



EFFECT OF THE ADDITION OF CHEMICALLY DEGRADED POLY(ETHYLENE TEREPHTHALATE) ON THE RHEOLOGICAL PROPERTIES OF BITUMEN

WPŁYW DODATKU POLI(TEREFTALANU ETYLENU) PODDANEGO CHEMICZNEJ DEGRADACJI NA WŁAŚCIWOŚCI REOLOGICZNE ASFALTU

Grzegorz Mazurek*, Mateusz Kaniewski, Marcin Podsiadło
Kielce University of Technology, Poland

Abstract

The study explores the feasibility of incorporating poly(ethylene terephthalate) (PET) plastomer into processed asphalt through chemical degradation. The depolymerization process involved subjecting the PET plastomer to aminolysis reaction with ethylenediamine. Consequently, the resultant monomer exhibited reduced rigidity and increased machinability. Enhancing its degree of fragmentation facilitated improved homogenization with bitumen. The resulting blend of bitumen and degraded plastomer underwent evaluation for creep resistance in accordance with the Multiple Stress Creep Recovery (MSCR) methodology at a temperature of 64°C. Moreover, fundamental standard tests were conducted, including penetration, softening point, and Fraass breaking point. The incorporation of additional amino groups in the form of degraded PET into the bitumen reduced its susceptibility ($J_{nr3200} < 0.5 \text{ kPa}^{-1}$) to the creep process and lowered the brittle temperature (approximately -3°C) in comparison to 50/70 neat bitumen. Furthermore, the proposed depolymerization technology for PET and its application to bitumen represents a viable approach for the utilization of PET plastomer.

Keywords: plastomer aminolysis, rheology, recycling, Maxwell model

Streszczenie

W pracy przedstawiono możliwość aplikacji plastomeru poli(tereftalan etylenu) PET do asfaltu przetworzonego poprzez zastosowanie chemicznej degradacji. Proces depolimeryzacji polegał na poddaniu plastomeru PET reakcji aminolizy z wykorzystaniem etylenodiaminy. W efekcie uzyskany monomer uzyskał mniejszą sztywność oraz był łatwy w obróbce mechanicznej. Zwiększenie jego stopnia rozdrobnienia umożliwiło lepszą homogenizację z asfaltem. Uzyskaną mieszaninę asfaltu i zdegradowanego plastomeru poddano ocenie odporności na proces pełzania zgodnie z metodyką MSCR w temperaturze 64°C. Ponadto wykonano podstawowe badania normowe takie jak: penetracja, temperatura mięknięcia oraz temperatura Fraassa. Wprowadzenie dodatkowych grup aminowych w postaci zdegradowanego PET do asfaltu zmniejszyło jego podatność ($J_{nr3200} < 0,5 \text{ kPa}^{-1}$) na proces pełzania oraz temperaturę lamliwości (około -3°C) w porównaniu do asfaltu 50/70. Ponadto zaproponowana technologia depolimeryzacji PET i jego implementacja do asfaltu jest sposobem, który można wykorzystać do utylizacji plastomeru PET.

Słowa kluczowe: aminoliza plastomeru, reologia, recykling, badania asfaltu, model Maxwella

1. INTRODUCTION

The bituminous binder constitutes a thermoplastic material that emerges as a byproduct during the distillation process of crude oil [1, 2]. The ultimate composition of bituminous binder is predominantly influenced by the origin of the crude oil. Within the climatic conditions of Poland, post-petroleum bitumen manifests in three rheological states: elastic-brittle, viscoelastic, and viscous. The influence of low temperatures renders bitumen susceptible to low-temperature cracking, whereas in its viscous state, it facilitates the development of plastic deformations within mineral and asphalt mixtures during the summer [3]. In recent decades, the most effective approach to augment the viscoelastic state range of bitumen has been the incorporation of polymers into the bitumen phase [4, 5]. The prevalent and efficacious approach involves the modification of bitumen utilizing styrene-butadiene-styrene copolymer (SBS). The notable compatibility of SBS with bitumen, attributed to its analogous solubility with the maltene phase, facilitates the formation of a stable mixture characterized by a cross-linked structure [6, 7]. Currently, the challenge associated with recycling synthetic materials necessitates the exploration of alternative management strategies. Within this framework, plastomer warrant particular consideration. The alteration of bitumen with plastomer necessitates supplementary measures due to the incomplete understanding of their effective integration into the bitumen matrix [8]. Based on an analysis of the literature, it can be inferred that the direct modification of bitumen using plastomer substantially diminishes the stiffness of the bitumen, thereby positively enhancing the rutting resistance of the asphalt mixture (mma) [9]. Nevertheless, this modification results in the degradation of the low-temperature characteristics of the asphalt mixture. Furthermore, the inclusion of a plastomer may compromise the stability of the bitumen-plastomer composite, leading to decreased ductility in the final product [10]. It is noteworthy that the incomplete compatibility between bitumen and plastomer is not the sole challenge faced by researchers during the homogenization process. An additional issue is the optimization of the mixing process, which is influenced by numerous factors [11]. A considerable disparity in the molecular weight, solubility, softening point, and chemical inertness of the plastomer presents a significant challenge to the effective homogenization with bitumen [12, 13]. Moreover, extended mixing duration leads to alterations in the polarity and stiffness

of the bitumen as a result of aging. Consequently, the technological modification process warrants careful consideration when homogenizing bitumen and plastomer [14, 15].

This study endeavors to explore a novel approach to modifying PET bitumen. Based on the experiments recorded in the paper [14], the modification procedure involved subjecting PET to depolymerization through an aminolysis reaction. The terephthalamide monomer produced through this process exhibited a reduced molecular weight and incorporated amino groups that enhance adhesion and compatibility with bitumen, thereby contributing to the stabilization of the mixture. Consequently, to achieve a stable bitumen and plastomer mixture, it is imperative to establish not only an optimal mixing technique but also a method for the application of a properly selected and pre-processed plastomer. The literature documents various attempts to modify the plastomer of poly(ethylene terephthalate), commonly referred to as PET, utilizing higher-order amines [16, 17]. Nevertheless, this methodology was primarily directed towards optimizing the recycling process of PET [18, 19]. This research primarily emphasizes the enhancement of the rheological properties of 50/70 neat bitumen, thereby rendering it more suitable for incorporation into asphalt mixtures, while concurrently exploring the potential application of the PET plastomer.

2. MATERIALS AND METHODS

2.1. Bitumen

The primary binder employed during the modification process was 50/70 neat bitumen. The bitumen under examination underwent fundamental rheological testing. The findings of the research, accompanied by the 95% confidence interval for the mean, are illustrated in Table 1.

Table 1. Test results of 50/70 neat bitumen

Feature	Result	Standard
Penetration at 25°C, 0.1 mm	61.8 ± 3.4	PN-EN 1426 [20]
Softening point TR&B, °C	49 ± 1	PN-EN 1427 [21]
Fraass breaking point, °C	-15.5 ± 1.2	PN-EN 12593 [22]
Elongation at 5°C, cm (declared)	2.2 ± 0.3	PN EN 14023 [23]
Cohesive energy, J/cm ²	1.7 ± 0.5	PN EN 14023
Viscosity at 60°C, Pas	-16.6 ± 2.0	ASTM D 4402 [24]
Viscosity at 90°C, Pas	-15 ± 1.5	
Viscosity at 135°C, Pas	60	

2.2. Poly(ethylene terephthalate) depolymerization process

The modification of road bitumen was conducted utilizing a PET plastomer (polyethylene terephthalate) depolymerized via aminolysis. Ethylenediamine was employed as the amine for conducting the aminolysis reaction. The PET plastomer in question belongs to the category of thermoplastic materials exhibiting a crystalline structure. According to the supplier's specifications, the PET polymer is characterized by a maximum pour point of $T_p = 256^\circ\text{C}$ and a glass transition point of up to 75°C . The aminolysis procedure was executed in a reactor fabricated by the author. A continuous nitrogen purge was maintained throughout the aminolysis process. The entire procedure was automated, allowing comprehensive regulation of temperature and mixing speed of the constituents. The maximum particle size of the PET flakes was recorded at 1 mm. This measurement system is illustrated in Figure 1.



Fig. 1. Measuring set for aminolysis reactions

Following the aminolysis process, which is documented in a patent application, the terephthalamide compounds denoted as TRA compound was synthesized. Its integration with asphalt was significantly more efficient compared to the utilization of pure PET [15]. The alteration in the structure of PET and its chemical composition is depicted through the spectroscopic spectrum presented in Figure 2.

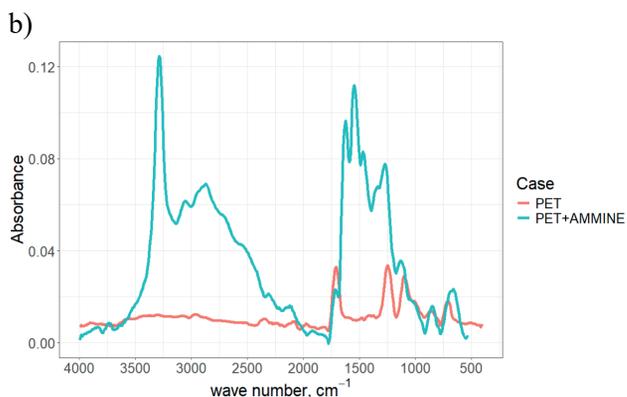


Fig. 2 Polyethylene terephthalate (PET) undergoing the depolymerization process: a) structural composition prior to modification (left) and subsequent to aminolysis (right); b) spectroscopic spectra of unmodified PET and aminolyzed (PET+Ammine) referred to as TRA (terephthalamide)

The initial material manifested a consistency similar to that of soap (Fig. 2a). The shredding process was executed with minimal effort compared to PET, which necessitated substantial energy and a highly efficient mixer for pulverization. Figure 2b illustrates the FT-IR spectrum of the initial PET plastomer and terephthalamide (TRA). It is noteworthy that the distinct spectral peak of the TRA component observed at 1720 cm^{-1} , corresponding to the tensile vibrations of the carbonyl ($-\text{C}=\text{O}$) ester group within PET molecules, has substantially diminished. Nevertheless, it should be highlighted that two additional peaks emerged, specifically at 1636 cm^{-1} and 1547 cm^{-1} , which correspond to the characteristic tensile vibrations of the carbonyl ($-\text{C}=\text{O}$) and bending vibrations of the $\text{N}-\text{H}$ of the amide group within the PET additive. Additionally, a novel characteristic peak at 3320 cm^{-1} has appeared, attributed to the tensile vibrations of $\text{N}-\text{H}$ in the amine group. This was accompanied by a peak for the wave number 2850 cm^{-1} , which can be ascribed to the asymmetric vibrations of the $-\text{CH}_2-$ groups. According to the observations of other researchers in the [25], [26] this result indicated a successful aminolysis of the PET. Moreover, the FTIR analysis presented herein demonstrated the formation of amide bonds between terephthalic residues and amines involved in the aminolysis reaction. This finding correlates with prior studies, which suggest that the depolymerization of PET in the presence of an excess of amines results in partial, secondary polymerization, manifesting as the formation of a copolymer between terephthalic acid and amines [27]. This phenomenon is posited to exert a favorable influence on the rheological properties of the initial bituminous binder matrix.

2.3. Mixing TRA with bitumen

Bitumen was combined with TRA at a constant rotational speed of the homogenizer mixer set at 3000 rpm for a duration of 60 minutes. The mixing procedure was executed at two different temperature levels: 150°C and 180°C. The proportion of TRA additive utilized was 2% based on the weight of the asphalt. Following the 60-minute mixing procedure, the TRA-modified bitumen was allowed to rest for 15 minutes, while maintaining a temperature of 150°C (irrespective of the main process mixing temperature) alongside a rotational speed of 30 rpm. An initial evaluation of the effectiveness of the TRA mixing process with bitumen was conducted utilizing an epifluorescence microscope (Fig. 3).

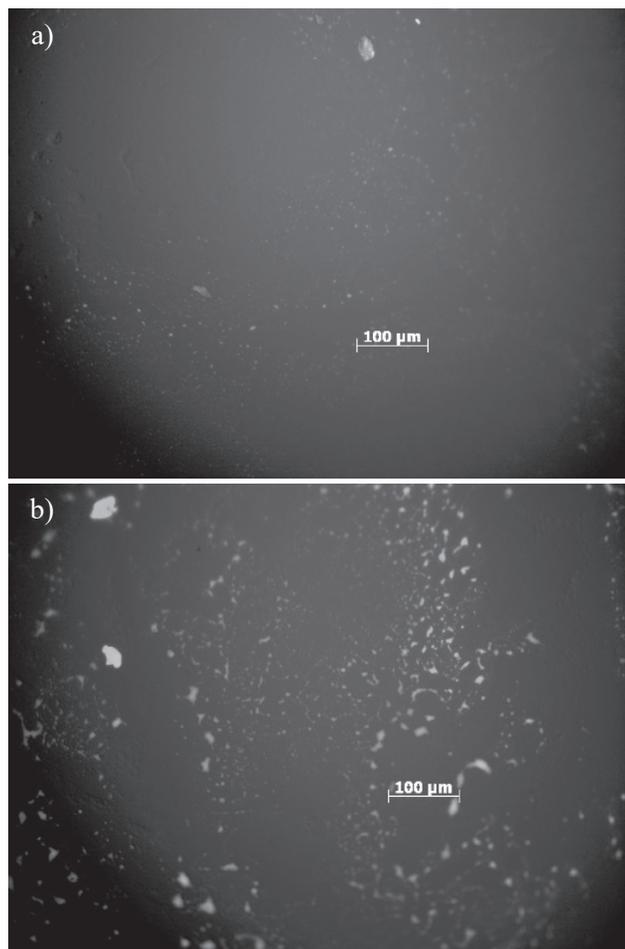


Fig. 3. Dispersion of degraded PET in bitumen phase using an epifluorescence microscope: a) mixing at 150°C; a) mixing at 180°C

Upon analyzing the data depicted in Figure 3, it can be inferred that the mixing temperature significantly influenced the results. When evaluating the TRA dispersion in bitumen across both mixing

temperatures, it is important to note that the level of homogenization achieved was high. Specifically, in the sample illustrated in Figure 3b, partial secondary polymerization of TRA in the bitumen was observed, which is projected to enhance the ductility of the bitumen. Conversely, the sample in Figure 3a exhibited a fine-grained nature of TRA dispersion in the bitumen, highlighting an indisputable advantage of utilizing TRA.

2.4. Rheological Properties Testing

2.4.1. MSCR test

The multi-stress creep recovery (MSCR) test was carried out according to EN 16659 [28] using a dynamic shear rheometer (DSR). It is a measuring system of parallel plates 25 mm in diameter with a gap of 1 mm. The test procedure involves repeatedly applying a load lasting 1 second to an RTFO-aged bitumen sample at constant stress. Each loading is followed by a recovery period of 9 seconds. A single creep-recovery cycle lasting 10 seconds is repeated 10 times at a stress of 0.1 kPa, further increased to 3.2 kPa. The measurement temperature was 50°C, 60°C and 70°C.

2.4.2. Generalised Maxwell model

The tested waste plastomer-modified bitumen underwent a detailed rheological analysis. Its purpose was to describe the strain during creep, using the MSCR test and utilizing the generalised Kelvin-Voigt model (Prony series). The good match of experimental data with this model suggests that the bitumen are within the linear viscoelastic (LVE) limits. The discrepancy in the model's results when compared to the experiment can point to non-linear behaviour τ - γ and provide information about the occurrence of substantial plastic strains. The (shear) relaxation function G is described with the following equation [29, 30] (1):

$$G(\psi^t) = G_0 \left(1 + \sum_{i=1}^n g_i \left[1 - e^{(-\lambda_i \psi^t)} \right] \right) \quad (1)$$

where: G_0 – instantaneous compliance, g_i – i -th compliance corresponding to the next Kelvin-Voigt element, λ_i – i -th retardation time, ψ^t – reduced time $\psi^t = \frac{t}{a_\sigma} = \frac{t}{a_\sigma}$, a_σ – shift time factor equaled to 1.

In general equation (1) the shift factor a_σ depends on temperature using nonlinear formula. For case where only one temperature case is used its value is equaled to 1. In the paper, the mastic samples

were subjected to pure shearing as part of controlled shear stress. In the analysis, it was assumed that the creep function within the LVE limits will be best represented by a generalised model, consisting of series of parallel Kelvin elements and a single Hooke element. Based on earlier analyses, it was decided that a setup of five Kelvin elements ($n = 3$) and a single Hooke element is sufficient to correctly describe the bitumen's strain changes. The estimation of the model's parameters required the use of the non-linear least squares method. In order to avoid issues related to the correct determination of the initial values, the MCalibration[®] program, utilizing an implemented set of solvers, was used for identification of Maxwell model parameters [31]. The model's quality of matching with the experimental data was determined using two qualitative measures, i.e.: the modified determination coefficient R^2 and normalized median absolute deviation NMAD [32].

2.5. FTIR Spectroscopy

Attenuated Total Reflection (ATR) Fourier Transform Infrared (FTIR) allows for determining the quantitative evaluation of the impact of certain bitumen additives or modifiers by measuring the spectrum of absorption of certain functional groups [33]. In the case of the bitumen sample's testing, some radiation is subject to reflection and some is absorbed by the sample. Taking into consideration the crystal's (diamond's) background, it is possible to obtain the sample's specific spectrum. The ageing effect and the presence of synthetic wax caused a change in the absorption bands' intensity in the analysed spectral range of $1.700 \div 1.724 \text{ cm}^{-1}$ in relation to the reference sample's spectrum. The test results' evaluation featured an analysis of the surface fields of the absorption bands obtained for given bitumens prior to and after foaming. The test was conducted using the FTIR spectrometer from Nicolet iS5, in the spectral range of $7.800 \div 350 \text{ cm}^{-1}$ with the ATR (Attenuated Total Reflectance) attachment. The obtained absorption spectrums were initially corrected by the introduction of a baseline correction and correction characteristic for the ATR attachment.

3. TEST RESULTS

The primary objective of the foundational research was to evaluate TRA modified bitumen in relation to 50/70 neat bitumen. Within the scope of this research, various tests were conducted, including penetration, softening point, and brittleness temperature acc. to

Fraass methodology. The findings were subsequently compared with the outcomes of 50/70 neat bitumen (denoted as 50/70 ref), which serves as the base bitumen for the modification. The research findings are visually represented in Figure 4.

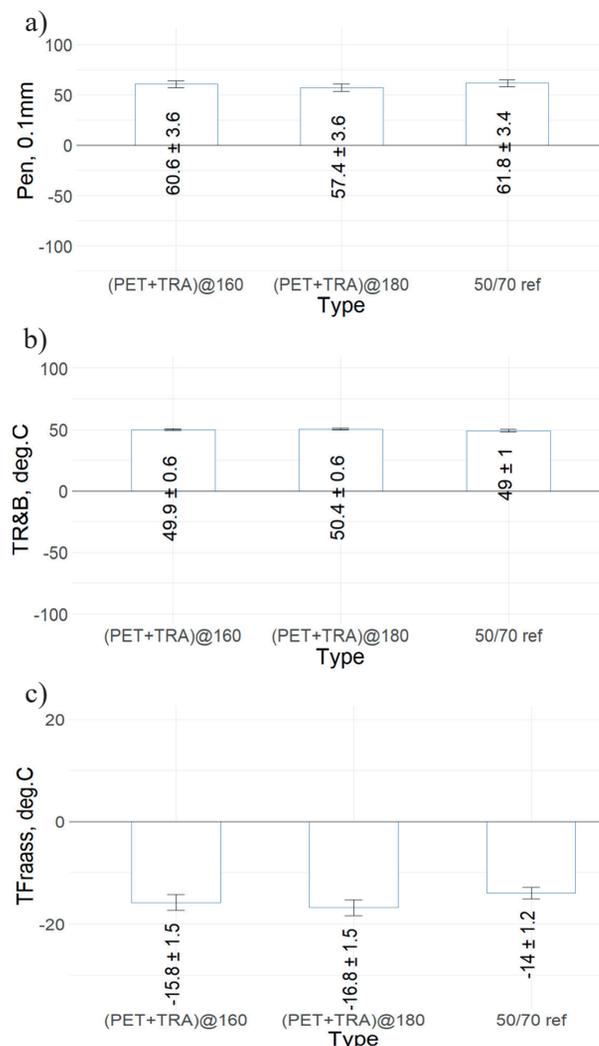


Fig. 4. Basic tests of TRA modified bitumen: a) penetration; b) softening point; c) Fraass temperature

Upon analysis of the test results, no substantial differences were identified in the evaluation of penetration or softening point, with an increase of approximately 1°C , when compared to 50/70 neat bitumen (Fig. 4a-b). Conversely, a notable reduction in the brittleness temperature was recorded, approximately -3°C , at a mixing temperature of 180°C (Fig. 4c). The conventional properties of bitumen did not deliver significant evidence to advocate for the integration of TRA into bitumen. Consequently, additional creep tests for bitumen were conducted in conformity with the MSCR procedure. The outcomes of the evaluation of the parameters J_{nr} and $\%ER$ for

50/70 neat bitumen and its modification with the TRA modifier, mixed at temperatures of 160°C and 180°C, are illustrated in Figure 5.

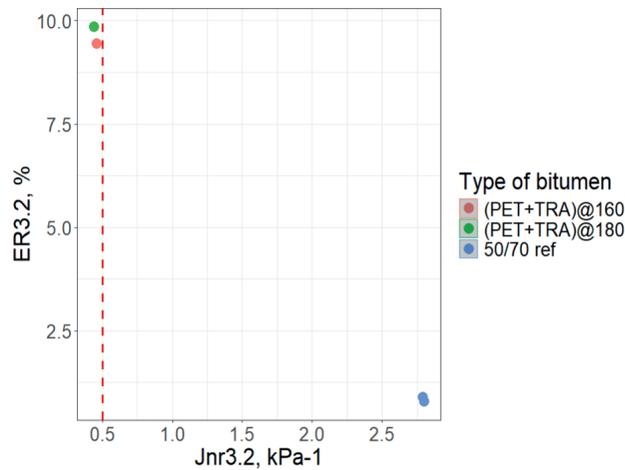


Fig. 5. MSCR test results

As indicated in Figure 5, the outcome of the irreversible component of the Jnr susceptibility was observed to be less than 0.5 kPa⁻¹ (red line), which implies that the 50/70+TRA bitumen is suitable for the production of mineral and bitumen mixtures designed for heavy traffic. According to the standards set by AASHTO M 332 and AASHTO T 350, such bitumen can be classified for application in extremely heavy traffic conditions (exceeding 30 million axles (ESAL) and vehicle parking speeds of less than 20 km/h), similar to SBS-modified bitumen. In contrast, the irreversible compliance value of 50/70 neat bitumen, when compared to 50/70+TRA, was five times greater, measuring less than 2.5 kPa⁻¹. It is noteworthy that the elastic recurrence value %ER also experienced a substantial increase, with levels fluctuating around 10%, whereas conventional 50/70 neat bitumen did not attain a value exceeding 1%. This phenomenon can be attributed to the incorporation of robust amine bonds by the addition of TRA to the bitumen, which enhanced the cohesion of the asphalt at elevated temperatures, while also marginally reducing the temperature according to Fraass breaking point temperature. To estimate the magnitude of the rheological effects accompanying the deformation of the tested bitumen, particularly those containing TRA, a simulation of the deformation trajectory over time, derived from the MSCR study, was conducted and described by calibrating the parameters of the generalized Maxwell model. The results of the parameter identification for the physical Maxwell model of bitumen incorporating TRA are presented in Figure 6.

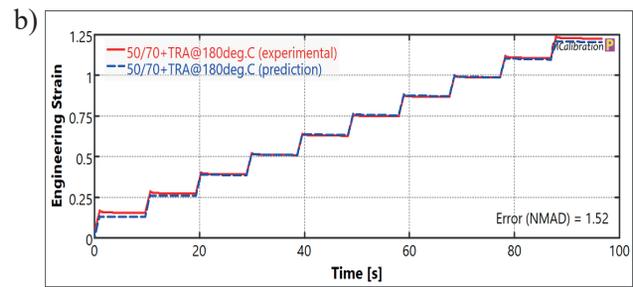
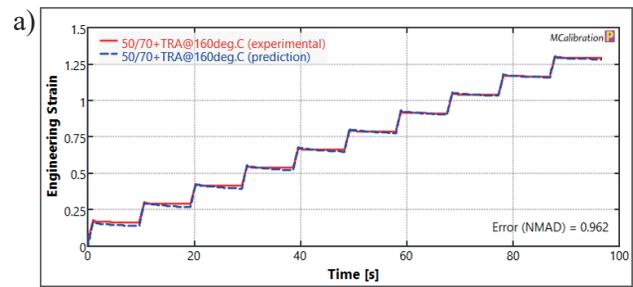


Fig. 6. Identification of the Maxwell model parameters for the deformation in time based on the MSCR test: a) 50/70+TRA at 160°C; b) 50/70+TRA at 180°C

A summary of the parameters of the generalized Maxwell model for the three elements of the Prony series is presented in Table 2.

Table 2. Generalised Maxwell model parameters

Parameter	50/70+TRA at 160°C	50/70+TRA at 180°C	50/70 referenced
G_0 , MPa	1.15	1.96	0.1
g_1	0.41	0.29	0.37
t_{1r} , s	3.3e-5	0.024	0.001
g_2	0.49	0.27	0.39
t_{2r} , s	0.03	2.7e-6	0.002
g_3	0.0004	0.0002	0.26
t_{3r} , s	9.9	0.0008	0.013
R^2	0.97	0.96	0.86
NMAD, %	0.96	1.52	7.6

In evaluating the outcomes of parameter identification for the generalized Maxwell model (refer to Fig. 2), it is pertinent to observe that the relaxation time for 50/70 neat bitumen, when TRA is incorporated, is decuple in comparison to 50/70 neat bitumen. This implies that the deformation rate will be definitely faster for 50/70 bitumen relative to TRA-modified 50/70 neat bitumen. Consequently, the inclusion of the TRA compound favorably mitigates the acceleration of deformation during the rutting process of asphalt mixtures (mma) subjected to vehicular traffic. Additionally, the instantaneous shear modulus, G_0 , in modified bitumen exceeds that of

50/70 neat bitumen by over 20-fold. This denotes that a substantial initial stiffness can be anticipated within the minor deformation spectrum, impacting both the formability of mma and the energy dissipation rate attributable to rheological phenomena. The integration of bitumen with TRA results in a novel composite material characterized by enhanced stiffness at elevated operating temperatures and diminished sensitivity to loading durations compared to 50/70 neat bitumen. Moreover, the modified bitumen exhibits a reduced brittle temperature, suggesting retention of a flexibility reserve at lower temperatures. This outcome is presumably attributable to the superior compatibility of TRA with bitumen and the noted secondary polymerization occurring in the degraded PET in bitumen phase. Hence, this approach presents itself as a viable alternative for recycling PET, acknowledged for its complexity in processing and chemical inertia. Future research endeavors will concentrate on the evaluation of alternative amines and the refinement of the TRA and bitumen amalgamation process.

4. CONCLUSIONS

On the basis of the research performed and the analysis of the results, the following conclusions were formulated:

- The aminolysis process of the PET plastomer led to the formation of terephthalamide with reduced mechanical strength, thereby enhancing its processability and subsequently improving the efficiency of the bitumen homogenization process with TRA.
- The modification of 50/70 neat bitumen via the incorporation of TRA resulted in a decrease in Fraass breaking point temperature by roughly -3°C , whereas the softening point demonstrated an increment of approximately $+1^{\circ}\text{C}$ in comparison to the conventional 50/70 bitumen.
- The incorporation of TRA into 50/70 neat bitumen resulted in a fivefold reduction in irreversible susceptibility compared to 50/70 neat bitumen, bringing it to below 0.5 kPa^{-1} . Conversely, the elastic recovery value in bitumen 50/70+TRA, as determined by MSCR, increased by approximately tenfold compared to 50/70 neat bitumen.
- The presence of TRA in bitumen resulted in a tenfold reduction in the relaxation rate of bitumen and an approximate twentyfold enhancement in stiffness relative to 50/70 neat bitumen. This modification is anticipated to beneficially influence the reduction of MMA deformation rate under high operational temperature conditions when TRA is utilized.

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