Evaluation of healing potential of bituminous binders using a viscoelastic continuum damage approach

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Abstract

A simplified viscoelastic continuum damage model was adopted to analyze experimental data obtained from healing tests consisting of continuous shear loading interrupted by single rest periods. Tests were conducted on various bituminous binders with the purpose of highlighting the aptitude of the proposed methodology in discriminating and ranking their healing potential. Healing was quantitatively assessed by introducing two indicators (HR_C and HR_S) derived from damage characteristic functions. Results revealed the existence of some irreversible damage after continuous shear loading. In addition, even though polymer modification enhances healing properties of base bitumen, in some cases neat binders may outperform polymer-modified binders.

Keywords: bituminous binder; polymer modification; healing; viscoelastic continuum damage theory.

1 Introduction

The capability to recover stiffness and strength after being damaged under loading represents a peculiar property of bituminous mixtures used in road paving applications, commonly referred to as healing. Healing is generally associated to phenomena of micro-crack repair that take place within the material when external loading is removed and sufficient rest time is given [1-5]. Since pavement structures are subjected to non-continuous vehicle loading due to the intermittent nature of traffic, microdamage healing occurring during non-loading phases contributes in extending their actual fatigue life. Experimental evidence of such an occurrence has been provided in several research studies, based both on laboratory testing [6-11] and field data [12-13].

Mechanisms driving microdamage healing are not yet fully understood. By referring to the model originally proposed for polymers [14], microdamage healing is depicted as a two-stage process consisting in the closure of micro-cracks due to wetting and in the subsequent gain of mechanical properties due to interdiffusion of molecules across crack surfaces. In their recent study, Sun et al. [15] were able to identify wetting and molecular diffusion stages by means of fluorescence microscope observations. They also found that healing rate at the wetting stage is lower than at the diffusion stage. Healing rates have been proven to increase as rest periods are extended [16-18]. In addition to time, temperature is the other external factor which significantly affects healing rates. An increase in temperature leads to the softening of the material which flows more quickly towards the crack surfaces. Moreover, at higher temperatures overall mobility of molecules is greater, thus promoting interdiffusion and consequent homogenization of interfaces in contact. Based on complex viscosity measurements, Tang et al. [19] identified the softening point as the optimal healing temperature of penetration-graded bitumen.

When focusing on intrinsic material-related factors, the key role played by chemical composition and molecular morphology needs to be considered. Sun et al. [20] showed that the healing rate of bituminous binders is highly sensitive to the content of aromatics. Kim et al. [6] linked healing capability to the length of molecules and to their degree of branching. In particular, the presence of molecules characterized by long chains and limited branching promotes interdiffusion, with the consequent improvement of healing properties. Correlation between self-diffusivity and molecular morphology was confirmed by molecular dynamics simulations conducted by Bhasin et al. [21]. Further confirmation, based on the results of laboratory testing, was provided by Santagata et al. [22] that found the healing ratio of neat binders to be positively influenced by the relative amount of long molecules within the oil phase. In addition to chemical composition and morphology of the base bitumen, the use of polymer modifiers may have a great impact on healing response. In fact, studies conducted on polymer-modified binders indicated enhanced performance for this type of materials in terms of stiffness recovery and fatigue life extension [23-24].

A number of methods and indexes have been proposed by researchers in order to evaluate healing properties of bituminous binders and mixtures. In the case of binders, test procedures are mostly based on the use of the dynamic shear rheometer. Shen et al. [25-26] characterized bituminous binders by subjecting them to intermittent loading sequences in which short rest periods (varying from 0 to 6 seconds) were inserted. Healing effects were then analyzed by adopting an approach that relies on the ratio of dissipated energy change concept. Tan et al. [17] used a healing test consisting of an initial fatigue test with continuous loading followed by a rest period (varying from 1 to 48 h) and by another fatigue test. Two healing indicators were introduced: the first one was calculated as the ratio of the initial modulus values after and before rest, while the second one was calculated as the ratio between the number of loading cycles applied in the two fatigue tests until failure. Results reported in the paper showed that both indicators well reflect healing potential. A similar protocol was employed by Shan et al. [27] that established three different indexes based on the curves of normalized modulus, elastic modulus and viscous modulus versus loading cycles. These indexes were found to lead to the same ranking of the studied materials. Santagata et al. [22-23, 28-30] used test procedures based on cyclic loading interrupted by the inclusion of either a single long rest period or multiple shorter rest periods at predefined levels of damage. In both cases, healing response was assessed on the basis of stiffness recovery exhibited by the binders during the unloading phases. Stimilli et al. [31] and Canestrari et al. [24] performed multiple rest tests and referred to the recovered number of cycles after a given number of rest periods as a criterion for the evaluation of healing properties.

The viscoelastic continuum damage (VECD) model represents a powerful tool in characterizing damage behaviour of viscoelastic materials under loading. Based on the work potential theory developed by Schapery [32-33], VECD theory relates the variation of pseudostiffness (C) to an internal state variable (S) resulting in a damage evolution law (C(S)) which has been demonstrated to be unique for each material and independent from testing conditions (mode of loading, frequency and amplitude) [34-36]. VECD modeling has been widely and successfully used to predict damage evolution of bituminous mixtures subjected to uniaxial cyclic fatigue tests [37-40]. More recently, it has been extended to bituminous binders and mastics tested under cyclic torsion [41-42]. Continuum damage mechanics also represents the theoretical framework used to determine fatigue laws from linear amplitude sweep (LAS) tests. LAS tests, along with a simplified version of the VECD model, have been adopted by Xie et al. [43] to characterize healing in bituminous binders.

This paper presents the results of an experimental study in which the VECD approach was used to analyze test data obtained from healing tests consisting of continuous shear loading interrupted by single rest periods. Two different indicators derived from damage evolution curves before and after rest were introduced to quantify healing. Experiments were conducted on a set bituminous binders of various origins and types with the purpose of highlighting the aptitude of the proposed methodology in discriminating and ranking the healing potential of materials with very different characteristics.

2 Materials and methods

Materials selected for the experimental investigation included two neat bitumens (NA and NB) which were sampled from refineries operating on crudes of different source, and two polymer-modified binders (PMBL and PMBH) which were originated from base bitumen NB by adding a low and high percentage of styrene-butadiene-styrene (SBS) elastomer, respectively (of the order of 2-3 % and 4-6 % by weight of base bitumen, according to the undisclosed processing scheme adopted by the manufacturer). Description of binders, along with their penetration grades and performance grades (PGs), is given in Table 1. All materials were evaluated after being treated with the Pressure Aging Vessel (PAV) in order to simulate long-term ageing, which is considered the most representative condition for the analysis of damage phenomena.

Table 1. Bituminous binders used in the experimental study

Binder code	Description	Penetration grade	PG
NA	Neat	70/100	64-16
NB	Neat	70/100	58-22
PMBL	Modified with SBS (low polymer content)	45/65	70-22
PMBH	Modified with SBS (high polymer content)	50/70	76-22

The experimental procedure followed in the study involved the use of fatigue and healing tests.

Fatigue tests consisted in continuous oscillatory shear loading applied in the stress-controlled mode and prolonged until complete specimen failure. They were conducted with the primary purpose of determining the characteristic damage behaviour of the materials to be assumed as a reference for healing analysis. Temperature was set at 20 °C, which represents a typical intermediate temperature used in fatigue studies. Frequency and stress amplitude were fixed at 10 Hz and 250 kPa, respectively. These testing conditions ensure a sufficiently high initial stiffness of materials which has been indicated as a crucial factor in preventing the occurrence of phenomena like instability flow and edge effects, thus highlighting true internal microdamage [44].

Healing tests consisted in two continuous oscillatory shear loading phases with an intermediate rest period. Temperature, frequency and stress level were set equal to those of fatigue tests. A single rest period of 6 h was adopted. Such a choice was suggested by the need of providing sufficient time to the materials to recover mechanical properties lost before loading removal, thereby allowing their healing potential to be highlighted [28]. The rest period was inserted in healing tests at predefined levels of material integrity loss expressed in terms of pseudostiffness reduction. This was practically done by interrupting the first oscillatory loading phase when shear strain amplitude exhibited by the specimen reached a specific threshold corresponding to the C value associated to the target integrity loss level as determined from reference fatigue test curves. The selected values of C before loading removal, C_{BR}, were 0.75 (25 % integrity loss) and 0.5 (50 % integrity loss) for all binders. Due to its peculiar behavior, in the case of heavily modified binder PMBH an additional C_{BR} value equal to 0.1 (90 % integrity loss) was used. It should be underlined that the approach adopted in this study is different from that of Xie et al. [43], who applied rest periods at various percentage values of the internal state variable at failure S_f. Before running fatigue and healing tests, test specimens were subjected to few oscillatory shear loading cycles at a strain level kept lower than the linear viscoelastic limit of the material. The norm of the complex modulus $|G^*|$ measured during this pre-loading phase was used to quantify sample-to-sample variability.

Fatigue and healing analysis was preceded by a preliminary rheological characterization in which standard frequency sweep tests at multiple temperatures were carried out in order to determine linear viscoelastic (LVE) properties of the considered binders. Temperatures varied from 4 to 82 °C with 6 °C increments; at each temperature, frequencies ranged in the 1-100 rad/s interval. Isothermal curves obtained from frequency sweeps were reduced to a reference temperature by means of time-temperature shifting which relied upon the WLF (Williams-Landel-Ferry) function [45] and entailed fitting of shifted data to the CA (Christensen-Anderson) model [46]. Operations illustrated above led to the construction of continuous master curves describing the variation of $|G^*|$ as a function of reduced loading frequency ω_R. An example of master curve determined at 20 °C for one of the binders (NA) is shown in Figure 1. Two rheological quantities of primary interest for VECD modeling were also derived from LVE analysis. The first one is the LVE modulus, $|G^*|_{LVE}$, which in fatigue and healing tests is used to calculate peak pseudo-strains from peak physical strains according to the elastic-viscoelastic principle. The second one is the slope of the relaxation modulus master curve in log-log space, m, which can be estimated from the slope of the steady-state modulus master curve in the log-log space [42].

Experimental measurements were performed by means of a Physica MCR 301 DSR from Anton Paar Inc., an air bearing stress-controlled device equipped with a permanent magnet synchronous drive (minimum torque equal to 0.1 μNm, torque resolution < 0.1 μNm) and an optical incremental encoder for the measurement of angular rotation (resolution < 1 μrad). Testing geometry adopted for fatigue and healing tests consisted of 8-mm parallel plates with 2 mm gap between the plates. In the case of frequency sweep tests, the abovementioned geometry was used at lower temperatures (4-34 °C), whereas at higher temperatures (34-82 °C) it was substituted by 25-mm plates with 1 mm gap. All measurements were performed in at least duplicate runs and average results were used for analysis.

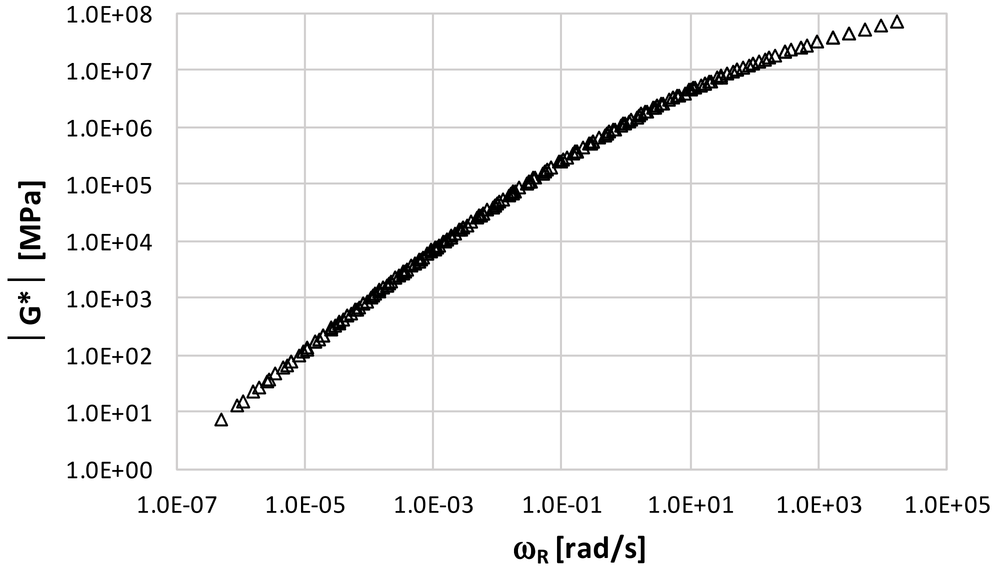


Figure 1. Master curve of the norm of the complex modulus at 20 °C (binder NA)

3 Analysis and results

Data obtained from fatigue and healing tests, combined with those gathered from LVE characterization, were analyzed following a methodology based on a simplified version of the VECD model proposed by Safaei et al. [42]. The aforementioned methodology consists of three subsequent steps that, starting from physical peak shear strains measured as a function of time, include determination of 1) peak shear pseudo-strain γ^R, 2) pseudostiffness C and 3) internal state variable S. Since pseudostiffness is a function of the internal state variable, the calculation process ultimately leads to the determination of the C(S) function which represents the damage characteristic function of the considered binder.

Calculation of pseudo-strain was performed by applying the elastic-viscoelastic principle [32] which allows the conversion of physical strain into pseudo-strain, thus eliminating time-dependent effects. Pseudo-strain is defined by Eq.1:

$$\gamma^R = \frac{1}{G_R}\int_0^{t_R} G(t_R - \zeta)\frac{d\gamma}{d\zeta}d\zeta \tag{1}$$

where G(t) is the relaxation shear modulus, t_R is reduced time, ζ is the variable of integration and G_R is a reference modulus. In the case of cyclic loading with zero mean strain, Eq.1 can be simplified in the following form (with G_R set equal to 1):

$$\gamma_p^R = \gamma_p * \left(|G^*|_{LVE}(\omega_R)\right) \quad (2)$$

where γ_p^R is the peak pseudo-strain at a given cycle, γ_p is the corresponding peak physical strain, $|G^*|_{LVE}$ is the LVE modulus at the reduced frequency ω_R. The use of Eq.2 instead of Eq.1 implies that analysis can be conducted without considering the full time history of stresses and strains, and this is the basic feature of simplified VECD modelling.

The ratio of peak shear stress τ_p to peak pseudo-strain γ_p^R gives the pseudo-stiffness C according to Eq.3:

$$C = \frac{\tau_p}{\gamma_p^R \cdot MR} \quad (3)$$

in which MR (Modulus Ratio) is the ratio between the modulus measured in the pre-loading phase of fatigue tests and the LVE modulus determined from frequency sweep tests. Such a parameter is introduced in Eq.3 to account for sample-to-sample variability.

The internal state variable S, which represents the state of damage within the specimen, is used to describe damage growth in viscoelastic materials according to the damage evolution law derived from Schapery's work potential theory and expressed by Eq.4:

$$\frac{dS}{dt} = \left(-\frac{\partial W^R}{\partial S}\right)^{\alpha} \quad (4)$$

in which α is the damage evolution rate and W^R is the pseudo-strain energy function.

Damage evolution rate was determined according to the relation α = 1+1/m, that in the case of bituminous binders has been proven to be more effective than the other one commonly used in VECD modelling (α = 1/m) [42]. The pseudo-strain energy function is calculated as follows:

$$W^R = \frac{1}{2} \cdot \left(\gamma_p^R\right)^2 \cdot C(S) \quad (5)$$

Derivation of the internal state variable as a function of time can be obtained by combining Eq.5 with Eq.4 and by solving via numerical integration, thus leading to the solution given in Eq.6:

$$S = \sum_{i=1}^{N} \left[\frac{DMR}{2} \cdot \left(\gamma_{p,i}^R\right)^2 (C_{i-1} - C_i)\right]^{\frac{\alpha}{1+\alpha}} \cdot [t_i - t_{i-1}]^{\frac{1}{1+\alpha}} \quad (6)$$

where i refers to the loading cycle number and t refers to time.

Damage characteristic functions of the investigated binders derived at a reference temperature of 20 °C from fatigue tests are displayed in Figure 2.

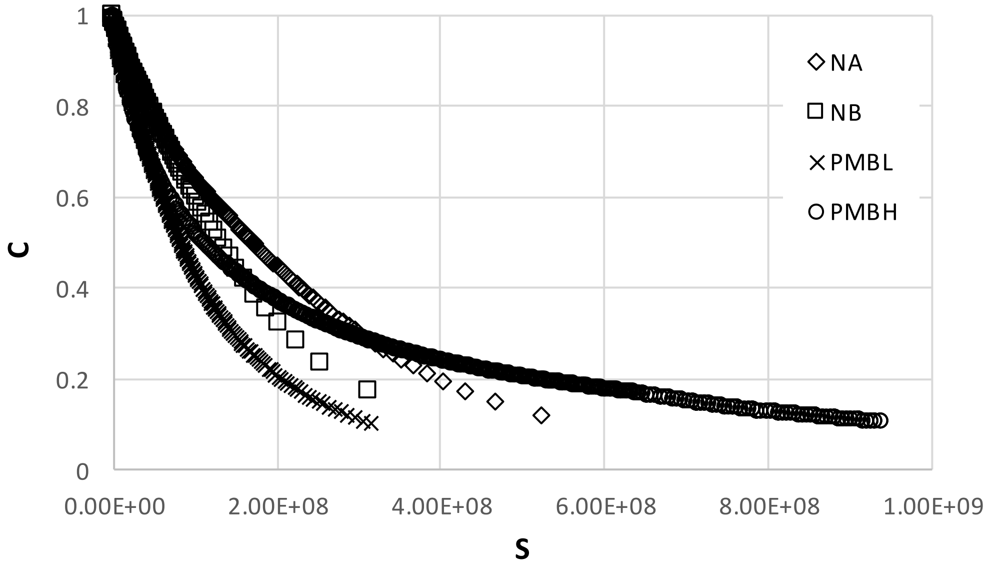


Figure 2. Damage characteristic curves obtained from fatigue tests for the investigated binders

All C(S) curves show an initial portion that follows an almost linear trend until a given level of damage, after which decrease of pseudostiffness takes place at a rate that is gradually reduced. Despite these common features, curves are clearly distinguished from each other, thus indicating substantial differences in damage behaviour exhibited by the various materials. In particular, changes in damage rates appear to be more pronounced for modified binders with respect to neat binders. This is due to the presence of the SBS polymer and can be linked to non-linearity effects which are usually observed for this type of materials during the earlier stages of continuous cyclic loading [47]. In the specific case of heavily modified binder PMBH, the C(S) curve shows in its final part a second linear segment which is characterized by a significantly reduced slope and suggests the presence of a steady-state damage stage extending towards very high levels of the internal state variable. During this steady-state stage, the binder is capable of accumulating a great amount of damage with a relatively low reduction of material integrity. As previously mentioned, with the purpose of evaluating healing response in this peculiar condition, the C_{BR} value of 0.1 was selected for this binder in addition to those considered for other materials.

Results of healing tests carried out on the binders are reported in Figures 3 to 6, which show the damage evolution curves obtained before and after rest periods inserted at various C_{BR} values.

In all cases, increase of C and decrease of S are observed following the unloading phase, clearly reflecting the occurrence of healing. This was quantitatively assessed by introducing two healing indicators, referred to as the integrity healing ratio (HR_C) and the damage healing ratio (HR_S). By definition, both healing parameters may range between 0 (no healing) and 1 (full healing).

The first indicator was calculated by means of Eq.7:

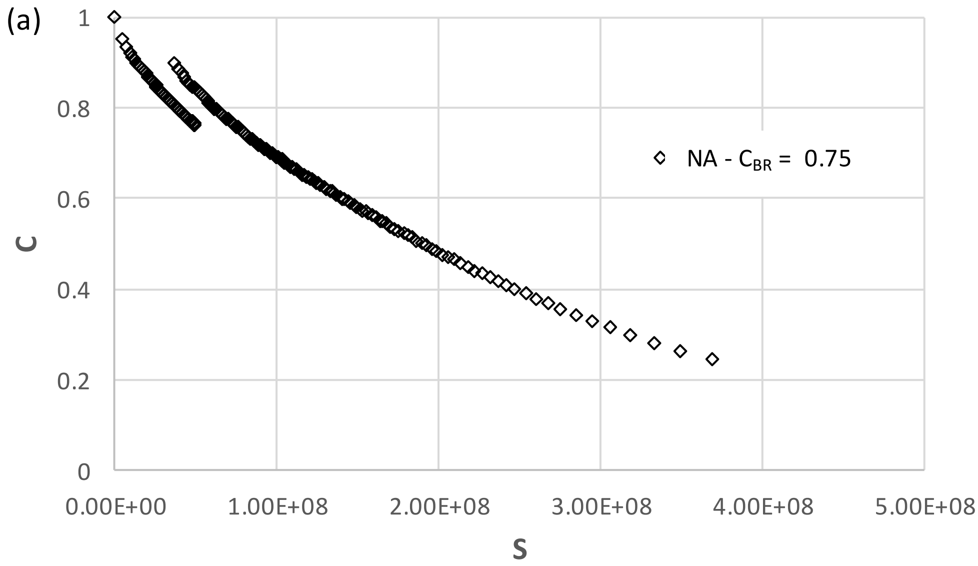
$$HR_C = \frac{C_{AR}-C_{BR}}{1-C_{BR}} \tag{7}$$

where C_{BR} and C_{AR} are the pseudostiffness values at the end of the first loading phase and at the beginning of the second loading phase, respectively. Such a parameter represents the relative amount of lost integrity which is recovered during the rest period.

The second indicator was calculated by means of Eq.8:

$$HR_S = \frac{S_{BR}-S_{AR}}{S_{BR}} \tag{8}$$

where S_{BR} and S_{AR} are the internal state variable values at the end of the first loading phase and at the beginning of the second loading phase, respectively. This indicator represents the relative amount of damage accumulated in the first loading phase which is then restored during the rest period.



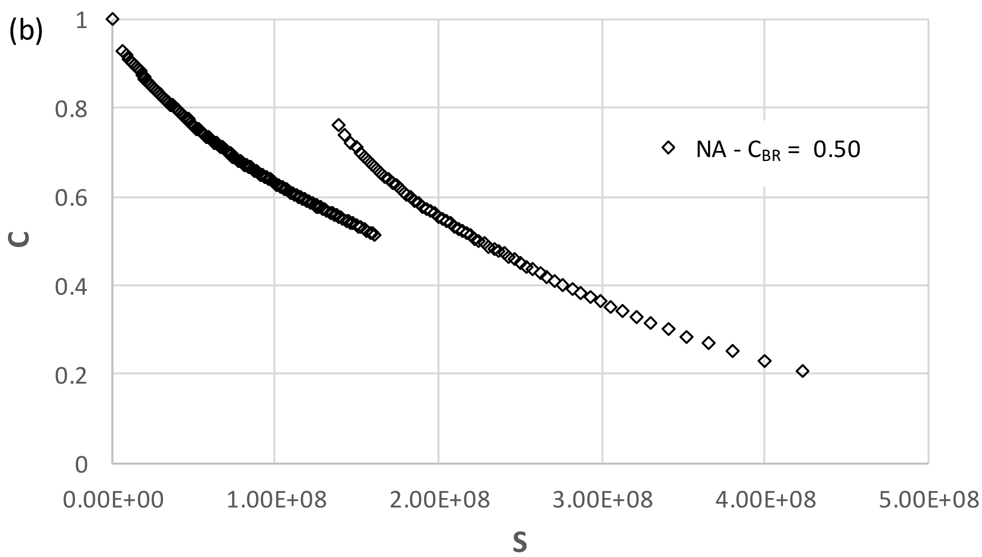


Figure 3. Damage characteristic curves obtained from healing tests for neat bitumen NA – (a) C_{BR} = 0.75 (b) C_{BR} = 0.50

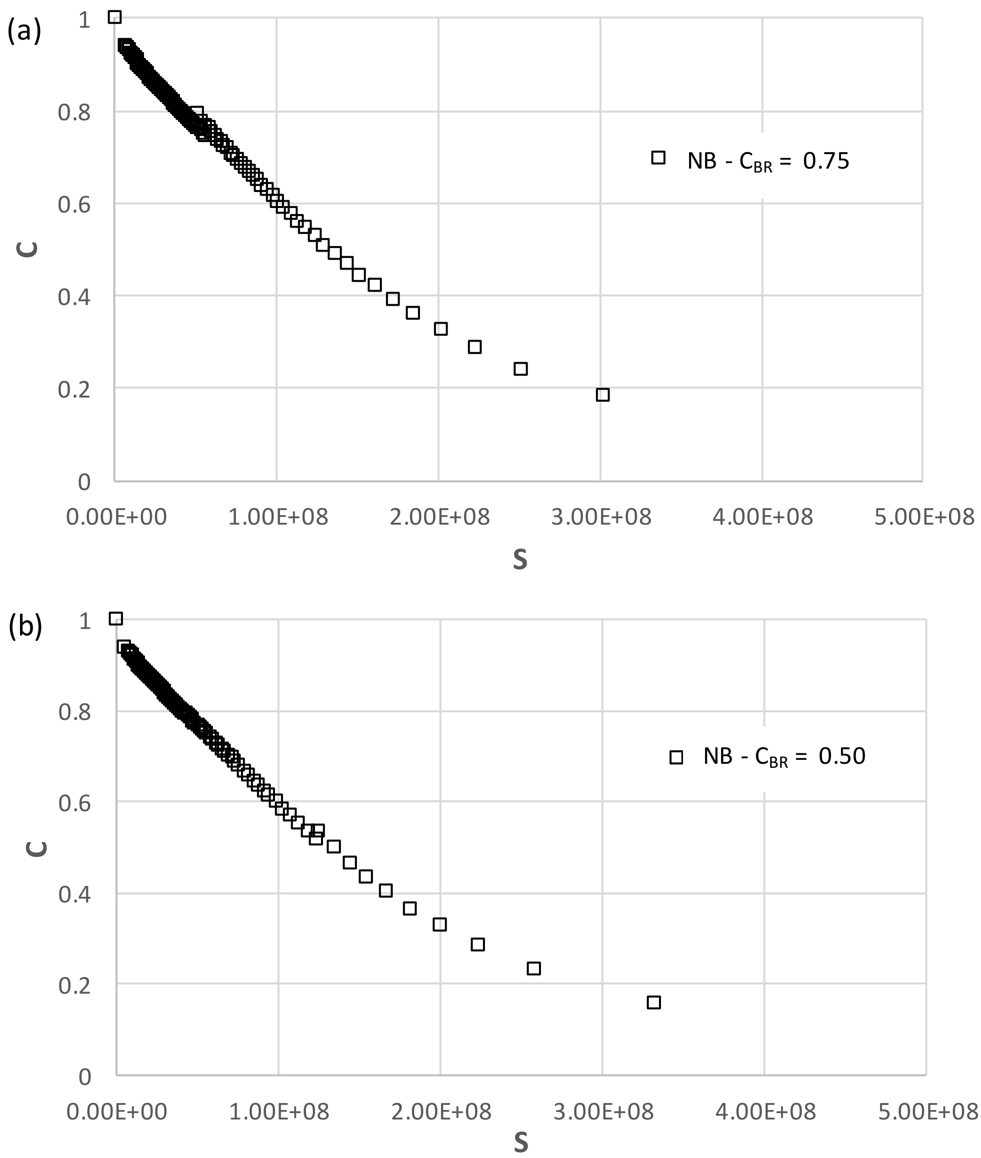


Figure 4. Damage characteristic curves obtained from healing tests for neat bitumen NB – (a) C_{BR} = 0.75 (b) C_{BR} = 0.50

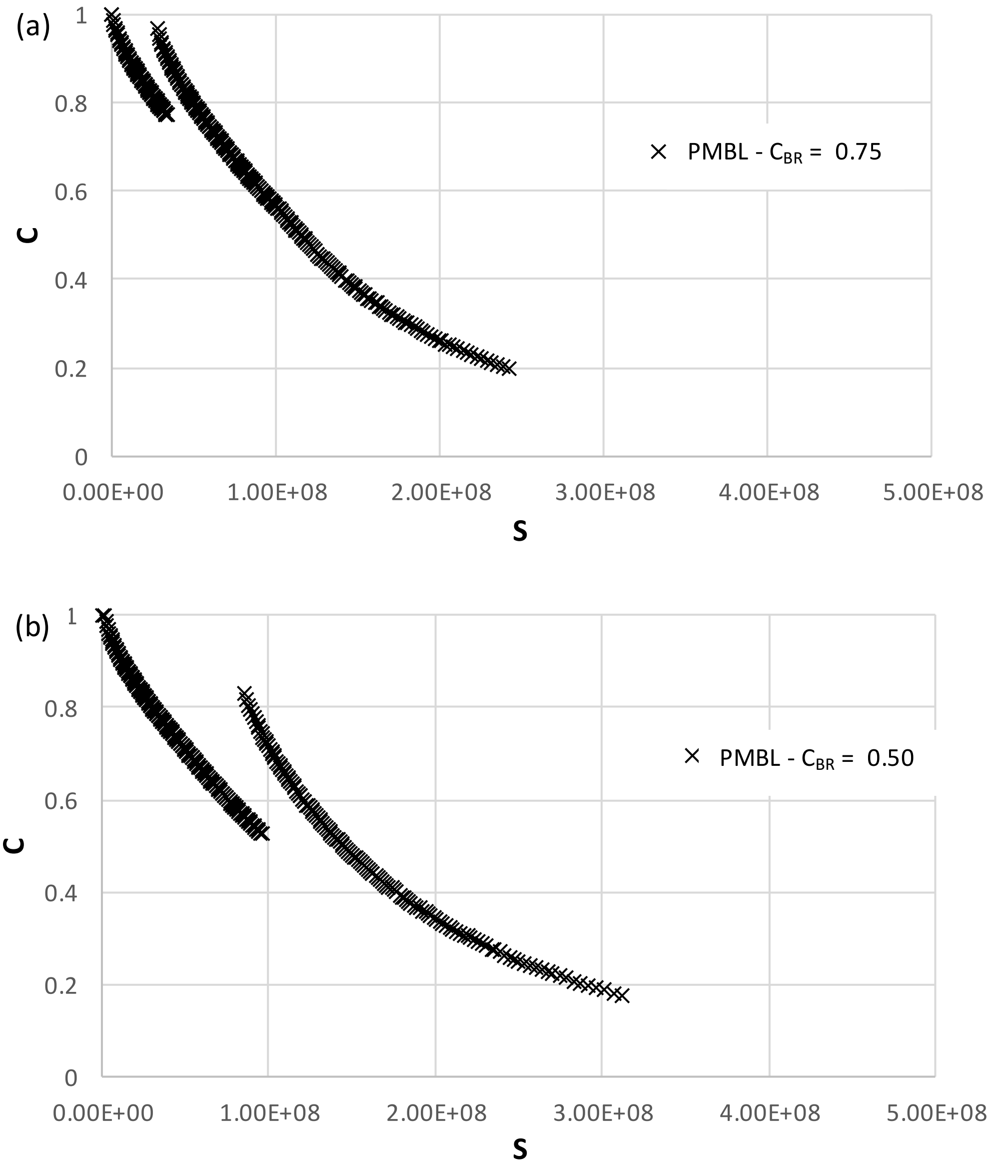


Figure 5. Damage characteristic curves obtained from healing tests for SBS-modified binder PMBL – (a) C_{BR} = 0.75 (b) C_{BR} = 0.50

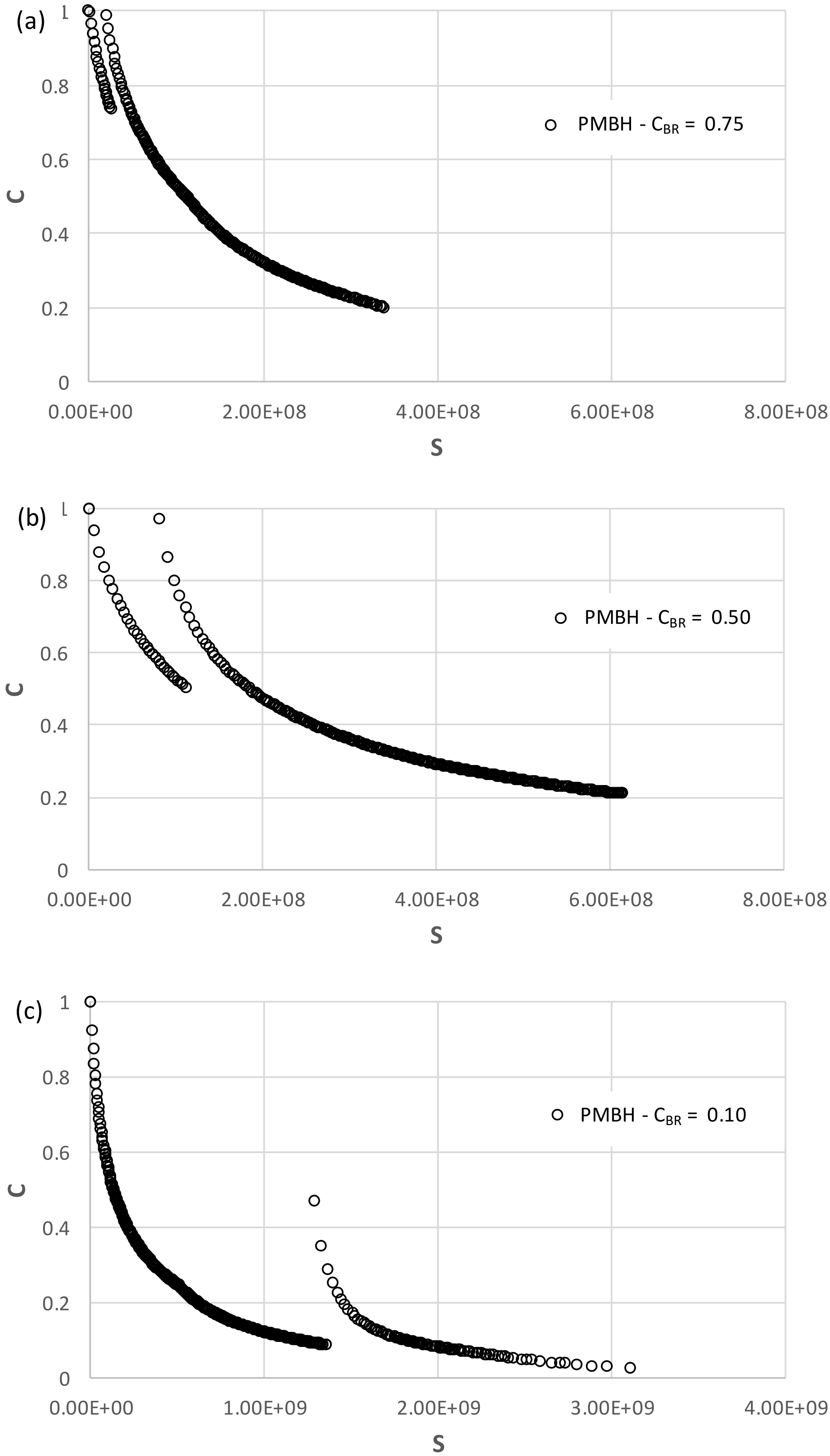


Figure 6. Damage characteristic curves obtained from healing tests for SBS-modified binder PMBH – (a) C_{BR} = 0.75 (b) C_{BR} = 0.50 (c) C_{BR} = 0.10

HR_C and HR_S values obtained for all tested binders are represented in Figure 7. It can be observed that recovery of material integrity and damage after rest periods depends upon the level of pseudostiffness loss imposed before loading removal. In particular, a lower percent of healing recovery is observed at a higher level of pseudostiffness loss. This indicates that part of total damage induced in the specimen by cyclic loading seems to be intrinsically irreversible and that such irreversible damage increases as total damage increases. In order to explain this outcome, it is speculated that bonds formed across cracked interfaces become progressively weaker as crack size becomes progressively larger. Crack size plays a prominent role in influencing rate of wetting and, therefore, the process underlying crack closing [5]. Effective wetting allows crack surfaces to come into contact, thus promoting subsequent randomization and interdiffusion of molecules across wetted surfaces. It follows that larger cracks are unlikely to heal as effectively as smaller cracks, thus hindering the recovery of the original internal structure and strength. As a consequence, the presence of larger cracks associated to greater levels of damage results in a reduced healing potential.

HR_C and HR_S values are higher in the case of neat bitumen NA with respect to neat bitumen NB when compared at the same level of imposed pseudostiffness loss. Even though belonging to the same penetration grade, these binders originated from crudes of different source and this reflects into very different aptitudes to heal. Healing properties of neat bitumen NB were greatly enhanced by SBS modification. These beneficial effects can be linked to the self-healing capability of the polymer and to the local bridging of microcracks induced by the polymer network [28]. Due to the higher SBS content, binder PMBH shows a superior healing performance than binder PMBL and displays a better healing response also when compared to the neat binders. It is worth nothing that binder PMBH, after being loaded up to 90 % pesudostiffness loss, has the residual capability to recover about 40 % of its initial integrity and 5 % of total accumulated damage.

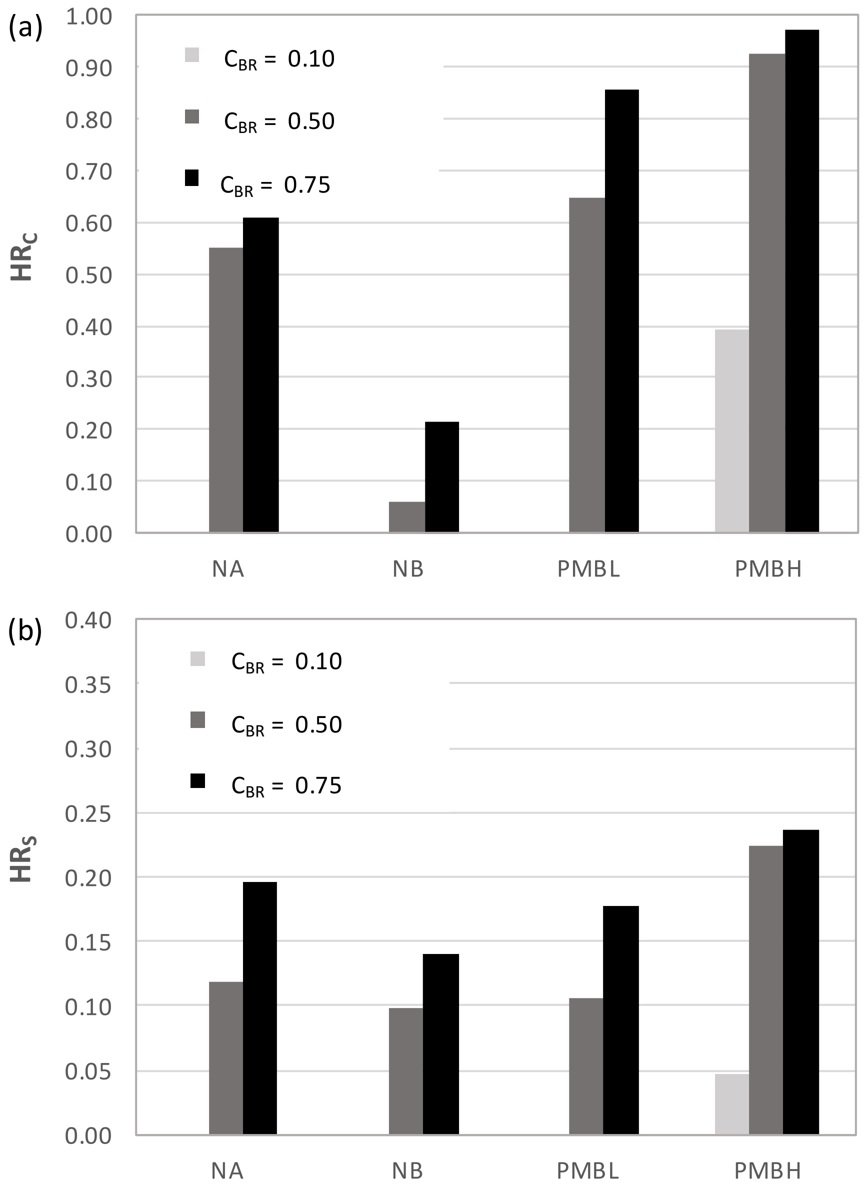


Figure 7. Healing indicators of the investigated binders – (a) HR_C (b) HR_S

In the case of binders PMBL and NA, relative ranking depends upon the healing indicator which is used for comparison. In particular, PMBL ranks higher than NA when referring to HR_C, while it ranks lower when considering HR_S. Some authors have highlighted the fact that the increase of stiffness immediately after rest periods does not necessarily result in a corresponding increase of strength; therefore, the use of stiffness recovery is not recommended for healing evaluation since it may lead to an overestimate of the true healing potential [5]. According to this rationale, it can be concluded that in the analysis it is preferable to refer to HR_S values and that, as a consequence, neat binder NA shows a slightly better healing performance than modified binder PMBL. This confirms that polymer-modified binders are not necessarily characterized by a better healing response when compared to neat bitumens [43], and that healing tests are mandatory in order to establish relative rankings when focusing on this specific performance-related issue.

4 Summary and conclusions

In the experimental study described in this paper, a viscoelastic continuum damage approach was used to evaluate and compare the healing potential of a set of different bituminous binders. Binders were characterized by means of healing tests consisting in two oscillatory shear loading phases with an intermediate rest period inserted at predefined levels of pseudostiffness reduction. Traditional fatigue tests were also carried out as a reference. Results revealed that the proposed test procedure coupled with viscoelastic continuum damage modelling provides an effective tool in discriminating and ranking the healing response of binders of various types and origins. Evidence of healing stems from the comparison of damage characteristic curves before and after rest periods that showed, for all considered binders and testing conditions, a recovery of material integrity (indicated by pseudostiffness increase) and of internal damage (revealed by internal state variable decrease). Healing was quantitatively assessed by introducing two indicators referred to as the integrity healing ratio (HR_C) and the damage healing ratio (HR_S).

Specific findings can be summarized as follows:

- Percent of healing recovery decreases as level of pseudostiffness loss imposed before loading removal increases, thus indicating the existence of some intrinsic irreversible damage, the relative amount of which increases with the increase of total damage.
- Polymer modification enhances the healing properties of base bitumen to an extent that depends upon polymer dosage.
- Healing ranking should be preferably based on the analysis of HR_S and in some cases shows that neat bitumens may outperform polymer-modified binders. Such an occurrence is related to the specific polymer-bitumen interactions which take place in the bituminous medium and needs to be directly assessed by experimental means.

In order to limit the experimental program to a reasonable extent, a single temperature and rest time were used in this study. Further research is needed to investigate the influence of temperature and duration of rest periods on the healing response of binders. In addition, application of the proposed approach to bituminous mastics also deserves consideration in future studies.

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