Fractional Viscoelastic Modeling of Antirutting Response of Bituminous Binders

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Abstract: A three-parameter fractional model, composed by a springpot in series with a dapshpot, was employed to describe the rheological behaviour of a set of polymer-modified bituminous binders obtained by combining a single-base bitumen with different types of syrene-butadiene-styrene copolymers at different concentrations. Experimental strain-time curves gathered from creep-recovery shear tests carried out by means of a dynamic shear rheometer at various temperatures were found to be in good agreement with model predictions. The fractional approach leads to a synthetic yet exhaustive description of the viscoelastic behavior of all materials. Variation of model parameters, each of which with a precise physical meaning, appears to be coherent with different binder compositions and testing conditions, reflecting the variations in terms of polymer concentration and temperature. **DOI: 10.1061/(ASCE)EM.1943-7889.0001081.** © *2016 American Society of Civil Engineers.*

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Introduction

The term rutting is commonly used to indicate one of the main distress types affecting asphalt pavements, consisting in the accumulation of permanent deformation produced by repeated traffic loading (Fig. 1). Formation of ruts on pavement surfaces produces low levels of comfort and safety for vehicles and users, thus reducing overall pavement serviceability. This failure mode is promoted by the presence of severe traffic and environmental conditions, such as those characterized by high percentages of heavy slow-moving trucks and high in-service temperatures. For these reasons, rutting resistance of pavements is greatly influenced by the rheological properties of bituminous binders used in the top bound layers, due to their time-dependent and temperature-dependent nature (Monismith et al. 1985; Sousa et al. 1991; Delgadillo and Bahia 2010).

Different experimental approaches have been proposed to evaluate the antirutting potential of bituminous binders. The one originally introduced in the framework of the SUPERPAVE grading system is based on viscoelastic parameter $G^*/\mathrm{sen}\delta$ (Harrigan et al. 1994), but its limits have been widely demonstrated by several studies (Bahia et al. 2001; D'Angelo and Dongre 2002). Other protocols currently adopted in binder characterization for the same purpose refer either to the zero-shear viscosity concept (Sybilski 1996) or to parameters derived from material response under repeated creep loading (D'Angelo et al. 2007). In both cases, however, experimental studies have highlighted the existence of significant drawbacks related to the fact that they seem not to be fully adequate to capture actual properties of specific types of binders, such as polymer modified ones (Morea et al. 2010; Zoorob et al. 2012).

More recently, Santagata et al. (2013, 2015) introduced a method based on single creep-recovery shear tests carried out at predefined loading and recovery times. Experimental findings showed the effectiveness of the method when applied to binders modified with polymers and crumb rubber since the high levels of induced strain allow differentiation between reversible and non-reversible components of response. Moreover, effects caused by variations of additive type, composition, structure, and dosage can be clearly highlighted.

While creep-recovery tests are relatively simple and quick to perform, modeling of the experimental data gathered from measurements appears not to be a trivial task. This is due to the fact that binders, as many viscoelastic materials, exhibit a creep-relaxation behavior of the power-law type (Mainardi 1994; Hilfer 2000; Mainardi 2010), with the consequent need of introducing a great number of elements (and thus of parameters) in fitting operations when trying to describe such a response by means of classical exponential-type functions. Furthermore, since parameters are subjected to several restrictions, the employed numerical algorithm may revert to a complex constrained least-squares problem (Sorvari and Malinen 2007).

Although power-law expressions for creep-recovery functions can be assumed a priori, without a physical sound derivation (Jäger et al. 2007; Füssl et al. 2014), they are implicitly generated by assuming a constitutive law of fractional type, i.e., involving time derivatives with noninteger order of strains and stresses (Carpinteri and Mainardi 1997). For this reason, approaches to viscoelasticity based on fractional calculus have attracted the attention of researchers for at least two decades, involving different materials and applications (Barpi and Valente 2003, 2004; Atanckovic et al. 2013,

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Fig. 1. Presence of ruts on pavement surface due to deformation in the top bound layers (image by Ezio Santagata)

2015, Deseri et al. 2014; Di Paola and Zingales 2012; Di Paola et al. 2013, 2014; Paggi and Sapora 2015; Zopf et al. 2015). Furthermore, in a wider context of engineering applications (not only related to viscoelasticity), fractional calculus has been widely recognized to be an efficient tool to model both memory effects and nonlocal interactions (for recent applications, see, for instance: Mehdinejadiani et al. 2013; Sapora et al. 2013; Tarasov and Trujillo 2013; Carpinteri et al. 2014; Ahmadian et al. 2015).

In the field of asphalt pavement engineering, few fractional approaches have been proposed for the study of the behavior of bituminous binders and mixtures (Oeser et al. 2008; Celauro et al. 2012; Fecarotti et al. 2012). In particular, a three-parameter model consisting in a dashpot in series with a springpot was recently introduced by Sapora et al. (2016) (see also Di Mino et al. 2016), with the goal of reducing the number of fitting parameters (Oeser et al. 2008) and of increasing prediction accuracy (Celauro et al. 2012; Fecarotti et al. 2012). Such a model was successfully applied to creep-recovery test data obtained at various temperatures (ranging from 58 to 76°C) for two different modified binders (containing thermoplastic polymer and crumb rubber, respectively).

In the research work reported in this paper, the study of the antirutting response of modified bituminous binders by means of a fractional viscoelastic approach presented in (Sapora et al. 2016) is extended to a wider array of materials, containing several polymers at different concentrations, over a larger range of temperatures (from 40 to 80°C). Results are analyzed with the specific goal of highlighting the mechanical meaning of model parameters and of linking them to the characteristics of the considered binders. Analogies and differences with the results shown in Sapora et al. (2016) are addressed.

The analysis presented herein lies in the framework of linear viscoelasticity of one-dimensional media, coherently with the experimental tests which were carried out: the solution is thus given in an analytical form, and the model parameters are obtained through a least-square fitting procedure. The numerical handling of fractional viscoelastic material models through finite-element analysis can be found in Müller et al. (2013).

Materials and Methods

Modification of bitumen by means of polymers has become very popular in the binder industry since it may provide a significant enhancement of performance-related properties (King et al. 1986; Collins et al. 1991; Wardlaw and Shuler 1991). For this reason, the use of polymer-modified binders (PMBs) in road construction represents one of the preferred design options in the case of heavy-duty pavements subjected to intense traffic flow and severe environmental conditions.

Table 1. Description of Modified Bituminous Binders Used in the Experimental Investigation

PMB code	Description: Type of modifier	Description: Dosage (%)
A-3	A-radial SBS with low styrene content	3
A-6		6
B-3	B-radial SBS with high styrene content	3
B-6		6
C-3	C-linear SBS with high styrene content	3
C-6		6

Among the various types of polymers employed as chemical agents for the production of PMBs, styrene-butadiene-styrene (SBS) synthetic rubbers are characterized by widespread diffusion: they are triblock copolymers belonging to the category of the so-called thermoplastic elastomers that form when adequately combined with a base bitumen, a cross-linked network promoted by thermal and chemical bonding mechanisms. The elastomeric network consists of polystyrene domains connected by elastic butadiene threads, which contribute to improve stiffness and elasticity of the material, especially at high temperatures. The ability of such polymers to form a completely cross-linked structure depends upon their concentration within the bituminous matrix and the thermodynamics of the combined system (Bahia et al. 2001).

Materials used in the experimental investigation described in this paper include several PMBs obtained from the factorial combination of a single base bitumen (70/100 pen grade), three different types of SBS, and two concentrations. SBS polymers (Atofina, Milano, Italy) employed for modification were commercially available products differing from each other in terms of chemical composition (low and high styrene content) and molecular structure (linear and radial chains). Preparation of PMBs was carried out in the laboratory by making use of a bath-oil mixer, operated at 180°C for 60 min with a mixing speed of 800 rpm. Polymers were added to bitumen at two different dosages, equal to 3 and 6% by weight of base bitumen.

A descriptions of the complete set of modified binders is given in Table 1. All of them were tested in short-term aged conditions simulated by means of the rolling thin-film oven test (RTFOT), according to the AASHTO procedure (AASHTO 2009). Examples of scanning electron microscope (SEM) images illustrating the microstructure of PMBs with higher SBS concentration are reported in Fig. 2.

The protocol adopted for binder testing consists in a creep phase, during which a shear stress of 100 Pa is applied for a loading time of 900 s, followed by a recovery phase, in which the load is removed and strain evolution is monitored for additional 900 s. Creep-recovery tests were carried out at different temperatures, selected depending upon the type of binder in order to emphasize its actual viscoelastic response. In particular, 40 and 60°C were adopted for PMBs containing 3% polymer (A-3, B-3, and C-3), whereas 60 and 80°C were used for those characterized by 6% concentration (A-6, B-6, and C-6).

The testing device employed to carry out creep-recovery tests was a Dynamic Shear Rheometer (DSR) (Physica MCR 301 DSR from Anton Paar, Graz, Austria), an air-bearing stress-controlled device equipped with a permanent magnet synchronous

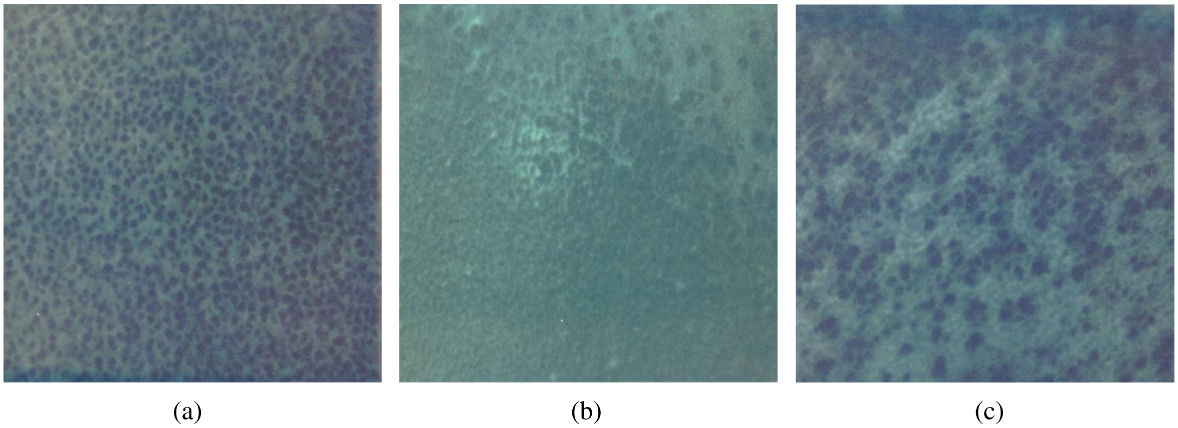

Fig. 2. SEM images corresponding to binders: (a) A-6; (b) B-6; (c) C-6

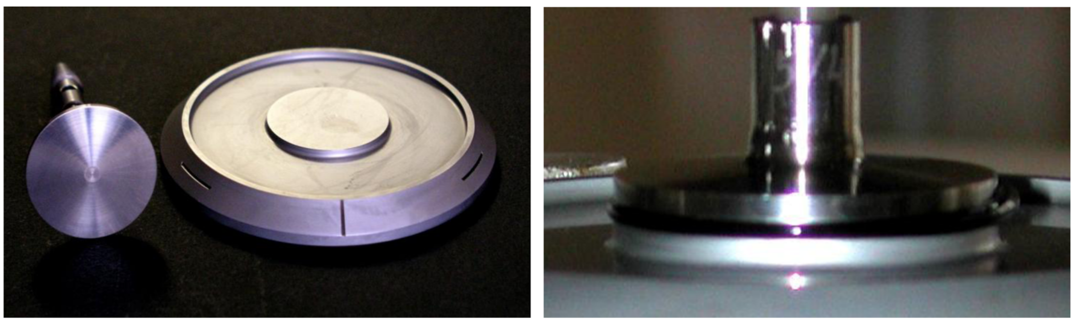

Fig. 3. Sensor system and test configuration

drive (minimum torque = 0.1 μNm, torque resolution <0.1 μNm) and an optical incremental encoder (Anton Paar, Graz, Austria) for the measurement of angular rotation (resolution <1 μrad). A cone-plate sensor system (Anton Paar, Graz, Austria) with a 35-mm diameter and 4° cone angle was used for measurements.

Test specimens were prepared by pouring the binder (preheated at 150°C for the time needed to achieve a sufficient fluidity) on the lower plate of the sensor system and by thereafter sandwiching it between the plate and the upper cone. The amount of material placed on the plate was slightly overdosed in order to allow the formation of a proper bulge at the periphery of the sample after reaching final measurement position. With the aim of preventing temperature gradients throughout the binder volume, specimens were conditioned at test temperature for 15 min before actual measurements.

The employed sensor system and the final test configuration reached after specimen preparation are displayed in Fig. 3.

Implemented Fractional Model and Parameter Calibration

The fractional model implemented in the present study (Fig. 4) includes a springpot connected in series with a dashpot (Sapora et al. 2016). While the dashpot simply accounts for time-dependent viscous response under loading, the springpot is a structural element with a fractional order α governing the transition from elastic to viscous behavior.

The strain-stress (ε-σ) constitutive law of the model is the following:

$$\frac{d\varepsilon(t)}{dt} = \frac{1}{b_1}\frac{d^{1-\alpha}\sigma(t)}{dt^{1-\alpha}} + \frac{1}{b_2}\sigma(t) \tag{1}$$

where b_1 and $\alpha \in (0, 1)$ = the springpot parameters; and b_2 = the dynamic viscosity related to the dashpot.

The definition of the fractional derivative of order α related to a generic function f can be expressed in the following form (Samko et al. 1993):

$$\frac{d^\alpha f(x)}{dx^\alpha} = \frac{1}{\Gamma(1-\alpha)}\int_0^x \frac{f'(y)}{(x-y)^\alpha}dy \quad \alpha \in (0, 1) \tag{2}$$

where Γ = the Euler-Gamma function.

According to Eq. (2), the α-derivative of a function reverts to the function itself for $\alpha = 0$, and to the first-order classical derivative for $\alpha = 1$. As α varies, the mechanical meaning of b_1 in Eq. (1) obviously changes, passing from a stiffness (Force $\times$ Length^{-2}) for $\alpha = 0$, to a viscosity (Time $\times$ Force $\times$ Length^{-2}) for $\alpha = 1$.

The creep function $D(t)$ related to the constitutive law (1) can be derived as

$$D(t) = \frac{t^\alpha}{b_1\Gamma(1+\alpha)} + \frac{t}{b_2} = \frac{t}{b_2}\left(1 + \frac{b_2}{b_1\Gamma(1+\alpha)}t^{\alpha-1}\right) \tag{3}$$

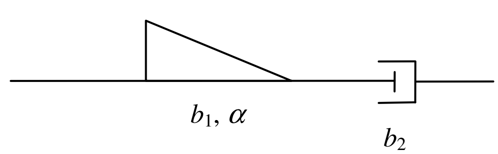

Fig. 4. Fractional model implemented in the analysis

Table 2. Fitted Parameters α, b_1 (Ns^{α}/m^2), $b_2(Ns/m^2)$, and Mean Percentage Error Corresponding to PMBs Containing 3% SBS Tested at 40 and 60°C

	A-3				B-3				C-3			
T (°C)	α	b_1	b_2	$\bar{\delta}$	α	b_1	b_2	$\bar{\delta}$	α	b_1	b_2	$\bar{\delta}$
40	0.570	0.966	2.14	1.44	0.567	0.596	3.12	1.37	0.731	0.0133	0.000	3.77
60	1.000	0.173	0.173	4.06	1.000	0.188	0.188	3.88	1.000	0.464	0.464	10.3

whereas the creep-recovery constitutive relationship can be written as

$$\varepsilon(t) = \left\{ \left[\frac{t^{\alpha}}{b_1 \Gamma(1+\alpha)} + \frac{t}{b_2} \right] U(t) - \left[\frac{(t-t^*)^{\alpha}}{b_1 \Gamma(1+\alpha)} + \frac{(t-t^*)}{b_2} \right] U(t-t^*) \right\} \sigma_0 \qquad (4)$$

where $U(t)$ = the unit-step (or Heavisde) function; t^* = the time at which unloading starts; and σ_0 = constant applied stress.

The limit cases of the considered fractional model are important to identify. Elastic response is maximized for $\alpha = 0$, in which case the springpot reverts to a spring and b_1 plays the role of a stiffness; in this case the model coincides with the well-known Maxwell model consisting in a spring in series with a dashpot. On the other hand, viscous behaviour is amplified the most for $\alpha = 1$; the model then reverts to two dashpots in series, each with the same viscosity ($b_1 = b_2$).

In order to fit the model to experimental data obtained from creep-recovery tests described in the previous section, the three model parameters α, b_1, and b_2 were determined by means of a numerical algorithm that evaluates the coefficients of a nonlinear regression function through a least-squares estimate. Due to possible settlement phenomena observed in the initial phase of creep, data recorded in the first 1% of the loading history were neglected (Sapora et al. 2016).

Values of α, b_1, and b_2 obtained from fitting, together with the corresponding absolute mean percentage error $\bar{\delta}$, are reported in Tables 1 and 2 for PMBs containing 3 and 6% polymer, respectively.

Discussion of Results

Shear strain test data obtained for PMBs characterized by the lower SBS content (A-3, B-3, and C-3) and theoretical results provided by Eq. (4) with fitted parameters reported in Table 2 are diagrammed as a function of time in Figs. 5–7. For each material and test condition, experimental and theoretical curves appear to be very close to each other, indicating that a good agreement was generally found. This is coherent with $\bar{\delta}$ values that are abundantly lower than 5%. The only exception is represented by Binder C-3 tested at 60°C, for which the creep experimental curve deviates from the theoretical one, showing a trend characterized by the increase of strain rate with loading time; the corresponding mean percentage error is nonnegligible, reaching a value of the order of 10%.

In all cases, materials tested at 40°C exhibit a certain amount of delayed elastic strain, which is partially recovered during the unloading phase. On the contrary, at 60°C, the accumulated strain at the end of the loading phase is fully irreversible. This evidence can be explained by considering that the raise in temperature produces the breakdown of physical crosslinks within polymer styrene domains; in such conditions the SBS added to the bituminous matrix acts as a simple filler that contributes to increase viscosity of the binder without improving its elasticity.

Such a transition from viscoelastic to viscous behaviour is reflected in the variation of fitted fractional parameters. In particular, values of α determined at 40°C range, approximately, between 0.55 and 0.75, whereas at 60°C they become equal to 1 with the fractional model being reduced to two dashpots in series ($b_1 = b_2$). In any case, values of α equal to 0.55 or higher indicate the predominance of viscous components in material's response even at 40°C (i.e., the springpot is more similar to a dashpot than to a spring) suggesting the added amount of elastomer (3%) is not sufficient to create a continuous polymeric network in accordance with results published elsewhere (Lu and Isacsson 1997). Moreover, the increase of the fractional exponent when passing from 40 to 60°C appears to be in contrast with findings of other studies (Di Mino et al. 2016), in which α is showed to decrease with temperature (together with b_1 and b_2). Both trends can be considered acceptable from a practical point of view since the global response

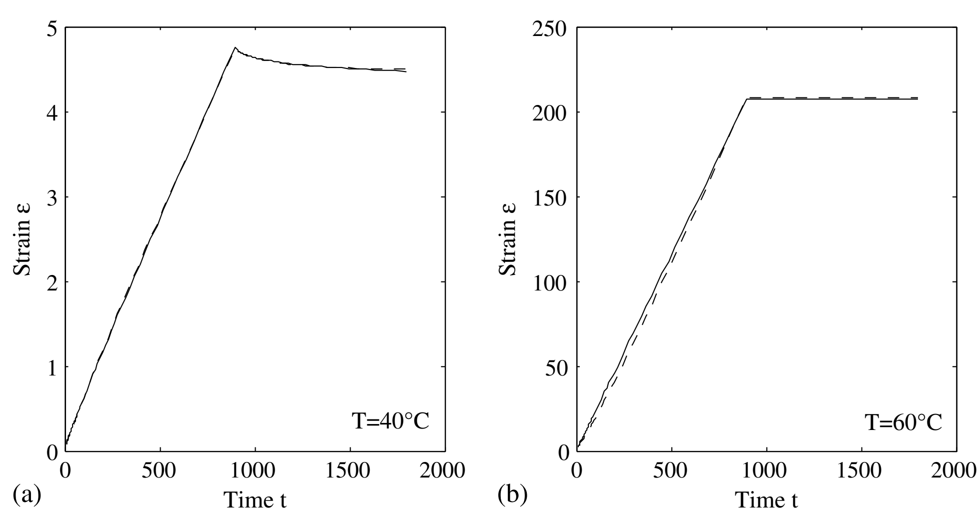

Fig. 5. Creep-recovery curves of Binder A-3 tested at (a) 40°C; (b) 60°C: experimental data (dotted line) and theoretical results (solid line)

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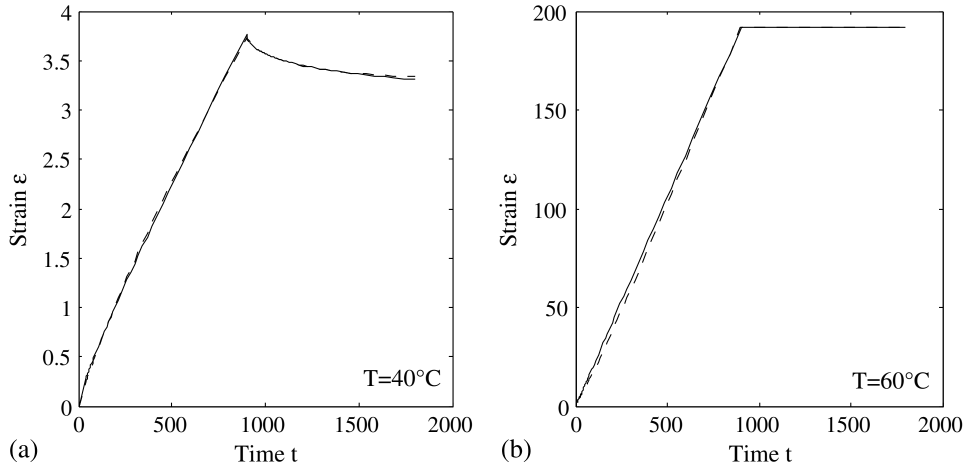

Fig. 6. Creep-recovery curves of Binder B-3 tested at (a) 40°C; (b) 60°C: experimental data (dotted line) and theoretical results (solid line)

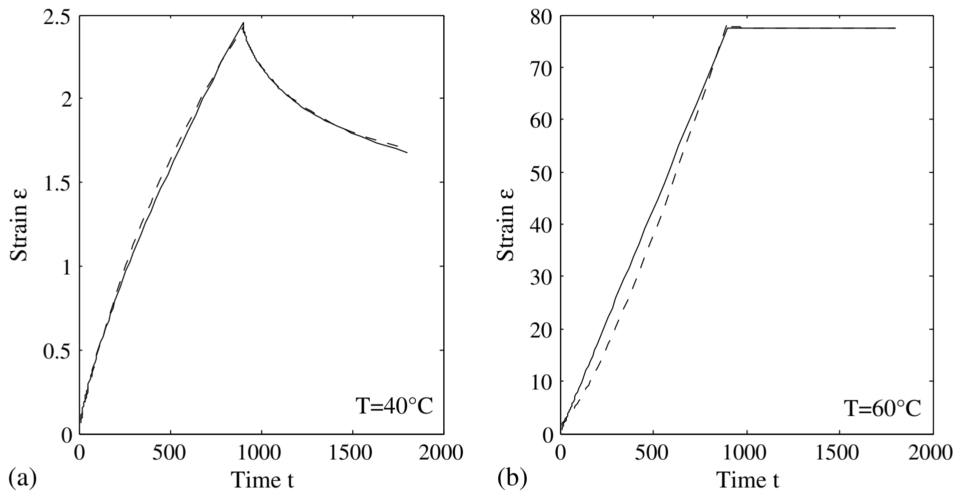

Fig. 7. Creep-recovery curves of Binder C-3 tested at (a) 40°C; (b) 60°C: experimental data (dotted line) and theoretical results (solid line)

Table 3. Fitted Parameters α, b_1 (Ns^{α}/m^2), b_2(Ns/m^2), and Mean Percentage Error $\bar{\delta}$ Corresponding to PMBs Containing 6% SBS Tested at 60 and 80°C

	A-6				B-6				C-6			
T (°C)	α	b_1	b_2	$\bar{\delta}$	α	b_1	b_2	$\bar{\delta}$	α	b_1	b_2	$\bar{\delta}$
60	0.259	0.044	6.41	1.75	0.104	0.063	61.4	0.69	0.156	0.0738	39.9	1.46
80	1.000	0.091	0.091	1.66	0.245	0.109	2.88	3.87	0.334	0.427	0.402	10.3

is described by the combined effects of α, b_1, and b_2 variations; however, the first one seems to be more logical from a theoretical point of view, matching the extreme cases described in the previous section. Similar considerations are valid for values reported in Table 3.

By comparing the materials to each other, it is clearly noticed that Binder C-3 exhibits the most elastic response at $T = 40$°C, its behaviour being described by a single springpot. On the other hand, Binders A-3 and B-3 present nearly the same value for α but different values of b_1 and b_2: the springpot contribution to overall deformation appears to be more significant for A-3, which presents a lower b_2/b_1 ratio [Eq. (3)], even though B-3 is less deformable in absolute terms. At 60°C, C-3 shows the highest value of b_2 that, as specified before, assumes the meaning of a dynamic viscosity, thus indicating the highest resistance to flow for such a material, followed, in order, by B-3 and A-3. This is coherent with the characteristics of the various SBSs used for modification and more specifically with the presence, in Type C and Type B, of a higher styrene content, which contributes to increase the hardness of the polymers and, consequently, of the corresponding polymer–bitumen blends.

Diagrams reporting the results obtained for highly modified binders (A-6, B-6, and C-6) are shown in Figs. 8–10. Also in this case a good fitting is observed ($\bar{\delta}$<5%) with the only exception, once again, given by binder containing Type C SBS tested at the higher temperature. In fact, the creep curve at 80°C shows a trend similar to that displayed in Fig. 7(b), which appears to be not well described by the proposed model (the mean absolute error exceeds 10%).

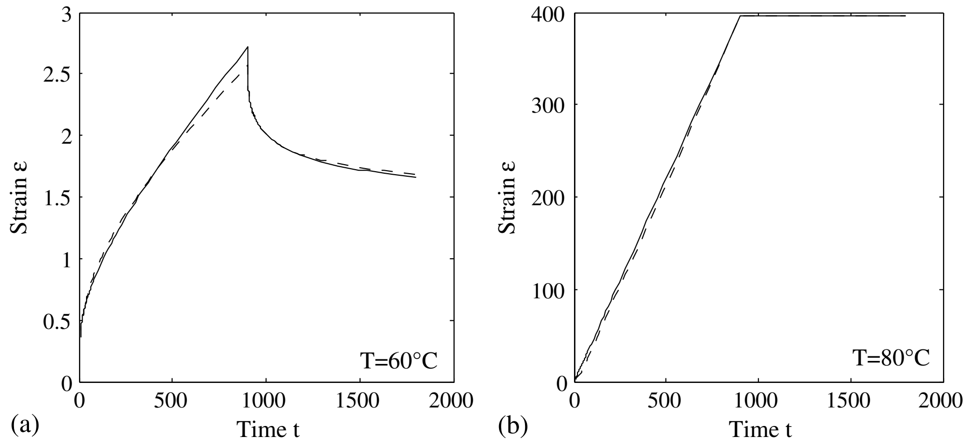

Fig. 8. Creep-recovery curves of Binder A-6 tested at (a) 60°C; (b) 80°C: experimental data (dotted line) and theoretical results (solid line)

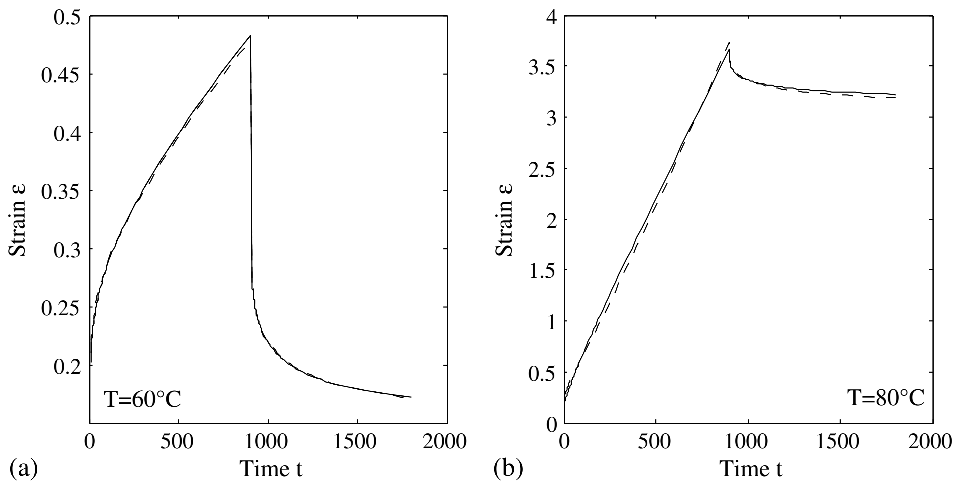

Fig. 9. Creep-recovery curves of Binder B-6 tested at (a) 60°C; (b) 80°C: experimental data (dotted line) and theoretical results (solid line)

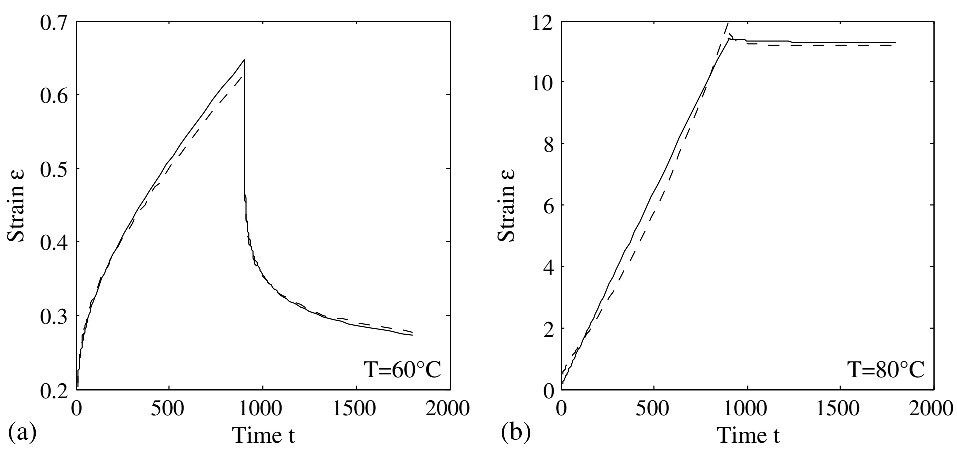

Fig. 10. Creep-recovery curves of Binder C-6 tested at (a) 60°C; (b) 80°C: experimental data (dotted line) and theoretical results (solid line)

This type of shear-thinning behaviour can be therefore considered peculiar of the linear SBS used in this investigation when subjected to high levels of shear deformations (Stastna et al. 2003).

In contrast with data illustrated before, α values are generally very low (except in the case of A-6 tested 80°C, which will be commented on later) with springpot elements approaching a condition corresponding to the spring case. This can be evidently attributed to the presence of a high amount of polymer that results in the formation of a diffused cross-linked structure with a pronounced rubberlike behaviour.

However, a distinction can be made between Binder A-6 and the Binders B-6 and C-6.

In fact, at 60°C, the latter ones behave like viscoelastic solids, which are able to recover most of the deformation experienced during the loading phase. This can be evidently attributed to the presence of a high styrene content, which promotes the formation of rigid polystyrene domains within the bituminous matrix. If compared to each other, B-6 prevails over C-6 in terms of stiffness and degree of elasticity, probably due to its radial/branched molecular structure. At 80°, as obvious, both materials become less stiff (lower values of $b_{2/}b_1$) and less elastic (higher values of α) even though they maintain a certain aptitude to recover deformation.

Binder A-6 at 60°C shows a typical viscoelastic behaviour characterized by a lower stiffness and degree of elasticity with respect to that of abovementioned Binders B-6 and C-6 (lower values of b_2/b_1 and higher values of α) and, as previously highlighted, at 80°C reaches viscous flow conditions ($\alpha = 1$ with $b_1 = b_2$). Such evidence can be linked with the low styrene content of the SBS used for the preparation of this type of PMB, that results in weak bonding between styrene blocks at very high temperatures, thus overcoming polymer networking effects.

Conclusions

The three-parameter (α, b_1, and b_2) fractional viscoelastic model consisting of a springpot in series with a dashpot was employed in the research study reported in this paper to model the creep-recovery experimental behaviour of several modified bituminous binders, differing in polymer type and concentration. While in a previous paper (Sapora et al. 2016) results were analyzed with the specific purpose of verifying the capability of the model to capture peculiarities of material response, the attention is here focused on linking the model parameters to variations in materials' composition and testing conditions.

From the original findings of the study, the following main conclusions can be drawn.

Experimental strain-time curves obtained from testing were found to be in good agreement with model predictions, as indicated by absolute mean percentage error values always being lower than 5%. The only exceptions are represented by binders containing the linear SBS polymer that showed a typical shear-thinning behaviour when subjected to high levels of deformations. This confirms the effectiveness of the fractional approach that leads to a synthetic but exhaustive description of the viscoelastic behavior of materials with a precise physical meaning of model parameters: whereas b_1 and b_2 describe the stiffness and dynamic viscosity of the binder, α governs the global elastic–viscous transition.

Variations of abovementioned parameters appear to be coherent with the composition of different materials and testing conditions, reflecting the variations in terms of polymer concentrations and temperatures. In particular, materials characterized by low polymer content exhibited high values of springpot parameter α, indicating the materials' response as being significantly governed by viscous components. These become even more predominant as temperature increases, with the concurrent decrease of b_2/b_1 ratio values. In the case of highly modified materials, α values drop dramatically, showing the evolution from a viscous to a pronounced viscoelastic behavior. This is especially true for binders containing SBS (both linear and radial) characterized by a high styrene content, the response of which can be assimilated to that of a viscoelastic solid. Such results suggest that SBS is able to form a continuous cross-linked network only if added to the base bituminous matrix in a sufficiently high amount. In such conditions, a binder containing radial SBS with high styrene content was found to prevail at both considered temperatures, as demonstrated by higher values of α and b_2/b_1 ratio.

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