Linear viscoelastic properties of mastics: results from a new annular shearing rheometer, and modelling.

B. Delaporte
Département Génie Civil et Bâtiment (DGCB), TOTAL France, Lyon, France

H. Di Benedetto
Département Génie Civil et Bâtiment (DGCB), ENTPE, Lyon, France

C. Sauzéat
Département Génie Civil et Bâtiment (DGCB), ENTPE, Lyon, France

P. Chaverot
TOTAL France, Centre de Recherche de Solaize (CReS), Solaize, France

ABSTRACT: The mastic, composed of bitumen and filler, is an intermediate material between bitumen and mix. It can be considered as the real binder in the mix. Measuring its mechanical behaviour will contribute to closing a gap when linking binder and mixture properties. A new specifically designed annular shearing rheometer was developed at the DGCB laboratory (ENTPE) to study bituminous mastics. The complex modulus G^* of mastics is measured in a large range of temperatures (-25°C to 70°C) and frequencies (0.03 to 10 Hz). The samples tested are larger than those currently tested (hollow cylinder: external diameter = 105 mm, thickness = 5 mm, height = 40 mm). An experimental campaign on the linear viscoelastic behaviour of different bituminous mastics was carried out at low strain level. The aim was to determine the effect of i) the filler size, ii) the filler concentration, and iii) the grading of the filler, iv) the filler type on the complex modulus of the materials.
A part of the analyses of the complex modulus data is proposed. A significant difference in behaviour between mastics and bitumen is observed: a decrease of the phase angle (complex modulus) of mastics at high temperatures is probably linked to the existence of contacts between filler particles. A unique rheological model (2S2P1D) developed at DGCB is used to model the experimental results. A function of the filler concentration is considered to describe the continuous change in properties from bitumen to mastic.

KEY WORDS: Mastic, Viscoelasticity, Filler.

1 INTRODUCTION

Bitumen does not occur not alone in bituminous mixtures; it is mixed with mineral filler forming the mastic. It is the intermediate phase between bitumen and mixture. In this paper, a new annular shearing rheometer (ASR) is presented. Then, the investigated properties of mastics in the Linear Viscoelastic (LVE) domain are simulated using a model called 2S2P1D (2 springs, 2 parabolic elements, 1 dashpot) (Olard & Di Benedetto, 2003; Di Benedetto et al., 2004). A link between the global LVE behaviour of bitumen and the LVE behaviour of mastic is proposed. The results presented in this paper are a part of a more general project realized within the framework of a partnership between the company TOTAL and the “Département Génie Civil et Bâtiment” (DGCB) of the “Ecole Nationale des Travaux Publics de l’Etat” (ENTPE) in Vaulx-en-Velin, France.

The mastic can be considered as the real binder in the mixture. Therefore, it has an important role in the performance of bituminous mixtures. The mixing of mineral filler and bitumen has been studied by several authors. The potential for reinforcement in mineral filler-asphalt composites may depend on different parameters such as filler type, filler size, filler concentration, nature of asphalt (Anderson & Goetz, 1973; Chen & Peng, 1998). The conclusions of Harris and Stuart’s work suggested that this reinforcement was well correlated to the fractional voids (Rigden voids) (Harris & Stuart, 1995). Other works have been conducted to measure and predict properties of asphalt mastics with micromechanics-based models (Hadrzynski, 1995; Buttlar et al., 1999; Shashidhar & Shenoy, 2002).

The devices currently used to measure the LVE properties of bitumen or mastics (complex modulus G^* or E^*) present two main limitations:

- the size of the specimen: it is generally admitted that the smaller size of the sample must be lower than 4 to 8 times the maximum size of aggregate. For classical geometries, this condition is not verified in the study of mastics.
- the range of complex modulus which can be measured: classical devices only cover a small range of complex modulus values. Then, the compilation of the global master curve requires several samples having different geometries (for example traction compression at low temperatures and annular shearing at high temperatures, or Dynamic Shear Rheometer tests on samples having different radii).

The two drawbacks are no longer of concern with the new specifically designed annular shearing rheometer (ASR), which has been developed at the DGCB to study bituminous mastics. With the same sample, it allows the measurement of the linear viscoelastic properties of mastics in a large range of temperatures and frequencies. It covers a range of complex modulus over 7 decades (1 kPa to 10 GPa). The size of the samples is larger than those currently used for the bitumen tests. Consequently, the mastic samples are well representative, even if filler and sand until 1 mm size are added. A great advantage of this apparatus is to use a unique solicitation type (annular shearing) on a unique sample for the whole range of temperatures and frequencies, ensuring a continuous measurement.

A general research topic aimed at analyzing and modelling the influence of the type, the size, the gradation, and the concentration of filler on mastic behaviour, for different types of bitumen, has started as the ongoing PhD work of the first author. In this paper, only a part of the campaign is presented. First, a general presentation of the new annular shearing rheometer is proposed. The experimental campaign is then detailed. Finally results and analyses of the data are presented. To simulate the LVE behaviour of the different mastics, the 2S2P1D model which derivates from the Huet & Sayegh model (Sayegh, 1965) is used. This model was found to fit quite well the experimental measurements in the present study.

2 THE NEW ANNULAR SHEARING RHEOMETER (ASR)

2.1 General presentation of the ASR

The principle of the annular shearing rheometer (ASR) consists in measuring sinusoidal shear stress and sinusoidal shear strain (distortion) applied on a hollow cylinder of bitumen or mastic, at different temperatures and frequencies. The complex modulus of the material can be obtained from the data. The hollow cylinder has a rather large size: 5 mm thickness, 105 mm outer diameter and 40 mm height. With these geometrical features, the test is homogenous as a first approximation, even with aggregates up to 1 millimetre. A schematic view of the apparatus is presented in Figure 1.

A sinusoidal cyclic excitation is applied in stress or strain mode using the control system of a 50 kN capacity press (hydraulic press) on which the ASR is fixed. Strain is measured by three displacement transducers placed at 120° around the sample. The control strain is taken as the mean value of the three measurements. The transducers measure the displacement between outer lateral surface and inner lateral surface of the bituminous hollow cylinder. The range of the amplitude of the cycles is from 1 µm to 250 µm. In terms of distortion γ, it corresponds to amplitudes of 2.10^{-4} m/m to 5.10^{-2} m/m. The inner surface of the sample adheres to the aluminium core, which is linked to the load cell. The outer surface sticks to the aluminium hollow cylinder linked to the mobile piston of the press.

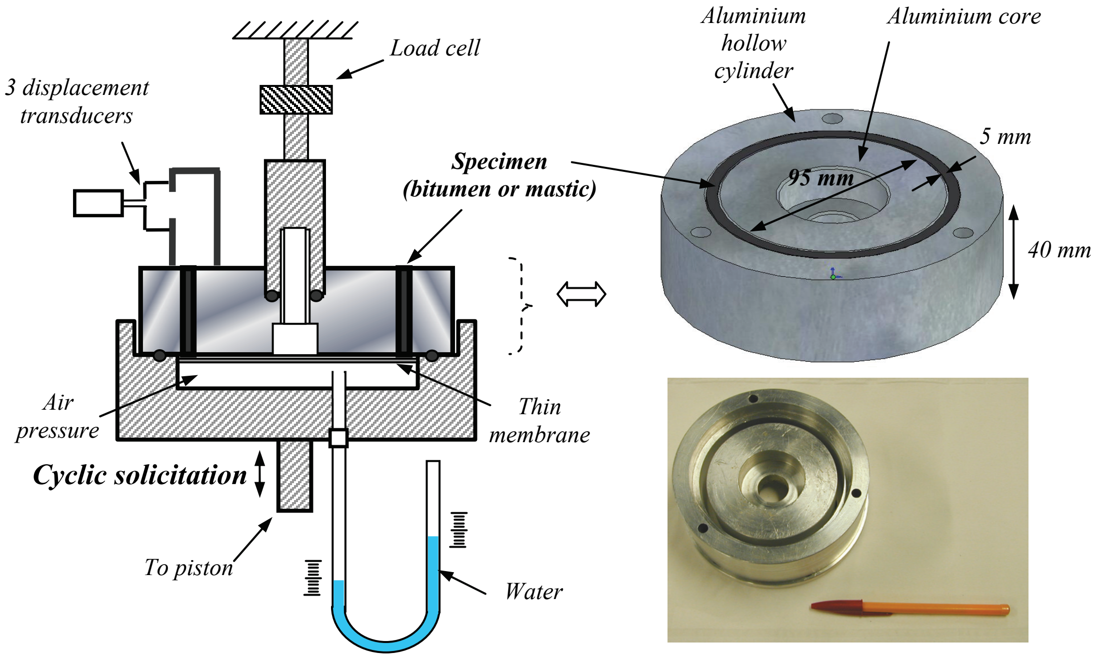


Figure 1. Schematic of the new annular shearing rheometer for bitumen and mastic

Complex modulus G^* ($|G^*|$ and phase angle ϕ) is written: $G^* = |G^*|\cdot e^{i\phi} = G_1 + \boldsymbol{i}G_2$. It is measured at 10 to 12 temperatures from about –25°C to 80°C and 6 frequencies from 0,03 Hz to 10 Hz. It contains elastic and viscous components, which are respectively designated as the storage modulus (G_1) and the loss modulus (G_2). $|G^*|$ is the ratio between the amplitude of distortion γ_0 with $\gamma(t) = \gamma_0 \cdot \sin(\omega t - \varphi)$, and the amplitude of shear stress τ_0 with $\tau(t) = \tau_0 \cdot \sin(\omega t)$. Phase angle φ is calculated by measuring the phase difference between shear stress and shear strain signals.

2.2 Load measurements

In order to cover the largest range of load (stress), a high sensitivity load cell (200 N) is used to measure the low complex values (at high temperatures). It can measure load cycles with amplitude as low as 1 N, which means a complex modulus close to 1 kPa for a distortion of 1%. This cell is placed in series with the main load cell of the press (50 kN). A by-pass system ensures a transition between the load cell of the press and the accurate load cell. When the tested material has a high modulus, the small load cell is disconnected as it is shown in Figure 2. The small load cell is capable of withstanding loads up to 2000 N to insure a factor of safety against physical damage.

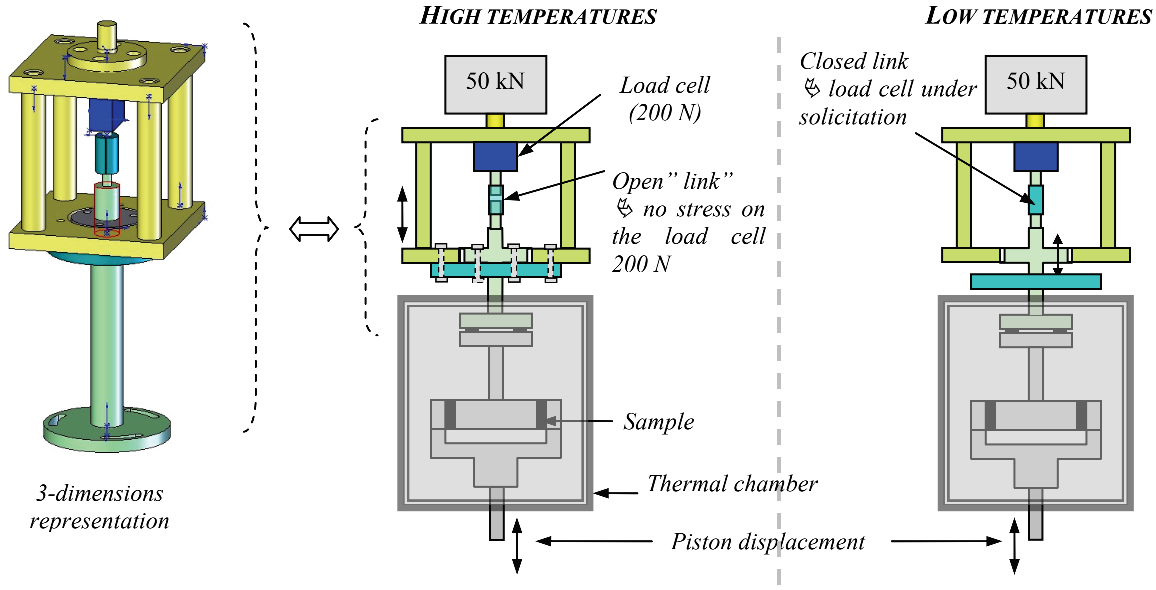


Figure 2. Schematic view of the by-pass system to make transition between low temperature measurements and high temperature measurements

2.3 High temperatures

At high temperatures (about 30 to 80°C), a specific procedure is used to prevent bituminous materials from creeping. Air pressure is applied under the specimen. The air pressure is obtained by a difference of water level in a U-shape tube (Figure 1). This pressure is applied through the medium of a thin membrane which is placed against the lower surface of the sample. The membrane has a quasi null stiffness. During the test, pressure is released to avoid air and water effects.

When the material stiffness is very low ($|G^*|$ less than 10 kPa), the rigidity of the displacement transducers is not negligible anymore in comparison with the one of the mastic. A specific test without material proves that this elastic rigidity was close to 150 Pa. Then, this value is systematically removed from the storage modulus of the tested material.

2.4 High filler content mastics

When filler volume concentration is high (over 50%), consistency is too high for the mastic to be moulded between the aluminium cylinders. A specific apparatus is designed to make the sample. The operation principle is presented in Figure 3.

After mixing bitumen and filler, the mastic is spread in the main support (Figure 3a). The aluminium cylinder is first fixed to this support. Then, the core is forced down by a nut and bolt system (Figure 3b). The mastic is squeezed up into the mould, until it is filled up (Figure 3c and Figure 3d).

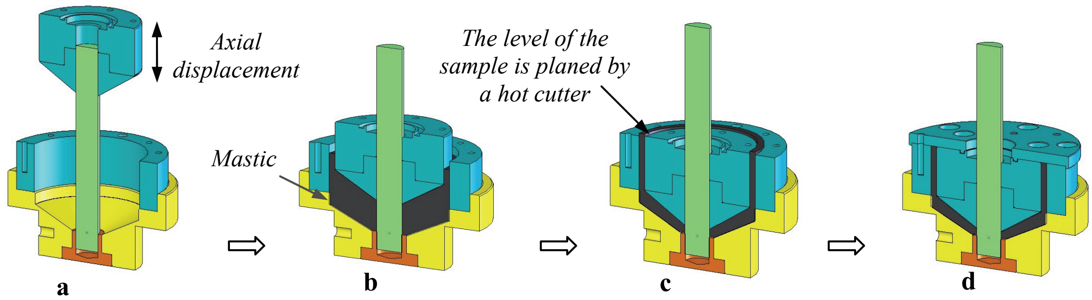


Figure 3. Schematic of moulding of high filler content mastics

3 EXPERIMENTAL CAMPAIGN

A 50/70 penetration grade bitumen has been used for the different formulations of mastic. Three different gradation types obtained from a unique reference limestone filler have been selected to be investigated in the mastics (cf. Figure 4):

- filler called "spread D_{max}" (or SDMX). It is a 0–100 µm grading, considered as a well-graded filler. The coefficient of uniformity defined by $C_U = D_{60} / D_{10}$ is approximately 5.6. Five volume filler concentrations (ϕ_f) have been selected: ϕ_f = 30, 40, 50, 55 and 60%. The volume concentration is the ratio of the filler volume and the mastic volume ($\phi = V_f/V_m$).
- filler sieved between 63 and 100 µm, called "centered D_{max}" formulation (CDMX): it is a 63 – 100 µm grading, called uniformly graded filler, with a low coefficient of uniformity C_U = 1.8. Four formulations have been used : ϕ_f = 30, 40, 50 and 55 %
- filler sieved at 15 µm called "intermediate" diameter D_i (DI). It is a 0 – 15 µm grading (well-graded filler, C_U = 5,1). Three concentrations have been tested: 30, 40 and 50%.

In this study, filler concentrations have been taken above 30%, because this value is close to the lowest concentration found in typical pavement mixes.

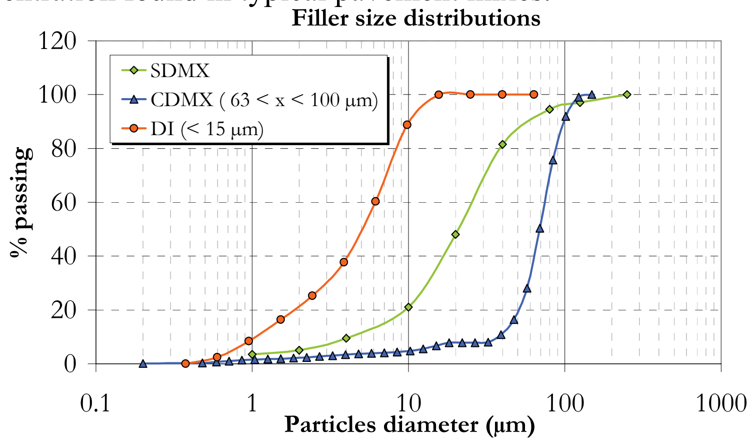


Figure 4. Particle size distribution of the different filler types

Thirteen materials have been tested during the experimental campaign. A mixture studied at the DGCB whose binder is bitumen 50/70 is also considered. Due to space limitations, only results from the uniformly graded filler (CDMX) are presented in this paper. Table 1 presents the tested materials: 4 mastics formulated with filler CDMX at different concentrations, the 50/70 bitumen and the mixture (French classification BBSG 0/10):

Table 1. Tested materials: 4 mastics from filler CDMX, bitumen 50/70 and mixture

Test name	Formulation	aggregate content (vol.)
B5070	Pure binder	0
M5070CDMX30	Mastic with filler CDMX	30% (filler)
M5070CDMX30	Mastic with filler CDMX	40% (filler)
M5070CDMX30	Mastic with filler CDMX	50% (filler)
M5070CDMX30	Mastic with filler CDMX	55% (filler)
50/70 MIX	Hot Mix Asphalt (HMA)	86%

All the materials presented in this study exhibited a thermorheologically simple behaviour: the data (ϕ, $|G^*|$) of each material have a unique curve in Black space. The complex modulus can be represented by graphs of the modulus $|G^*|$ and the phase angle versus the frequency on logarithmic scale. The shift factor $a_T(T)$ used for the construction of the master curve (cf.

Figure 5) is a function of the temperature. Its evolution can be simulated by the Williams, Landel and Ferry (WLF) equations:

$$\log(a_T) = -\frac{C_1 \cdot (T - T_r)}{C_2 + T - T_r} \qquad \text{Eq. 1}$$

where C_1, C_2 are WLF constants, and T_r the reference temperature. The reference temperature T_r is chosen equal to 10°C.

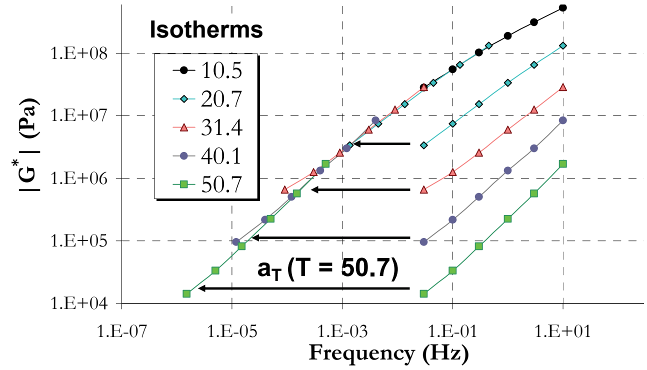


Figure 5. Construction of a master curve with a_T factor (temperature in °C).

4 RESULTS AND MODELING

4.1 Shift factor a_T

In previous works, it has been suggested that a_T binder and a_T corresponding mix could be considered as equal (Boutin et al., 1995; Di Benedetto & Des Croix, 1996; Di Benedetto et al., 2004). As would be expected for the mastics investigated during this experimental campaign, the translation coefficients a_T of the different tested mastics are very close (cf. Figure 6). As a consequence, the binder may completely define the shift factor values whatever the type and content of mineral aggregates.

The a_T data can be approached by the WLF law as presented in Figure 6. Coefficients C_1 and C_2 are equal respectively to 18.8 and 141.2.

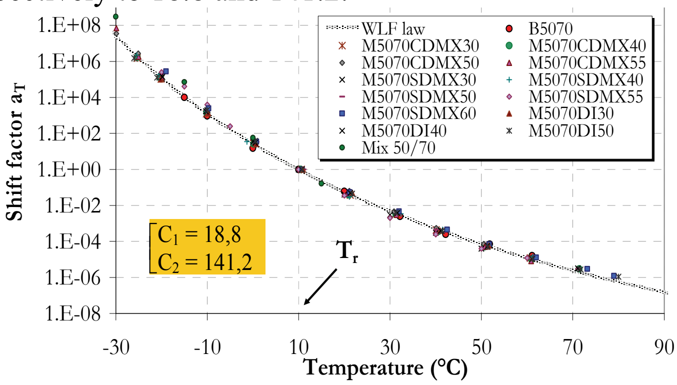


Figure 6. Shift factor a_T of all the tested materials formulated from the binder, at T_r = 10°C.

4.2 High temperatures

When temperature increases (or frequency decreases), bitumen tend to be a purely viscous material. The phase angle approaches 90° and the slope of the master curve approaches 1 (see Figure 7 and Figure 8). With regards to mastics, a significant change in behaviour can be deduced from the experimental curves at very low frequencies, compared to pure binders:

- the slope in the modulus master curve is decreasing at low frequencies,
- phase angle is decreasing at low frequencies (see Figure 8).

In this case, the mastic behaviour exhibits characteristics resembling the behaviour of a mixture. It is not purely viscous at high temperature. The physical meaning of this change of behaviour may be the existence of a solid contact between particles, when filler concentration is high. It is conveyed by an elastic modulus G_{filler}. Then, at very low frequencies, the modulus of the mastic tends towards $G_0 \neq 0$ and the phase angle tends towards 0.

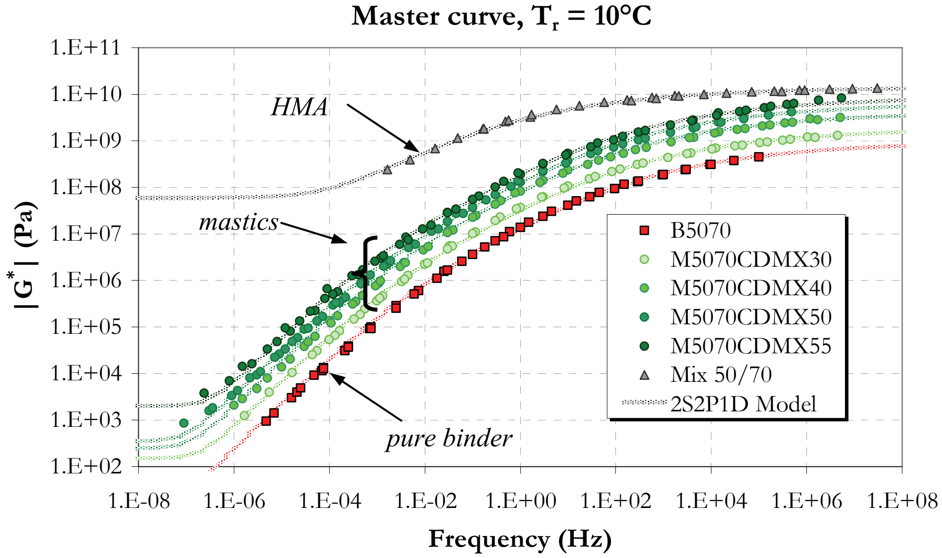


Figure 7. Modulus master curves (T_r = 10°C) of bitumen 50/70, 4 mastic formulations based on CDMX filler (30%, 40%, 50% and 55%), and Hot Mix Asphalt (HMA). Straight lines are simulation with the 2S2P1D model (cf. section 4.3)

Figure 8 shows that phase angle is the same for the 4 mastics and the binder on a large range of frequencies ($f_r > 10^{-4}$ Hz). Note that this result has also been validated for the 2 other gradations of filler. If we do not consider low frequencies ($<10^{-4}$Hz) where filler concentration has a large influence, this result shows that the binder completely defines the phase angle of the mastic. In addition, it can be observed that the difference in the phase angle curves between mastics and HMA is very high, even when high filler concentrations are considered.

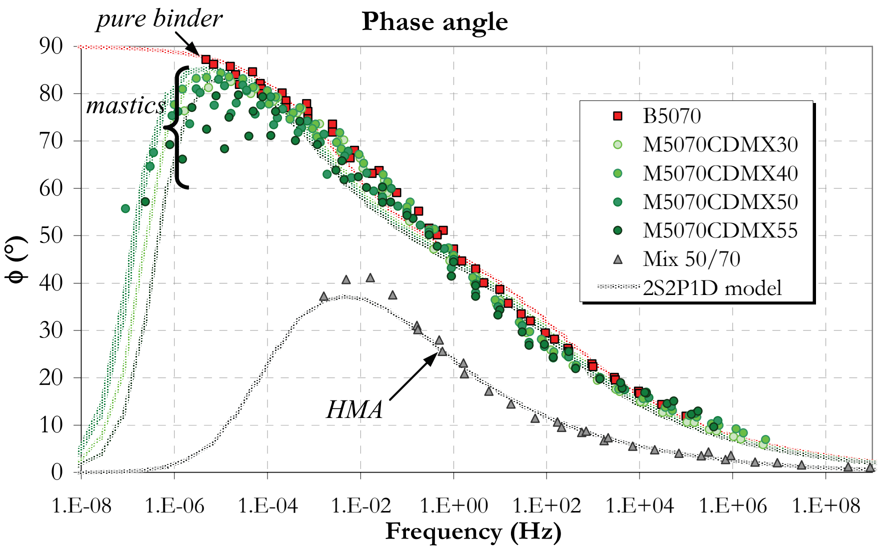


Figure 8. Phase angle master curves of bitumen 50/70, 4 mastic formulations based on CDMX filler (30%, 40%, 50% and 55%), and Hot Mix Asphalt (HMA). 2S2P1D Model.

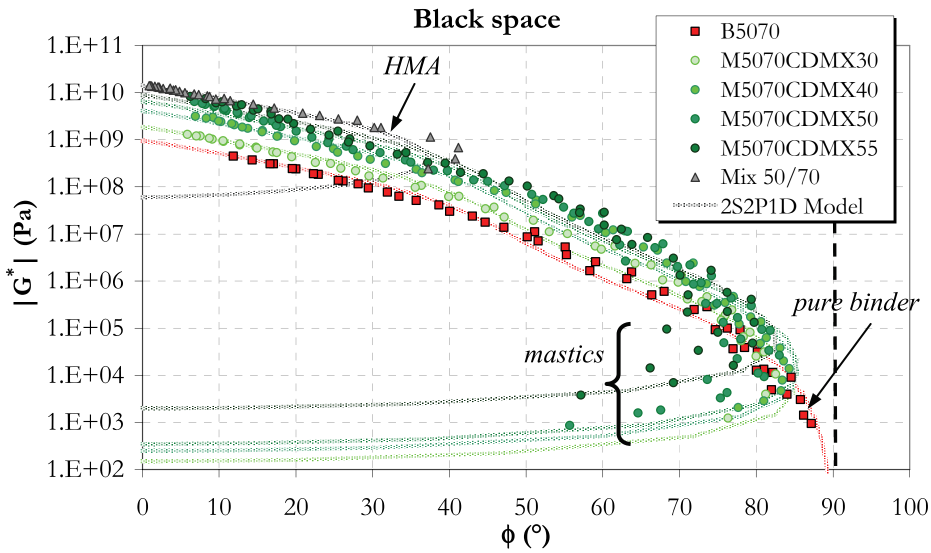


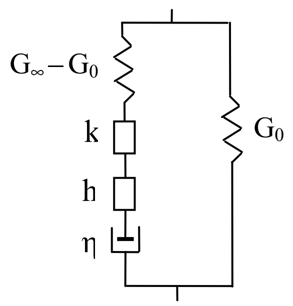
Figure 9. Complex modulus in Black space of bitumen 50/70, 4 mastic formulations based on CDMX filler (30%, 40%, 50% and 55%), and Hot Mix Asphalt (HMA).

4.3 Modelling of mastic rheological behaviour: model 2S2P1D

The 2S2P1D model, developed at the DGCB, consists of a generalization of the Huet-Sayegh model (Sayegh, 1965). It is valid for any bituminous material: binder, mastic or mix (Di Benedetto et al., 2004). This model 2S2P1D (2 Springs, 2 Parabolic, 1 Dashpot) is based on a simple combination of physical elements (spring, dashpot and parabolic element). The Huet-Sayegh model has been adapted by adding a linear dashpot in series with the two parabolic elements and the spring of rigidity E_∞-E_0 (cf. Figure 10).

At a given temperature, the introduced 2S2P1D model has 7 constants and its complex modulus is given by the following expression:

$$G^*(\omega) = G_0 + \frac{G_\infty - G_0}{1 + \delta(\boldsymbol{i}\omega\tau)^{-k} + (\boldsymbol{i}\omega\tau)^{-h} + (\boldsymbol{i}\omega\beta\tau)^{-1}} \qquad \text{Eq. 2}$$



with $\boldsymbol{i}$: complex number defined by $\boldsymbol{i}^2 = -1$,
ω: the pulsation = $2\pi \cdot f_r$ (f_r is the frequency),
k, h: exponents such as $0 < k < h < 1$,
δ: constant,
G_0 the static modulus when $\omega \to 0$
G_∞ the glassy modulus when $\omega \to \infty$,
τ: characteristic time, function of the temperature
β: constant, defined so that: $\eta = (G_\infty - G_0) \cdot \beta \cdot \tau$
η: newtonian viscosity

Figure 10. Representation of the 2S2P1D model

At a given temperature, 7 constants are required to completely characterize the LVE properties of the considered material. τ evolution may be approximated by a WLF type law (Di Benedetto et al., 2004). If the Time-Temperature Superposition Principle (TTSP) holds, $\tau(T) = a_T(T) \cdot \tau_0$ where $a_T(T)$ is the shift factor at temperature T. Then $\tau_0 = \tau(T_r)$ is determined at the reference temperature $T_r = 10°C$. The number of constants amounts to 9 including the 2 WLF constants (C_1 and C_2 calculated at the reference temperature).

The simulations of the behaviour of bitumen, mastics with filler CDMX and mixture are plotted in Figure 7, Figure 8, and Figure 9. The model constants used to simulate the materials are reported in Table 2. Simulations of complex modulus with the 2SPS1D model fits quite well with the experimental points for $|G^*|$ and Φ.

Table 2. Parameters of the 2S2P1D model for the binder, the corresponding mastics composed of filler CDMX and the mixture.

Material	G_0 (Pa)	G_{inf} (Pa)	k	h	δ	$\tau_0 = \tau(10°C)$	β
B5070	0	9.50E+08	0.21	0.55	2.3	9.18E-05	450
M5070CDMX30	150	1.85E+09	0.21	0.55	2.3	1.40E-04	450
M5070CDMX40	250	4.10E+09	0.21	0.55	2.3	1.73E-04	450
M5070CDMX50	350	6.30E+09	0.21	0.55	2.3	1.83E-04	450
M5070CDMX55	2000	8.80E+09	0.21	0.55	2.3	2.18E-04	450
Mix 50/70	6.00E+07	1.40E+10	0.21	0.55	2.3	7.00E-02	450

The reference bitumen is the only material tending to a Newtonian behaviour at very low frequencies. Its elastic modulus G_0 is equal to zero and only 8 constants (with C_1 and C_2) of the model are not nil for the pure binder. Mastics have a non nil static modulus G_0. The physical meaning is probably the existence of solid contacts between filler particles (simulated by a spring). Therefore, G_0 is increasing with the number of contacts which is linked to the volume filler concentration. The change between the bitumen behaviour and the behaviour of mastic related to a matrix structured by filler (whose solid particles are in contact) having a G_0 not null happens at a threshold value of filler concentration below 30%.

Table 2 highlights that parameters δ, k, h and β of the model are the same for each material. They are completely determined by the pure bitumen, as C_1 and C_2. The only parameters varying between materials having different filler or aggregate content are the static modulus (G_0), the glassy modulus (G_∞), and τ_0. This observation is very important as only 3 constants of the model (G_0, G_∞ and τ_0) are needed to obtain the complete complex modulus (from very low frequencies to high frequencies, and from low temperatures to high temperatures) of any mastic or mixture, once the behaviour of the bitumen is known.

The parameter τ_0 of the mastics depends on the considered filler concentration. Figure 11 shows that the following equation can be obtained from our data: $\ln(\tau_{0mastic}/\tau_{0bitumen}) = 1.56\phi_f$. This equation allows predicting the parameter τ_0 of any mastic, if $\tau_{0binder}$ is known.

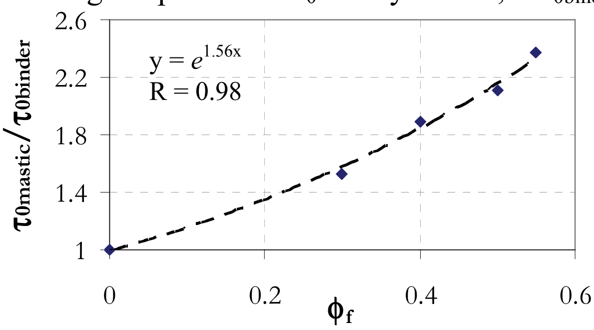


Figure 11. Evolution of the characteristic time of mastics as a function of the filler content

CONCLUSION

A new Annular Shearing Rheometer is presented in this paper. It can measure linear viscoelastic properties of bitumen and mastic on a large range of temperatures and frequencies using one single sample (over 7 decades for $|G^*|$). A part of a general experimental campaign on mechanical properties of mastics is presented. Some conclusions can be pointed out:

- as observed on asphalt mixtures, the shift factors of all the tested mastics have values very close to those of the binder.
- for filler content from 30% to 55%, the master curves ($|G^*|$) of the corresponding mastics display a gradual increase in complex modulus when filler content increases (Figure 7).
- for filler content from 30% to 55%, the master curves of phase angle of the corresponding mastics are identical below 70° (equivalent frequencies up to 10^{-3} Hz at 10°C) (Figure 8).
- for equivalent frequencies lower than 10^{-3} Hz at 10°C, a decrease of the phase angle is observed. This decrease is more important at high filler content (Figure 8). It is probably explained by the existence of contacts between filler particles.

In the prediction of mastic G^*, the 2S2P1D model (developed at the ENTPE) was found to compare favourably to experimental results to give good simulations. Another important finding is that, among the 9 constants needed by the model, 6 are given by the pure bitumen.

ACKNOWLEDGEMENT

The authors thank Bill Buttlar, Gilles Gauthier and Jean-Pascal Planche for their valuable comments in editing this paper.

REFERENCES

Anderson D. A. & Goetz W. H., 1973. *Mechanical Behavior and Reinforcement of Mineral Filler-Asphalt Mixtures*. Journal of the Association of Asphalt Paving Technologists, Vol. 42, 37 - 66.

Boutin C., De La Roche C., Di Benedetto H. & Ramond G., 1995. *De la rhéologie du liant à celle de l'enrobé bitumineux, Théorie de l'homogénéisation et validation expérimentale.* Eurobitume Workshop 1995.

Buttlar W. G., Diyar B., Al-Khateeb G. & Waldhoff A. S., 1999. *Understanding Aspahlt Mastic Behavior Through Micromechanics*. Transportation Research Record, 1681, 157 - 169.

Chen J.-S. & Peng C.-H., 1998. *Analyses of Tensile Failure Properties of Asphalt-Mineral Filler Mastics*. Journal of Materials in Civil Engineering, Vol. 10, 256-262.

Di Benedetto H. & Des Croix P., 1996. *Binder-mix rheology: limits of linear domain, non linear behaviour*. Eurasphalt & Eurobitume Congress 1996.

Di Benedetto H., Olard F., Sauzéat C. & Delaporte B., 2004. *Linear viscoelastic behaviour of bituminous materials: from binders to mixes*. Road Materials and Pavement Design.

Hadrzynski F., 1995. *Etude de la rhéologie d'un bitume modifié par des polymères et prévision du comportement mécanique des mélanges avec fines, sables et graviers. Université de Paris-Nord*.

Harris B. M. & Stuart K. D., 1995. *Analysis of Mineral Fillers and Mastics Used in Stone Matrix Asphalt*. J. Assoc. Paving Technol., Vol. 64, 54-95.

Olard F. & Di Benedetto H., 2003. *General "2S2P1D" model and relation between the linear viscoelastic behaviors of bituminous binders and mixes*. Road Materials and Pavement Design, Vol. 4.

Sayegh G., 1965. *Variation des modules de quelques bitumes purs et enrobés bitumineux.* Thèse de doctorat d'ingénieur, *Faculté des Sciences de l'Université de Paris*.

Shashidhar N. & Shenoy A., 2002. *On using micromechanical models to describe dynamic mechanical behavior of aspahlt mastics*. Mechanics of Materials, Vol. 34, 657 - 669.