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3 authors:

Gaspare Giancontieri
Università degli Studi di Palermo


David Hargreaves
University of Nottingham


Davide Lo Presti
Università degli Studi di Palermo

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Gaspare Giancontieri[a], David Hargreaves[b] and Davide Lo Presti[a,b*]

[a]*Faculty of Civil Engineering, University of Palermo, Palermo, Italy;* [b]*Nottingham Transportation Engineering Centre, University of Nottingham, Nottingham, UK*

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Modified bituminous binders allow asphalt technologists to design asphalt mixtures with superior performance. However, several recent studies highlighted that due to the complexity of these material, their characterisation can be challenging since common procedures used to characterise neat bitumen might not be adequate. For instance, during high temperature rotational viscosity testing of recycled tyre rubber modified binders (RTR-MB), a number of changes may occur to the sample leading to the here-defined sample stability which in turn provides misleading results. In this study the authors want to first provide a deeper understanding of this phenomenon by a numerical analysis using a bespoke Computational Fluid Dynamics (CFD) model to simulate the laboratory tests and use innovative visual aids to monitor the sample stability of heterogeneous bituminous binders during the rotational test. The numerical analysis was complemented by a laboratory campaign aiming at proving the occurring of sample stability during viscosity measurement of heterogeneous bituminous binders with a standard testing setup (SC-27). Furthermore, a dual helical ribbon (DHR) is here introduced as a solution to overcome the issue. Hence, laboratory tests were undertaken also with DHR and differences in viscosity measurements of neat bitumen, SBS-MB and RTR-MB were recorded. Results of this combined numerical and empirical approach proved that the standard setup for rotational viscosity measurements seems not be adequate for RTR-MB and depending on the level of modification and test temperatures, might not be best suited for SBS-MB either. The DHR seems to solve the issue and authors strongly recommend the adoption of this testing geometry to obtain more realistic high-temperature viscosity measurement of heterogeneous bituminous binders.

**Keywords:** sample stability; dual helical ribbon; modified bitumen; CFD; recycled tyre rubber; rheology

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Corresponding author:

E-mail addresses Dr Davide Lo Presti davide.lopresti@unipa.it

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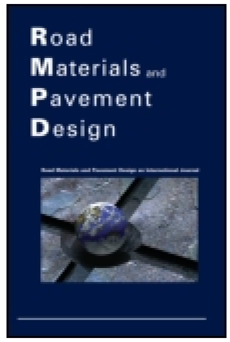


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Gaspare Giancontieri[a], David Hargreaves[b] and Davide Lo Presti[a,b*]

[a] *Faculty of Civil Engineering, University of Palermo, Palermo, Italy;*

[b] *Nottingham Transportation Engineering Centre, University of Nottingham, Nottingham NG7 2RD, UK*

Contacts:

Gaspare Giancontieri: gaspare.giancontieri@unipa.it

Dr David Hargreaves: david.hargreaves@nottingham.ac.uk

*Dr Davide Lo Presti (corresponding author): davide.lopresti@nottingham.ac.uk

# Are we correctly measuring the rotational viscosity of heterogeneous bituminous binders?

Modified bituminous binders allow asphalt technologists to design asphalt mixtures with superior performance. However, several recent studies highlighted that due to the complexity of these material their characterisation can be challenging since common procedures used to characterise neat bitumen might not be adequate. For instance, during high temperature rotational viscosity testing of recycled tyre rubber modified binders (RTR-MB), a number of changes may occur to the sample leading to the here-defined sample stability which in turn provides misleading results. In this study the authors want to first provide a deeper understanding of this phenomena by a numerical analysis using a bespoke Computational Fluid Dynamics (CFD) model to simulate the laboratory tests and use innovative visual aids to monitor the sample stability of heterogeneous bituminous binders during the rotational test. The numerical analysis was complemented by a laboratory campaign aiming at proving the occurring of sample stability during viscosity measurement of heterogeneous bituminous binders with a standard testing setup (SC-27). Furthermore, a dual helical ribbon (DHR) is here introduced as a solution to overcome the issue. Hence, laboratory tests were undertaken also with DHR and differences in viscosity measurements of neat bitumen, SBS-MB and RTR-MB were recorded. Results of this combined numerical and empirical approach proved that the standard setup for rotational viscosity measurements seems not be adequate for RTR-MB and depending on the level of modification and test temperatures, might not be best suited for SBS-MB either. The DHR seems to solve the issue and authors strongly recommend the adoption of this testing geometry to obtain more realistic high-temperature viscosity measurement of heterogeneous bituminous binders.

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## 1. Introduction

The experimental characterization of bituminous material's rheological properties is of fundamental importance during their design as well as for predicting the asphalt mixtures performance. In order to assess whether adequate coating of aggregate occurs, as well as for ensuring pumpability and compactability of the mixtures, bitumen technologists are used to monitoring the high-temperature viscosity of bituminous binders (in the range 100–200°C) during product development and quality control (Caltrans, 2011). In a laboratory, usually rotational viscometers with coaxial cylinder configuration are used for this exercise. This testing setup consists of a tube into which the test fluid is poured, and a spindle is inserted and then rotated at a selected rotational speed. The instrument will convert the value of torque applied into a viscosity reading. This setup was specifically designed for homogeneous systems (i.e. bitumen), however nowadays is used for any type of bituminous binders, no matters the complexity. In fact, the use of polymer modified bitumen is widely increased in pavement engineering. These binders are often able to improve the performance of asphalt mixtures (e.g. resistance to cracking, noise reduction etc.), (Huang, 2006; Peralta *et al.*, 2010; Visser *et al.*, 2017; Zhang *et al.*, 2018) nevertheless, their rheological characterisation can be challenging due to complex structure of such materials. In fact, deviation from Newtonian behaviour, including shear rate dependent viscosity, elasticity and time-dependent rheological behavior can provide misleading viscosity measurements (Celauro *et al.*, 2012). The issue here is that some modified types of bitumen are multi-phase system that can present internal structure behaving differently from the bitumen matrix. This complexity sometime is highlighted by a lack of compatibility and in that case during hot storage the modifier could separate, either top or bottom of the container. This is a practical issue that bitumen technologists have to cope within asphalt plants, however if the difference between densities of modifier and bituminous matrix is significant, the same issue might be encountered also during laboratory measurements. This lack of sample stability may produce significantly inconsistent data.

These types of issues are encountered in other fields and for many other materials such as chocolate, plastic, rubber, ceramic, food, cosmetic, detergent, paints, glazing, lubricants, inks, adhesives, sealants, (Dow, 1946; Lin *et al.*, 1985; Gallegos *et al.*, 1999; Tabilo-Munizaga *et al.*, 2005; Malkin, 2009). For these reasons the rotational viscometers are equipped with supplementary impellers designed to help rheologist for some of these scenarios. For instance, a vane spindle allows performing measurements with paste-like materials, gels, and fluids where suspended solids migrate away from the measurement surface of standard spindles. Furthermore, the Brookfield Helipath Stand is designed to slowly lower or raise a T-bar spindle so that it describes a helical path through the test sample. These accessories are all fit for purpose although they are not able to guarantee the sample stability of complex systems with high level of heterogeneity such as Recycled Tyre Rubber Modified Bitumen (RTR-MB) or other materials having the tendency to stratify during rotational tests due to phase density differences (Giancontieri *et al.*, 2018).

In a first attempt to address this issue, (Lo Presti *et al.*, 2014) successfully designed, manufactured and tested a prototype of a Dual Helical Impeller (DHI) for Brookfield Rotational Viscometers. Experimental studies were carried out to evaluate whether the DHI was able to improve the degree of homogenisation of high viscous fluids, in turns producing more realistic viscosity measurements. In comparison to the Brookfield standard cylindrical geometry (spindle SC-27), the DHI always predicted a different "apparent" viscosity. This result was explained by the capability of the DHI to create what have been likened to convective-like flows as opposed to the axisymmetric swirling flow induced by the standard SC-27 spindle. In a further study, (Lo Presti *et al.*, 2017), the authors developed a bespoke numerical model able to reproduce the previous experimental campaign aimed to provide an insight into the flow field created by the DHI.

Results proved that the modified impeller improved the overall grade of homogenisation within the sample, but also showed that a further optimisation of the DHI was possible. In fact, especially for the case of Recycled Tyre Rubber Modified Bitumen (RTR-MB), which can be considered as a heterogeneous blend of fluid and particles with different densities and size, testing the sample with an adequate thickness (gap size) is of primary importance to avoid misleading results. Rotational viscometers are equipped with spindles of different diameters and while this helps obtaining an adequate sample thickness, they still do not ensure sample stability. The DHI allowed a good mixing of the volume fractions but due to the tiny gap between the inner and outer helix, it can suffer of clogging especially when RTR-MB is used.

Considering the issues mentioned above, Giancontieri *et al.* (2018) carried out a shape optimisation of the DHI. Therefore, a testing configuration that improves the coaxial cylinders setup with a Dual Helical Ribbon (DHR) as impeller was developed. **Error! Reference source not found.** shows the DHR adapted for a rotational viscometer. The DHR was designed, manufactured and then validated using the Otto-Metzner method testing a wide range of non-Newtonian fluids. The optimised shape was designed to avoid issues of clogging as well as guaranteeing sample stability by creating an upward pumping within the inner region of the blades and allowing a downward stream on the peripheral side of the container. This leads to an effective convective flow of the fluid within the chamber with no risk of clogging and with improved dispersions of the particle.

To provide the reader with further information for the interpretation of the presented results and conclusions, the following sections will provide a background on measuring viscosity by means of non-standard equipment and a brief review on the use of CFD for modelling mixing of multi-phase fluids.

### *1.1 Background – Measuring viscosity using non-standard equipment*

For rheological measurements by means of rotational viscometer with coaxial cylinders, there is a theoretical basis for the calculation of the viscosity. Consider a cylinder of radius Rs, length, L, rotating at an angular velocity, ω, inside a stationary cylinder of radius, Rc (Mezger, T.G., 2006). The shear rate at the surface of the spindle,$\dot{\gamma}$ , is

$$\dot{\gamma} = \frac{2\omega R_c^2}{R_c^2 - R_s^2} \quad (1)$$

and the shear stress, $\tau$, is

$$\tau = \frac{T}{2\tau R_s^2 L} \quad (2)$$

where $T$ is the torque input by the machine. The viscosity, $\mu$ is

$$\mu = \frac{\tau}{\dot{\gamma}} = \frac{T(R_C^2 - R_S^2)}{4\pi\omega R_C^2 R_S^2 L} \quad (3)$$

So, since all the variables on the right-hand-side of Equation 3 are measurable, the viscosity can easily be determined. Now, when a non-cylindrical impeller is used in the viscometer, there are no tractable equations for the shear rate or shear stress akin to Equations 1 and 2. This presents a challenge since, while the impellers may be good at mixing, there is no simple way of deriving the viscosity of the mixture. The only quantities that can be measured directly are the torque, $T$, and the angular velocity, $\omega$. In this case, the only way to proceed is to use the impeller design in a range of single and multi-phase fluids of known viscosity, which have been measured with the standard spindle viscometer. Then, the viscosity becomes a function of these two measurable variables

$$\mu = \mu(T,\omega) \quad (4)$$

There is no information about the shear rate or the shear stress that can be derived because the complex 3D nature of the flow precludes that. After testing a wide range of fluids of different viscosities, a surface fit could be made and then for a given $T$ and $\omega$, the viscosity of an unknown fluid could be calculated. The situation is further complicated in that the calibration procedure would then have to be done for each class of fluid (Newtonian, shear thinning, shear thickening, etc.) and a judgement would have to be made by the user as to which class of fluid they had in the viscometer.

A different approach is that promoted by Brookfield and utilized, for example, in (Celauro *et al.*, 2012). Each different spindle geometry provided by Brookfield for use in their viscometers is ascribed a Spindle Multiplier (SMC) value. For example, the SC-27 has an SMC of 25. Further, each viscometer model, here we are using the LVDV-E, has a Torque Constant, $TK$, which allows the apparent viscosity to be found from

$$\mu = \frac{0.1}{N} \cdot TK \cdot SMC \cdot T\% \tag{5}$$

where $N$ is the rotational speed (rpm) and $T\%$ is percentage of the maximum spring torque that the viscometer can deliver. In addition, the Shear Rate Constant (SRC) is defined as

$$\dot{\gamma} = SRC \cdot N = SRC \cdot \left(\frac{60}{2\pi}\right)\omega \tag{6}$$

For a design such as a DHI, this is a simplification but one which can produce values of apparent viscosity that are of practical use. Lo Presti (2014) attempted, with some success, to match the behaviour of the DHI to the range of SC-XX spindles offered by Brookfield. The SC-XX range are of the cylindrical type and each has unique SRC and SMC values. The Brookfield viscometer allows only the selection of a spindle code, which has associated SRC and SMC values, thus limiting fine adjustment to a discrete set of values. The authors were nevertheless able to find a spindle code that matched closely the DHI and did this by testing the DHI against a number of standard liquids of known viscosity. The SC4-28 spindle was found to most closely match the DHI with values of

SRC of 0.28 and SMC of 50. The validity of this approach is open to question because of the radically differing spindle geometries of the SC4-28 and DHI.

Thus, computational fluid dynamics was seen as an alternative method of obtaining values of SRC and SMC for the DHI spindle. For those reasons, another approach was used by Giancontieri *et al.* (2018) to calibrate the DHR geometry. The calibration procedure involved the use of the Metzer-Otto method for non-Newtonian fluids (Metzner *et al.*, 1957) that introduces the concept of apparent viscosity, $\eta_{app}$, which is linked to an effective shear rate $\dot{\gamma}_{\text{eff}}$. They suggested this effective shear rate to be proportional to the rotation frequency $N$:

$$\dot{\gamma}_{eff} = K_s N \tag{8}$$

Where the $K_s$ is a function of the geometry and the flow index. The idea behind the calibration procedure was to find a correlation between the $K_s$ and the flow index $n$.

Metzer-Otto also stated that:

$$\frac{\boldsymbol{\eta_{nN}}}{\boldsymbol{\mu N}} = \frac{\boldsymbol{M_{nN}}}{\boldsymbol{M_N}} \tag{9}$$

Where the $\eta_{nN}/\mu N$ is the ratio between the viscosity of a non-Newtonian and a Newtonian one calculated by means of dynamic shear rheometer. $M_{nN}/M_N$ is the ratio between the torque of a non-Newtonian fluid and a Newtonian calculated by means of rotational viscometer. From eq. 9 is easy to calculate the non-Newtonian viscosity:

$$\eta_{nN} = \mu N \left(\frac{M_{nN}}{M_N}\right) \tag{10}$$

From eq 10 it was possible to calculate the apparent shear rate of the non-Newtonian fluid using the Ostwald–de Waele power law relationship:

$$\dot{\boldsymbol{\gamma}}_{\boldsymbol{eff}} = \left(\frac{\boldsymbol{\eta_{nN}}}{\boldsymbol{k}}\right)^{\frac{\boldsymbol{1}}{\boldsymbol{n-1}}} \tag{11}$$

where *n* and *k* are input parameters, *k* is a measure of the average viscosity of the fluid (the consistency index); n is a measure of the deviation of the fluid from Newtonian (the power-law index). The value of n determines the class of the fluid:

- n = 1 → Newtonian fluid
- n > 1 → shear-thickening (dilatant fluids)
- n < 1 → shear-thinning (pseudo-plastics)

For those reasons, firstly an experimental campaign using a parallel-plate system aimed to find out the *n* and the consistency index *k* was carried out. Then, the DHR was adapted to the rotational viscometer and for each material the torque as a function of the rotational speed (0 to 100 rpm at 25 °C for approximately 45minutes) was measured. Finally, coupling the results of the parallel-plate and the rotational viscometer, $K_s$ was calculated for each material using the eq 8. Moreover, combining Equations 5 and 10 it was possible identify the unique constant for the DHR when used with the rotational viscometer.

### *1.2 Background – Mixing optimization and using CFD simulation of mixing processes*

Before the development of numerical simulations, mixers were normally evaluated experimentally using various measures: power consumption (Takahashi et al. 1982; Takahashi, Yokota, and Konno 1979, 1984; Cordobes, Brito de la fuente, and Gallegos 1998) mixing time and circulation time ( Smith 1970; Carreau, Patterson, and Yap 1976). However, none of these methods gives an understanding of the spatial variation of the phases or the nature of the transport processes, making the understanding and hence the efficient optimisation of the mixer designs difficult. Numerical simulations by means of

Computational Fluid Dynamics (CFD) offer a greater flexibility in analysing and visualizing the mixing. In both the previous (Lo Presti, Giancontieri, and Hargreaves 2017) and the present work the authors performed several simulations producing basic scenario which provide a lower level of accuracy but guarantee conceptual information (flow fields and volume fractions) as well as performing long-lasting and more accurate simulations providing absolute values of torque, which in turns provided the computational viscosity values.

In recent years, the simulation of mixing vessels is widely used to optimize mixer geometries and get better insights of the complex flow patterns generated by the impeller-vessel wall interaction (Delaplace *et al.*, 2000; Paul *et al.*, 2004). From the perspective of numerical analysis, one of the pioneering works focused on the mixing performance of helical ribbon impellers in cylindrical vessels is the contribution made by Tanguy (Tanguy *et al.*, 1992). They developed a three-dimensional model which was validated experimentally, based on the finite-element method for the analysis of a Helical Ribbon-screw impeller. The authors reported good liquid circulation at low impeller speeds (10 rpm) and showed evidence of poor pumping in the vessel bottom. They noticed that the segregation increased upon increasing the impeller speed. Subsequently, numerical works were developed for several helical ribbon geometries and fluids (Bertrand *et al.*, 1999). The numerical modeling of mixing in a stirred tank has attracted a great deal of attention and a recent review of the state-of-the-art in CFD simulations of stirred vessels can be found in Giancontieri et al (2018) and Sommerfeld (2004). The rapid development of numerical techniques and computational power has unleashed the possibilities of computational Fluids Dynamics (CFD) in this area. CFD is now an important tool for understanding the mixing in the stirred tanks (Um *et al.*, 2008). Lo Presti et al. (2017) introduced the use of CFD model to reproducing an experimental campaign of viscosity measurements of complex bituminous binders. Results proved that CFD is a tool able to provide insights into the measuring chamber that otherwise it would be impossible to obtain. Modelling of the complex flow in the presence of a rotating impeller is a computational challenge because of the complex geometry of rotating impeller and the nature of the flow in stirred tanks. Nevertheless, the developed CFD model, regardless of the testing geometries, speed and type of material tested, was able to provide values of torque that practically matched those obtained from the laboratory investigation.

Although CFD codes have made remarkable steps towards the solution of several engineering problems over the last decade, it still remains a difficult task to use such codes to help the design and the analysis of multi-phase systems.

### *1.3 Aim of the study*

This study aims to provide evidence that the standard coaxial cylinders setup for rotational viscosity measurements of bituminous binders might not be adequate for modified binders, particularly when dealing with high level of heterogeneity. In fact, when using standard rheological measurements for such complex systems, a number of changes, such as phase separation, sedimentation and agglomeration, may occur in the measuring chamber. These changes all contribute to undermine the here-defined sample stability that if altered could provide significantly misleading results.

Instability of the modified bitumens at the usual temperature for rotational tests (135 °C), can be minimal or none. However, this investigation wants to prove that if equilibrium times are considered, rather than high-level of heterogeneity of the sample, higher testing temperatures and high spindle speeds, the sample stability is compromised also in these conditions. This could lead to significant mistakes in further steps such as choosing wrong temperatures to guarantee appropriate aggregate coating during mixing and/or ensuring good workability during compaction of asphalt concrete.

This issue is particularly relevant within design of rubberised binders by means of low-shear blending protocol that sees the adaptation of the rotational viscometer as a mixing device offering a continuous monitoring of the viscosity (Celauro et al., 2012; Subhy, et al. 2015). In fact, in this scenario there is a contingency of high rotational speed and high-level of heterogeneity due to the high temperature where the bitumen viscosity is quite low (180 °C) and rubber particles are not swollen yet and tend to agglomerate in layers, mainly at the bottom of the tube.

In order to avoid both the above-presented issues, this study also presents an innovative testing configuration that replaces the coaxial cylinders setup with a Dual Helical Ribbon (DHR) as impeller. The investigation will offer a comparison of experimental and numerical results to highlight whether the DHR helps ensuring sample stability during rotational test of complex bituminous binders.

## 2. Methodology

The experimental programme showed in **Error! Reference source not found.** was tailored to allow a comparison between the DHR and the SC-27 systems by means of a combined laboratory-based and computational approach. The laboratory work will provide results of rotational viscosity tests on neat bitumen, an SBS-MB and an RTR-MB. The numerical simulations will provide a qualitative insight into the measuring chamber, by means of velocity fields and distribution of volume fractions within the container. The results of this combined approach will be used to understand whether lack of sample stability occurs and will also allow clarifying whether using the DHR ensures it as well the improving of rheological measurements.

### *2.1 Laboratory experimental campaign*

Three different binders were included in the experimental programme: a neat bitumen and two highly modified bitumen with different level and type of modifier (**Error! Reference source not found.**). The RTR-MB was produced by adding 15% of crumb rubber size 0,300mm in 35/50 grade bitumen, while the PMB by adding 7.5 of SBS 30% rad in 180/240 grade bitumen. All fluids being tested at 5, 20 and 100 rpm in a range of temperature between 120 to 200 °C. Due to the expected high torque for the lower range of temperatures, which would be close to the viscometer's limits, two Brookfield DV-II PRO Digital Viscometer were used: a low torque (LV model) and high torque (HA model). The two equipment differ mainly for the viscosity measurements range. The LV model is designed for low viscosity fluids (100% torque = 673.7 dyne cm), while the HA model is more appropriate for medium–high viscous fluid (100% torque = 14,374 dyne cm). Torque measurement accuracy specified by manufacturer is 1% of the full-scale range. Tests were conducted according to Subhy (2016), which are based on international standards on viscosity measurements of rubberized bitumens (ASTM D4402, ASTM D6114). The tests were undertaken for a total of about 45 minutes, where the first 15 min where allowed to reach the thermal equilibrium (0 rpm).

Then, impellers were quickly submerged in the blend and the viscometer was turned on to carry out the test, during which torque and angular velocity were measured and the viscosity calculated from Eq. (5). Tests were performed in coaxial cylinder setup with both the standard spindle SC-27 and the DHR as impellers. Each of the reported results is obtained as average of at least two replicates.

### *2.2 Numerical simulations by means of CFD*

In parallel with the laboratory experiments, a CFD study was conducted with the aim of offering a qualitative insight of the measuring chamber in order to assess whether lack of sample stability occurs and eventually assessing the reasons. Version 17 of ANSYS Fluent was used in this work.

### *2.2.1 Governing equations*

The modelling involves the solution of the Navier–Stokes equations. In order to simulate the behavior of the two-phase fluids in the present application, the mixture model was used. Here, the momentum and continuity equations are solved for the mixture, the volume fraction equations for the secondary phases, while algebraic expressions are used for the relative velocities and inter-phase drag.

First the mixture continuity equation:

$$\frac{\partial \rho_m}{\partial t} + \nabla\cdot(\rho_m \mathbf{u}_m) = 0 \quad (12)$$

where $\mathbf{u}_m$ is the velocity, $\rho$ is the density and the subscript $m$ indicates a quantity defined for the mixture. The momentum equation is:

$$\frac{\partial}{\partial t}(\rho_m \mathbf{u}_m) + \nabla(\rho_m \mathbf{u}_m \mathbf{u}_m) = \nabla p + \nabla\left[\eta_m\left(\nabla \mathbf{u}_m + \nabla \mathbf{u}_m^T\right)\right] + \rho_m \mathbf{g} + \nabla \left(\sum_{k=2}^{n} \alpha_k \rho_k \mathbf{u}_{dr,k} \mathbf{u}_{dr,k}\right) \quad (13)$$

where $\mathbf{g}$ is the gravitational acceleration and $\mathbf{u}_{dr,k}$ is the drift velocity for the secondary phase $k$.

$$u_{dr,k} = \mathbf{u}_k - \mathbf{u}_m \quad (14)$$

The mixture model in ANSYS FLUENT uses an algebraic slip formulation, which is based on the work of Yap (1979). It uses the slip velocity, which is related on the drift velocity defined in Eq. (14). The velocity of the $k^{th}$ phase is calculated from algebraic expressions, rather than a separate momentum equation. So, the final term on the right-hand side of Eq. (13) imparts a momentum source or sink to the mixture momentum equation, based on the relative motion of the primary and secondary phases.

### 2.3 CFD model setup

The geometry of the spindle (**Error! Reference source not found.**) was developed based on the Brookfield Sc-27 and realised with ANSYS DesignModeler. A mesh consisting of tetrahedra and triangular prisms (in the inflation layers adjacent to walls) was generated using ANSYS Meshing and the volume mesh associated with the surface mesh shown in figure contains 110,000 elements.

The DHR geometry was designed according to Giancontieri et al (2018). It allows creating a convective flow able to provide adequate mixing of complex blends also at low RPM. The idea is that the outer helix pumps the fluid downwards while the inner helix pumps it upwards. Based on the prototype design, DesignModeler and Meshing software were used to produce a mesh, which, due to the increased complexity, contained approximately 150,000 tetrahedral and triangular prism cells. The CAD model and associated surface mesh can be seen in figure 3. The Viscous Model was selected in Fluent, it allows to set parameters for inviscid, laminar, and turbulent flow. It is accepted practice in stirred or mixing vessels to use the mixing Reynolds number, $Re_m$ to characterise the flow:

$$Re_m = \frac{\rho D^2 N}{\mu} \tag{15}$$

where $D$ is the diameter of the impeller and $N$ is the number of revolutions per second. For $Re_m$ >10000, the flow is assumed to be fully turbulent. In the present application, with the least viscous fluid and the highest impeller speed, the mixing Reynolds number is approximately 100, which indicates all simulations are laminar. Fluent gives the option to select different viscosity model, in this work the non-Newtonian-power-law was selected, therefore, a non-Newtonian flow was modelled according to Equation 11.

Values $n$ and of $K_s$ (**Error! Reference source not found.**) have been calculated for each material by dynamic shear rheometer and used in Fluent. Then, the effective shear rate Vs the rotational speed for both SC-27 and DHR was calculated (**Error! Reference source not found.**).

Furthermore, a "cut-off viscosity" was defined in the power-law model to run the simulation, this viscosity arbitrarily given will be the highest viscosity predicted by the software when simulating at very low shear rates. This is necessary because Ostwald-de Waele model gives an infinitive viscosity at shear rates close to zero. Finally, particles were added by defining the diameter (0.300 mm) and the concentration (15%). Therefore, non-Newtonian behaviour is applied to the continuous phase, and then crumb rubber particle particles were added to simulate their motion within continuous phase (bitumen). A moving mesh approach was used, where the entire fluid domain was rotated about the vertical z axis at the appropriate angular velocity, while the outer wall was held stationary relative to the moving zone. The reason for doing this, rather than simply moving the outer wall relative to a stationary fluid domain was so that animations for the DHR impeller could be produced with the helices being seen in motion. There was no significant computational overhead associated with the approach used. Then rotational speed was set from 0 to 100 Rpm. For both the geometries; no slip wall boundary conditions were applied to the inner, outer and bottom walls. The upper boundary was set as a symmetry plane as the shear between air and bitumen is insignificant. Standard solver settings were used throughout: the SIMPLE algorithm was used for pressure–velocity coupling. Least Squares Cell Based discretization for gradients and second order differencing for the momentum and volume fraction equations were used. Due to the use of a moving mesh, the solution was necessarily transient, and it was found that the simulations had to be run up to 45 min of real time to achieve a stationary solution, where the mean velocity and volume fraction did not drift with time, as measured at a number of monitoring points. To simulate the movement of the DHR with accuracy, a maximum angular change of 2º was allowed per time-step. For example, at 100 Rpm this means 1 revolution every 0.6 sec, 1 revolution is 360º therefore the DHR needs 180-time steps per revolution to complete a revolution, so it needs a time steep of 0.6\180= 0.00333 sec.

## 3. Results & Discussion

### *3.1. Laboratory campaign*

#### *3.1.1 Measurements on neat bitumen*

As mentioned above the whole experimental programme was carried out within a range of temperatures between 120 °C and 200 °C. **Error! Reference source not found.** and **Error! Reference source not found.** shows viscosity measurements carried out on a 35/50 pen bitumen at 140 °C and 180 °C, by using both the SC-27 and the DHR impellers at different rotational speed. These two test temperatures were selected because close to the typically used to measure rotational viscosity at elevated temperatures of neat bitumen (ASTM D4402) and rubberized binders (ASTM D 6114-97). As a result, with a homogeneous material such as the neat bitumen DHR provides the same readings of the Sc-27.

#### *3.1.2 Measurements on modified bitumens: apparent viscosity vs time*

After the positive tests performed on neat bitumen, the SC-27 and the DHR have been used to measure the apparent viscosity of RTR-MB containing 15% (size 0.300 mm) and produced in high shear. Same tests were carried out for an SBS-MB produced by adding 7.5% of SBS 30% radial in 180/240 grade bitumen. Each test lasted 45 min, and in the first 15 mins the system was left reaching the thermal equilibrium (0 RPM). Considering the shear thinning nature of the modified bitumen, to evaluate the response of the two geometries tests were conducted respectively at 5, 20, 100 Rpm. **Error! Reference source not found.** shows the apparent viscosity measurement of the modified binders performed at 180 °C with a rotational speed of 20-100 Rpm. The difference in terms of viscosity between the two geometries at the end of the thermal equilibrium is due to the higher initial momentum required by the SC-27. **Error! Reference source not found.** instead shows the differences in terms of percentage between the two testing geometries at 1, 5 and 30 min. Considering that for both the materials similar trends were recorded at low rpm (5 rpm) for simplicity only the graph at 20 and 100 rpm are reported. It can be observed that the SC-27 impeller provides measurements that pass through a wider transition period before achieving a stable measurement, while DHR allows obtaining measurements with a more stable trend. This is quite evident with both SBS-MB and RTR-MB, however higher speeds (100 rpm) seems diminishing the unstable trend showed from the SC-27. Moreover, during the whole campaign the SC-27 provided values of apparent viscosity (after 45 mins) that are always lower than the DHR. If compared with the measurements carried out on the neat bitumen, it seems that the modified bitumen is in some way affected during the measurements.

Phenomena like phase separation and sedimentation may occur during the test leading to inconsistent results. **Error! Reference source not found.** highlights how the DHR instead provides a stable trend of results, which seems not be affected with time.

### *3.1.3 Measurements on modified bitumens: apparent viscosity vs temperature*

**Error! Reference source not found.** provides a numerical assessment of the differences recorded between the reading obtained with both SC-27 and DHR. It has to be underlined that differences between the two measurements increases with the increase in heterogeneity of the materials: neat bitumen > SBS-MB > RTR-MB.

In other words, using the SC-27 in standard conditions provides values of apparent viscosities that could be significantly different from those obtained with the DHR. Lo Presti et al. (2014), arrived at this same conclusion, with this investigation we are coupling the laboratory campaign with a numerical approach that could provide us with an insight of the phenomena happening inside the measuring chamber with the ambition of arriving to figure out the reasons behind this.

## ***3.2 Numerical simulations***

### *3.2.1 Validation of the model*

The CFD was configured to reproduce the whole laboratory campaign, although since both binders shows similar trend this simulation focused only on the most severe instability of the RTR-MB scenario. The numerical simulations want to provide further insight of the sample instability phenomena by assessing the velocity fields, volume fraction and numerical measurements distribution within the measuring chamber. In order to use these simulations as a reliable source of information, at first a validation of the model was carried out. The validation consisted in comparing numerical measurements of the torque with those obtained from the laboratory test. **Error! Reference source not found.** and **Error! Reference source not found.** show that the results obtained from the laboratory campaign and the numerical simulations, are in good agreement, therefore validating the reliability of the numerical model. Indeed, the software allows to compute the moments about a specified centre, and the coordinates of the centre of pressure for selected wall zones making possible the comparison with the experimental results. In support of this methodology, the reader should consider that rotational viscometers measure the viscosity by using a mechanical system to impose a controlled force or displacement onto a sample, hence the torque is the actual quantity directly measured during rotational tests. Once the model was considered able to reproduce the complex scenarios of rotational test even of complex binders, several investigations have been carried out. Each of them had the aim of providing a visual understanding of the phenomena happening within the measuring chamber while rotational measurements are performed. The next paragraphs offer the details of these simulations run only on the RTR-MB at 180 °C at 20 rpm, the binder with the highest level of heterogeneity



### *3.2.2 Velocity field distribution within the chamber*

A study of the velocity field distribution within the sample (**Error! Reference source not found.**) highlighted that the DHR shape allows a better distribution of the velocity flow field thanks to the pumping effect induced by the axial force. Indeed, the inner blades push the fluid upwards while the outer push the fluid downward allowing a continuous recirculation of the flow. Instead, the standard concentric spindle due to its geometry creates low velocity field at the top and the bottom of the tube, while all the stresses are distributed around the vertical walls of the spindle. This confirms that the vertical film of sample sandwiched along the container and the spindle is the only portion tested during the measurements. Also, there is no axial pumping but only tangential forces that do not allow creating a convective flow. Phenomena like phase separation, sedimentation and agglomeration may occur due to those issues.

### *3.2.3 Apparent viscosity measurements distribution within the chamber*

Complementary to the previous simulations, this section presents an analysis of the viscosity measurements made directly using the numerical model at different height of the container: top, middle and bottom thirds (**Error! Reference source not found.**). The figure also provides the comparison with the experimental data obtained within the laboratory campaign (last column). As a result, for the SC-27 high values have been found in the top and the bottom region, while in the middle region due to the tangential forces the values of viscosity reported are lower. This is in line to what was shown in the previous section showing that velocity fields are concentrated in the middle section; hence the measured torque will strongly depend from the tangential forces in this section. In other words, as shown in **Error! Reference source not found.** and **Error! Reference source not found.** , the values of viscosity reported by the instrument when the SC-27 is used, are related to the sample in the middle region. With the DHR the scenario is simplified. In fact, both **Error! Reference source not found.** and **Error! Reference source not found.** show that DHR is able to offer a better mixing of the sample, which in turns provide a more homogeneous distribution of the velocity fields and viscosity readings over the entire length of the container.

### *3.2.4 Evaluating the effect of time on sample stability*

On the basis of the lesson learned in the previous section, the simulations in **Error!**

**Reference source not found.** focused on understanding whether the viscosity measurements can be affected also by lack of sample stability over time. The idea was to reproduce the same methodology used for the experimental campaign, but with the additional feature of obtaining visual data by plotting the volume fractions distribution within the container and over time. As a result, in both scenarios, seems evident that within the thermal equilibrium (from 0 to 15 min), the particles tend to settle down to the bottom due to the gravity force. From 15 to 45, min the viscometer motor is turned on and the impellers start rotating at 20 Rpm. In this phase, the SC-27 is not able to keep the particles evenly distributed within the tube leading to a high concentration of particles at the bottom. It is also important to observe that the fluid along the clearance between the wall and spindle - which as discussed seems to be the actual sample tested with the SC-27 - is heterogeneous for the entire length of the test. Instead, once the DHR starts rotating, it assures an evenly distribution of the particles within few minutes. For this reason, the value reported by the viscometer using a standard geometry cannot consider realistic. On the other hand, the DHR allows testing a well-distributed sample and this is surely contributing in guaranteeing a stable trend of results, as observed during the laboratory campaign.

At last, simulations were run to understand whether the sample stability can be connected also the nature of the sample itself. In other words, simulations were run without the geometry in the tube for 45 min (**Error! Reference source not found.**). Results showed that there is a high level of instability within the sample due to the particles settling down for the gravity. As a result, sample of RTR-MB lack of sample stability over time, hence misleading results can be obtained if no agitation is provided during rotational tests.

## 6. Conclusions

This investigation offers an innovative approach consisting in a combined laboratory-based and computation approach aimed at understanding whether standard testing geometry for rotational tests are suitable for measuring high-temperature rotational viscosity of modified bituminous binders with high level of heterogeneity. The study also offers a solution to overcome some of the presented issues and this consists in changing the standard coaxial cylinder testing setup by substituting the standard spindle (SC-27) with a dual helical ribbon (DHR). In other words, the investigation aims at answering the following research questions:

Do heterogeneous modified bituminous binders undergo lack of sample stability during viscosity measurements at high-temperatures?

- The laboratory campaign provided evidence that highly heterogeneous modified bitumen do provide an un-stable trend of measurements during the testing with SC-27. The final values of apparent viscosities can be significantly different from those obtained with the DHR, which instead provided stable readings already after few minutes.
- The numerical simulations clarified that heterogeneous bituminous blends undergo sample instability already during thermal equilibrium, simply because they are stored at high temperatures in a vertical container. The lack of sample stability is not reversed when SC-27 is used at 20 rpm. In this testing configuration, higher rotational speed (100 rpm) might help decreasing but not eliminating the issue.

Do we obtain different results if sample stability is ensured?

- The DHR instead is able to ensure sample stability even at low speed (5 rpm). This is due to an axial pumping induced by the rotating impeller within the sample.
- The laboratory campaign provided evidence that increasing the complexity of the bituminous blends is directly proportional to an increase in misleading readings of apparent viscosity of the blends, up to almost 40%. This trend is mainly governed by the material type as well as to differential values of densities and viscosities of the blend's components.
- The CFD analysis, provided the additional opportunity to have readings over three thirds of the vertical container. Results highlighted that SC-27 provides readings mainly governed by the middle region, while the viscosity results over the length of the tube are quite uniform when the DHR is used as impeller.

Hence, as general conclusion can be stated that bitumen technologist are invited to not underestimate sample stability arising while performing high-temperature viscosity measurements of heterogeneous modified bitumen with the standard coaxial cylinders setup. Sample stability certainly occurs when testing recycled tyre rubber modified bitumen, and it might be significant also for polymer modified bitumen such as SBS-MB. The DHR impeller offers a solution to ensure sample stability, hence asphalt technologists are strongly recommended to consider this solution within rheological characterization of complex and heterogeneous bituminous binders at high-service temperatures.

This study was tailored for the rotational viscometer environment; however, the authors have already developed the same concept for Dynamic Shear Rheometers. Research is currently undergoing. Future recommendations can lie in using a similar approach to explore a wider range of modified binders and impellers. Also, this type of combined laboratory/computational approach is strongly recommended since provides new set of data and visual aids that could lead towards the definition of virtual laboratory testing protocols.

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## 8. Author contributions

The authors confirm contribution to the paper as follows:
study conception and design: Davide Lo Presti, Gaspare Giancontieri,
Data collection: Gaspare Giancontieri; Davide Lo Presti
Numerical simulations: Gaspare Giancontieri; David Hargreaves
Analysis and interpretation of results: Gaspare Giancontieri, David Hargreaves, Davide Lo Presti;
Draft manuscript preparation: Gaspare Giancontieri, Davide Lo Presti
All authors reviewed the results and approved the final version of the manuscript.



## 9. References

ASTM. Standard test method for viscosity determination of asphalt at elevated temperatures using a rotational viscometer. American society for testing and materials, a. D4402/d4402m

ASTM. Standard test method for measurement of apparent viscosity of asphalt rubber or other asphalt binders by using a rotational hand-held viscometer. American society for testing and materials, b. D7741/d7741m.

Bertrand, F. *et al.* (1999) 'Numerical modeling of the mixing flow of second-order fluids with helical ribbon impellers', *Computer Methods in Applied Mechanics and Engineering*, 180(3–4), pp. 267–280. doi: 10.1016/S0045-7825(99)00169-3.

Caltrans (2011) 'Quality Control Manual for Hot Mix Asphalt - State of California', *State of California, Department of Transportation, Division of Construction*, (June). Available at: http://www.dot.ca.gov/hq/construc/publications/qcqaman1.pdf.

Carreau, P. J., Patterson, W. I. and Yap, C. Y. (1976) 'Mixing of viscoelastic fluids with helical-ribbon agitators I- Mixing time and flow patterns.', *Can. J. Chem. Eng.*

Celauro, B. *et al.* (2012) 'Definition of a laboratory optimization protocol for road bitumen improved with recycled tire rubber', *Construction and Building Materials*. Elsevier Ltd, 37, pp. 562–572. doi: 10.1016/j.conbuildmat.2012.07.034.

Cordobes, F., Brito de la fuente, E. and Gallegos, C. (1998) 'Effect of rheological properties on power input and effective shear rates when mixing with different impellers.', *Anales de química*. Springer, 94(4–5), pp. 274–278. Available at: http://cat.inist.fr/?aModele=afficheN&cpsidt=10064702 (Accessed: 20 April 2015).

Delaplace, G., Leuliet, J. C. and Relandeau, V. (2000) 'Circulation and mixing times for helical ribbon impellers. Review and Experiments', *Experiments in Fluids*, 28(2), pp. 170–182. doi: 10.1007/s003480050022.

Dow, R. B. (1946) 'The rheology of lubricants', pp. 81–91.

Gallegos, C. and Franco, J. M. (1999) 'Rheology of food, cosmetics and pharmaceuticals', *Current Opinion in Colloid and Interface Science*, 4(4), pp. 288–293. doi: 10.1016/S1359-0294(99)00003-5.

Giancontieri, G., Hargreaves, D. and Lo Presti, D. (2018) 'Are we correctly measuring the rotational viscosity of rubberized bituminous binder ?', in *Proceedings of the Rubberised Asphalt - Asphalt Rubber 2018 Conference*.

Huang, S. C. (2006) 'Rheological characteristics of crumb rubber-modified asphalts with long-term aging', *Road Materials and Pavement Design*, 7(January 2015), pp. 37–56. doi: 10.1080/14680629.2006.9690057.

Lin, S. and Brodkey, R. S. (1985) 'Rheological Properties of Slurry Fuels', *Journal of Rheology*, 29, p. 147. doi: 10.1122/1.549814.

Malkin, A. Y. (2009) 'The state of the art in the rheology of polymers: Achievements and challenges', *Polymer Science Series A*, 51(1), pp. 80–102. doi: 10.1134/S0965545X09010076.

Metzner, A. B. and Otto, R. E. (1957) 'Agitation of non-Newtonian fluids', *AIChE Journal*, 3(1), pp. 3–10. doi: 10.1002/aic.690030103.

Mezger, T.G. (2006) *The Rheology Handbook*.

Paul, E. L., Atiemo-obeng, V. a and Kresta, S. M. (2004) *Handook of industrial mixing*.

Peralta, J. *et al.* (2010) 'Changes in Rubber Due to its Interaction with Bitumen when Producing Asphalt Rubber', *Road Materials and Pavement Design*, 11(4), pp. 1009–1031. doi: 10.1080/14680629.2010.9690317.

Lo Presti, D. *et al.* (2014) 'Toward more realistic viscosity measurements of tyre rubber–bitumen blends', *Construction and Building Materials*. Elsevier Ltd, 67, Part B(0), pp. 270–278. doi: http://dx.doi.org/10.1016/j.conbuildmat.2014.03.038.

Lo Presti, D., Giancontieri, G. and Hargreaves, D. M. (2017) 'Improving the rheometry of rubberized bitumen: experimental and computation fluid dynamics studies', *Construction and Building Materials*, 136, pp. 286–297. doi: 10.1016/j.conbuildmat.2016.12.200.

Smith, J. . (1970) 'The Mixing of Newtonian and non- Newtonian fluids', *J. Soc. Cosmet. Chem*, 552, p. 242. Available at: https://books.google.com/books/about/The_Mixing_of_Rubber.html?id=NV7L3Jb1URwC&pgis=1.

Sommerfeld, M. and Decker, S. (2004) 'State of the art and future trends in CFD simulation of stirred vessel hydrodynamics', *Chemical Engineering and Technology*, 27(3), pp. 215–224. doi: 10.1002/ceat.200402007.

Subhy, A., Lo Presti, D. and Airey, G. (2015) 'Rubberised bitumen manufacturing assisted by rheological measurements', *Road Materials and Pavement Design*, 16(November), pp. 1–21. doi: 10.1080/14680629.2015.1079549.

Subhy, A., Lo Presti, D. and Airey, G. (2016) 'Rubberised bitumen manufacturing assisted by rheological measurements', *Road Materials and Pavement Design*, 17(2), pp. 290–310. doi:

10.1080/14680629.2015.1079549.

Tabilo-Munizaga, G., Barbosa-Cánovas, G. V and Tonir, S. (2005) 'Rheology for the food industry', *Journal of Food Engineering*, 67(1–2), pp. 147–156. doi: 10.1016/j.jfoodeng.2004.05.062.

Takahashi, K. *et al.* (1982) 'Effects of Geometrical Variables of Helical Ribbon Impellers on Mixing of Highly Viscous Newtonian Liquids.', *Journal of Chemical Engineering of Japan*, 15(3), pp. 217–224. doi: 10.1252/jcej.15.217.

Takahashi, K., Yokota, T. and Konno, H. (1979) 'Mixing of pseudoplastic liquid in a vessel equipped with a variety of helical ribbon impellers', *Journal of Chemical Engineering of Japan*, 45(1978), pp. 63–68.

Takahashi, K., Yokota, T. and Konno, H. (1984) 'Power Consumption of Helical Ribbon Agitators in Highly Viscous Pseudoplastic Liquids.', *Journal of Chemical Engineering of Japan*, 17(6), pp. 657–659. doi: 10.1252/jcej.17.657.

Tanguy, P. a *et al.* (1992) 'Finite element analysis of viscous mixing with a helical ribbon-screw impeller', *AICHE Journal*, 38(6), pp. 939–944. doi: 10.1002/aic.690380614.

Um, B. and Hanley, T. R. (2008) 'A CFD model for predicting the flow patterns of viscous fluids in a bioreactor under various operating conditions', 25(5), pp. 1094–1102.

Visser, A. T., Duhovny, G. S. and Sachkova, A. V. (2017) 'Comparison of bitumen–rubber use in extreme conditions in Russia and South Africa', *Road Materials and Pavement Design*, 18(5), pp. 1190–1199. doi: 10.1080/14680629.2016.1201524.

Yap, C. Y., Patterson, W. and Carreau, P. J. (1979) 'Mixing with helical ribbon agitators: Non-Newtonian Fluids', *AIChE Journal*, 25(3), pp. 516–521.

Zhang, Y. *et al.* (2018) 'Comparison of the partitioning of evapotranspiration – numerical modeling with different isotopic models using various kinetic fractionation coefficients', *Plant and Soil*. Plant and Soil, 430(1–2), pp. 307–328. doi: 10.1007/s11104-018-3737-z.