  $J_{nr}$  at 3.2 kPa creep stress.

to changes in the applied loads (Nuñez et al., 2014). In Figure 16, binder B presented the lowest absolute values for  $A$  and  $B$ , which indicate lower resistance to fatigue damage and less dependence on the load strain (lower slope). However, polyolefin curves (except PP) tend to undergo the base binder at strain levels above 6%, indicating that irradiated modified binders presented lower fatigue resistance under higher load intensities, which can be related to the brittle-type behaviour gained after exposure to  $\gamma$ -ray. Therefore, these circumstances must be carefully considered, when choosing these modified binders for heavy traffic load conditions.

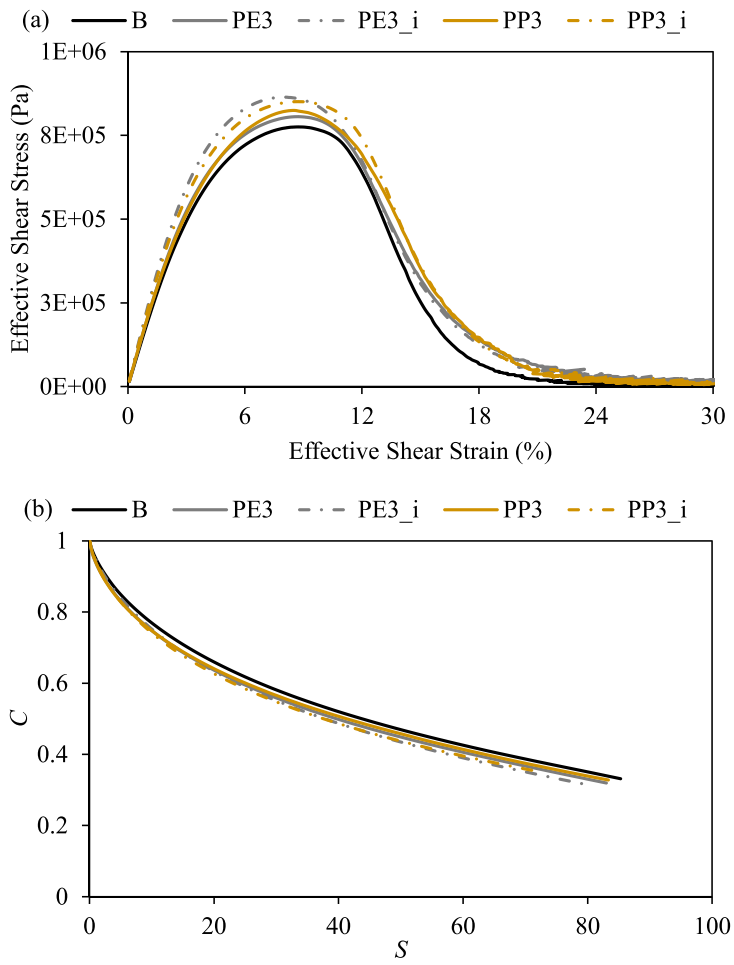


Figure 15. (a) Effective Stress-strain curve and (b) damage characteristic curves.

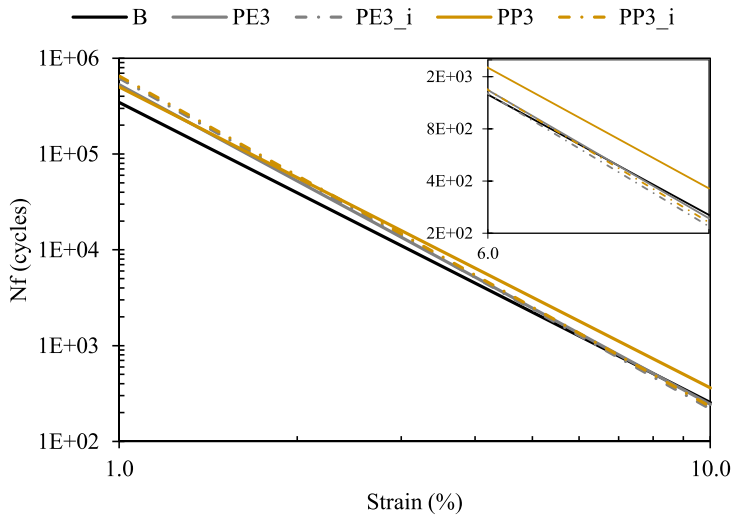
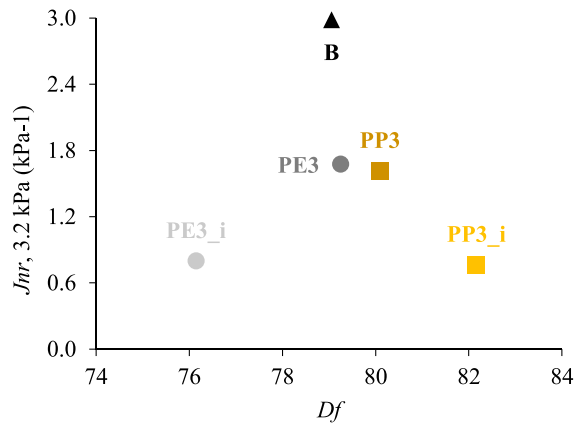


Figure 16. Asphalt binders fatigue resistance curves.

**Table 3.** LAS test parameter results.

Binder	A	B
B	3.76E + 05	-3.12
PE3	5.35E + 05	-3.29
PE3_i	7.21E + 05	-3.50
PP3	5.32E + 05	-3.24
PP3_i	1.06E + 06	-3.59

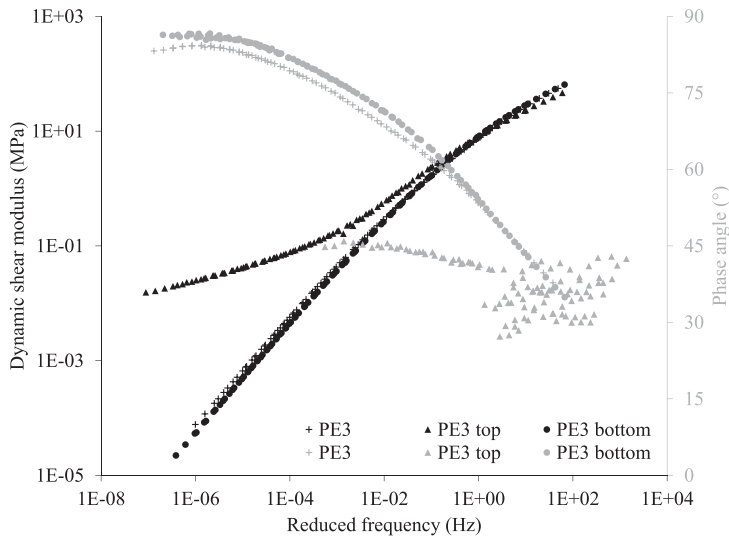
**Figure 17.** Relationship between rutting resistance and damage parameters.

Joohari and Giustozzi (2022) proposed a plot that relates the MSCR and LAS results of several hybrid polymer-modified binders, to rank these materials using a ‘balanced’ design approach. In Figure 17, the correlation between both parameters is presented,  $J_{nr}$  at 3.2 kPa creep stress and the damage accumulation in the specimen at failure ( $D_f$ ), that corresponds to a 35% reduction in undamaged  $|G^*| \cdot \sin \delta$  (AASHTO T391-20, 2020). The binders positioned at the bottom left area in the chart represent the best scenario regarding rutting and fatigue resistance. Therefore, PE3\_i granted the best balance between fatigue and rutting resistance, among the investigated binders. Although PE3/PP3 and B assume similar fatigue responses, based on the  $D_f$  criteria, the non-irradiated modified plastics provided the lowest susceptibility to permanent deformation.

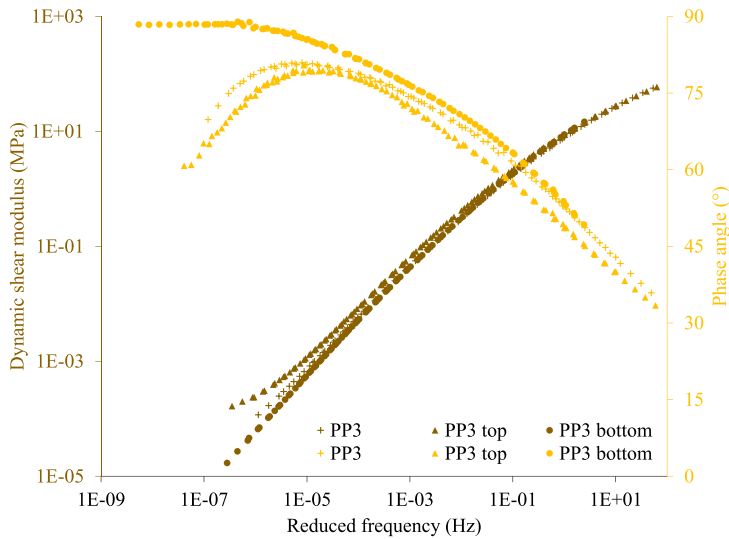
#### 4.2.3. Storage stability and rheological evaluation

To evaluate the polymer separation tendency in the asphalt matrix, the modified binders were conditioned for 48 h at 163°C to simulate a high-temperature storage state, according to ASTM D7173 (2020). After cooling down at -10°C, the aluminium tubes were cut into three equal portions, and the top and bottom parts were tested, based on their rheological behaviour, and compared to the respective homogenised polyolefin-modified binder, as reference. Despite the performance gains after irradiated polyolefins incorporation, no benefits were observed regarding the modified binders’ stability. The dynamic shear modulus and the phase angle master curves, Figures 18–21, exhibit a lack of stability for both investigated conditions, before and after irradiation. It is noted that the data points showed a high degree of dispersion concerning the phase angle. While dynamic shear modulus values are not significantly affected by signal processing (as the stress and strain peaks are relatively insensitive to the experimental data reading rate), the phase angle must be measured precisely between the corresponding stress and strain peaks. This may explain why phase angles are dispersed for the samples with a high particle concentration, represented by the top portion of the tube.

For all tested materials, the modified binder in the bottom section of the tubes showed lower stiffness when compared to the top section, with dynamic shear modulus values approximately 75% lower



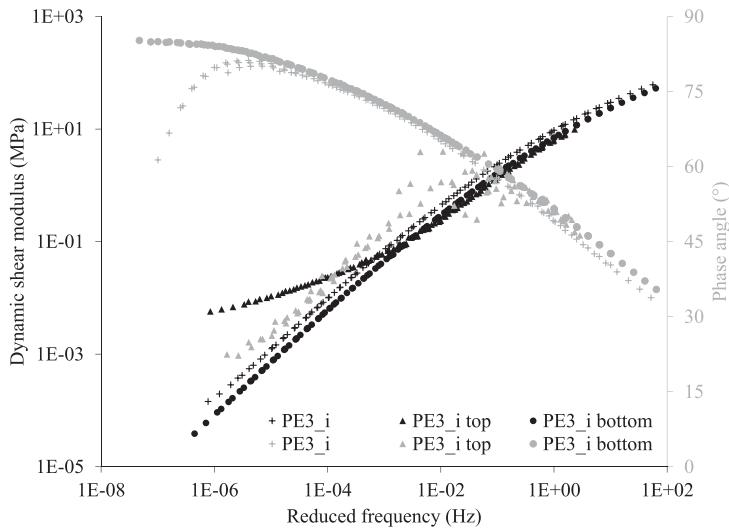
**Figure 18.** Frequency sweep results of reference PE (homogeneous) compared to top and bottom portions after high-temperature storage.



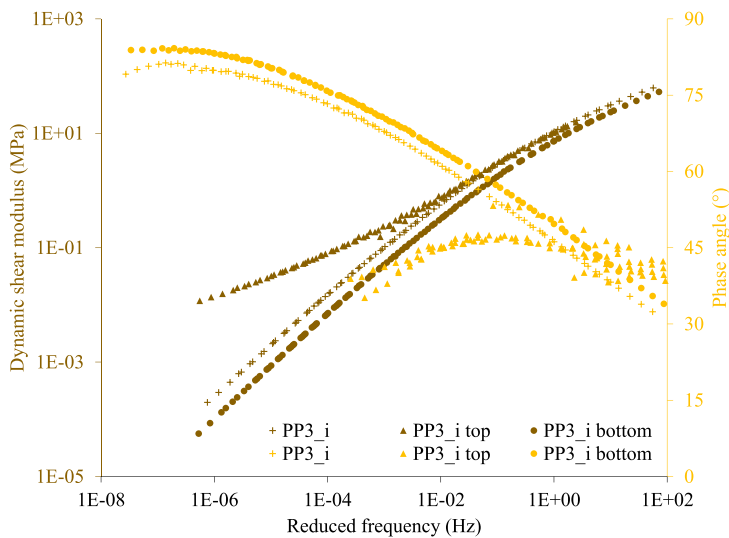
**Figure 19.** Frequency sweep results of reference PP (homogeneous) compared to top and bottom portions after high-temperature storage.

than the reference (homogenous) material, as seen in Figure 22. The polyolefin particles' movement from the bottom to the top section of the storage samples indicates 'weak' compatibility between recycled plastics and the asphalt matrix, becoming more evident at lower frequencies, with dynamic shear modulus differences up to 100%.

The viscoelastic behaviour showed disturbances and abrupt variations in the data collected, representing inconsistencies during the test (Airey, 2002). These viscoelastic deviations can also provide information about possible binders' phase changes, indicating the high presence of polymeric components (King et al., 2012; Rowe et al., 2014). In addition, the increment of the irradiated PP dynamic shear modulus difference might reflect the effects of the oxidation process, which can occur after the

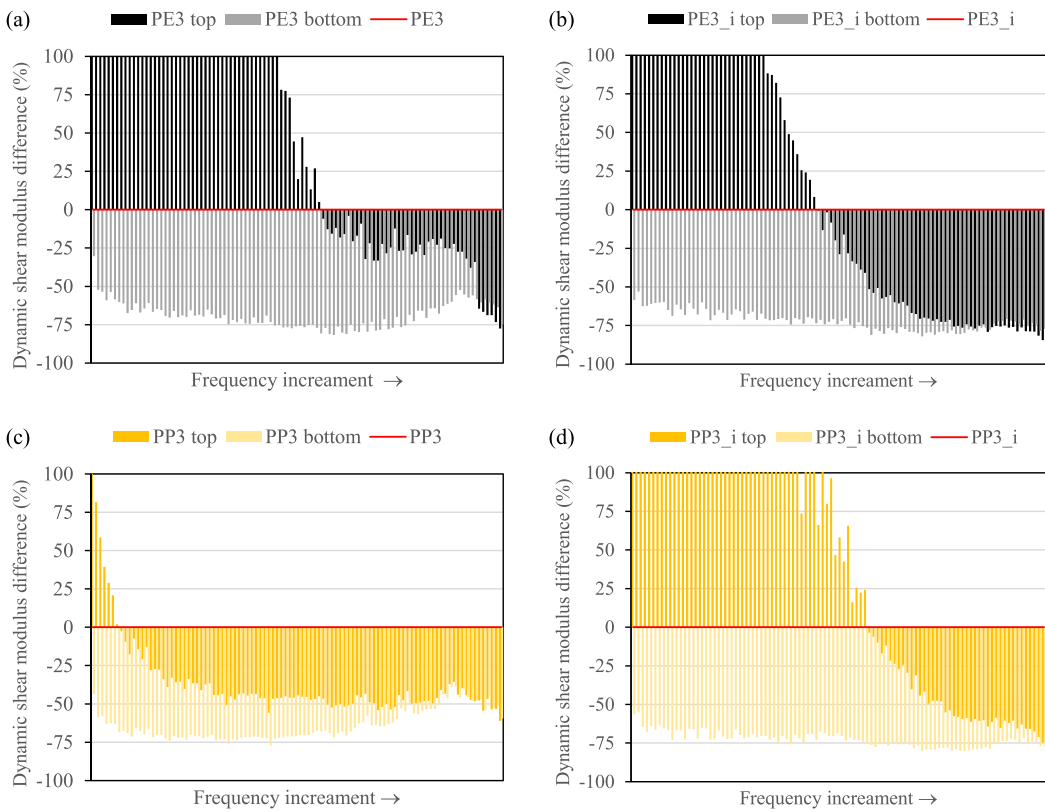


**Figure 20.** Frequency sweep results of reference irradiated PE (homogeneous) compared to the top and bottom portions after high-temperature storage.



**Figure 21.** Frequency sweep results of reference irradiated PP (homogeneous) compared to the top and bottom portions after high-temperature storage.

$\gamma$ -ray treatment. Before the irradiation, as seen in Figure 19, the PP-modified binder top portion presented a phase angle curve similar to the respective homogeneous material. However, in Figure 21, the collected data showed a high degree of dispersion after irradiation. Previous studies investigated the effect of  $\gamma$ -ray irradiation over PP and observed that when exposed to gamma rays under atmospheric conditions, the polymer undergoes degradation through the  $\beta$ -scission mechanism (Sirin et al., 2022). Additionally, the radicals formed on the polypropylene chains react with atmospheric oxygen, leading to oxidative degradation of the polyolefin (Cota et al., 2007; Sirin et al., 2022), and possibly reducing the interactions of the modified binder matrix's constituents. Despite some chemical bonds between



**Figure 22.** Percentage  $|G^*|$  difference of (a) PE, (b) irradiated PE, (c) PP and (d) irradiated PP, from the top and bottom portions, compared to the respective homogeneous polyolefin modified binder.

binder and polyolefin particles may occur (Wang et al., 2018), due to their nonpolar nature and typically high crystallinity, they are nearly completely immiscible with asphalt (Polacco et al., 2015), even after the  $\gamma$ -ray treatment.

## 5. Conclusions

A diversity of waste polymers currently generated by industries and post-consumers shows the importance of further investigation into how such addition affects the final behaviour of asphalt binders. Hence, the present study investigated possible effects on the rheological properties of an asphalt binder modified by two recycled post-consumer polyolefins, PE and PP, submitted and not to the  $\gamma$ -ray irradiation technique. The research conclusions are listed.

- Raman spectrometry and thermal analysis proved to be efficient tools for identifying impurities in recycled polyolefins. The Raman spectra confirmed the presence of PET in recycled PE, while evidence based on the material's melting point revealed the presence of both PET and PP. These findings highlight significant challenges in plastic recycling, particularly concerning cross-contamination. Such contamination underscores the need to enhance sorting processes, invest in more advanced detection technologies, and educate consumers and recyclers on the importance of proper waste segregation.
- The Raman spectra analysis indicated complex chemical changes. The recycled polyolefins became more amorphous, and their crystallinity decreased after  $\gamma$ -ray irradiation, a condition associated

with stiffness loss. However, the semi-crystalline characteristic was enhanced, where PE and PET exhibited both conformations. In the case of PP, the irradiation treatment may have contributed to the observed increase in stiffness and elasticity.

- Thermal analysis indicated that neither of the recycled polyolefins was compromised by the modified binders blending process temperature; they were able to withstand it without undergoing degradation. Regarding heat flow intensities, irradiated PE required a higher amount of heat energy to melt compared to regular recycled PE, while irradiated PP required lower temperatures to reach its melting point for the same amount of heat energy.
- Frequency-temperature sweep tests demonstrated several benefits in the linear viscoelastic responses following the incorporation of polyolefins, including reduced thermal susceptibility, increased stiffness, and enhanced elastic behaviour. Non-irradiated and irradiated polyolefins improved the high PG classification by one and two grades, respectively.
- All binders tested exhibit low cracking susceptibility, with PE modified binders displaying similar *GR* results before and after irradiation. The irradiation treatment leads to lower thermal susceptibility in modified binders, as evidenced by higher *R* values, consistent with the  $|G^*|$  master curves.
- In general, the performance of all tested binders improved in terms of permanent deformation and fatigue resistance under lower strain levels, with careful considerations required when selecting these binders for heavy traffic load conditions. These conclusions align with previous literature studies and highlight the benefits of using recycled polyolefins as asphalt binder modifiers, with additional advantages observed when the materials are submitted to  $\gamma$ -ray irradiation.
- The frequency sweep analysis, performed after the static storage stability test, demonstrated a strong tendency for phase separation when conditioned at a high temperature, even when subjected to  $\gamma$ -ray exposure. This conclusion emphasises the need for further optimisation of the material formulation and/or processing methods to enhance compatibility and prevent phase separation.

The conclusions relate only to the materials, methods, and test conditions employed in this current research. To solve storage stability limitation, further investigations will consider different sources of post-consumer recycled polyolefins and additives. In addition, it is important to consider the effects of irradiation on aging aspects and the low-temperature performance of the modified binders.

## Disclosure statement

No potential conflict of interest was reported by the author(s).

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