Durability assessment of bio-extended bituminous binders by rheological characterisation after long-term ageing

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Contribution theme
02. Binder performance and testing

Executive summary

The pursuit of sustainable road infrastructure is prompting extensive research into the utilization of bio-extended bituminous binders. This paper investigates the durability of bio-extended bituminous binders (both unmodified and polymer-modified) containing a plant-based bio-oil, aiming to assess their long-term performance. The research employed laboratory-scale tests and advanced rheological techniques to evaluate viscoelastic properties of the binders after long-term ageing. Accelerated ageing protocols were used to simulate prolonged exposure to environmental factors, including the Rolling Thin Film Oven Test (RTFOT) and Pressure Ageing Vessel (PAV) test. Dynamic Shear Rheometer (DSR) tests measured the shear modulus and phase angle of the binders under various conditions while Bending Beam Rheometer (BBR) tests measured the flexural creep stiffness at different low temperatures. Various performance indicators and specification parameters were analysed for the investigated binders regarding their resistance to both fatigue cracking at intermediate temperatures and thermal cracking at low temperatures. The results indicate that the bio-extended binders passed all the analysed criteria after RTFOT+PAV ageing. Using the investigated bio-oil in bituminous binders had largely a positive effect on the ΔT_c and did not affect the standard performance low-temperature grade significantly. The bio-extended binders followed the same relationships between several durability-related performance indicators as the reference binders did. Certain slight negative effects could be noticed at low temperatures when the bio-oil content was high, but a further verification on the asphalt mixture scale will be necessary to assess if these effects are significant enough to impact the asphalt mixture performance in practical applications.

Keywords
bio-bitumen, polymer-modified bitumen, rheology, durability, cracking resistance

1. INTRODUCTION

Bituminous binders are commonly used in the construction and maintenance of asphalt pavements. They are typically composed of bitumen – a viscous, black, and sticky substance that is derived from crude oil – and, if necessary, other additives such as polymers to improve their properties. Bituminous binders are used to hold the materials together in asphalt mixtures and provide strength to the resulting pavement. However, the use of traditional petroleum bitumen has several environmental and sustainability concerns, including its dependence on non-renewable resources and relatively high energy consumption and greenhouse gas emissions during production. As a result, there is a growing interest in the development of alternative binders that are partially or completely derived from renewable sources and thus have a lower environmental impact.

Biomass resources have been investigated as potential alternatives to traditional petroleum bitumen for paving purposes [1]. In this context, plant biomass would be a preferred source over animal biomass. This is because plant biomass is more readily available and has a lower environmental impact compared to animal biomass. Plant biomass, such as lignin and hemicellulose, can be extracted from various sources such as wood, agricultural waste, and energy crops. These materials can be converted into bio-binders or bio-oils, some of which can then be used as bitumen extenders or replacements. Among the related investigations so far, the use of plant-based bio-oils as bitumen extenders has shown the most promising results [2, 3]. The success of this approach is attributed to several factors. Firstly, the use of bio-oils, especially at low content, has not shown a significant negative impact on binder properties. Secondly, bio-oils can replace a relatively large portion of bitumen compared to other approaches, leading to greater benefits. Lastly, replacing petroleum bitumen (as opposed to other components in the asphalt mixture) is more effective in addressing the environmental and sustainability concerns.

Plant-based bio-oils are renewable resources that can be obtained from various biomass sources, such as wood, agricultural residues, or waste cooking oil. By blending the bio-oil with bitumen, the resulting binder can provide a more sustainable and environmentally friendly alternative to purely using traditional bitumen derived from crude oil in asphalt pavements. In addition, previous studies [4, 5] concluded that certain plant-based bio-oils can even help improve some properties of bituminous binders, for example, the flexibility, adhesion, and workability of binders.

However, since bio-oils have different chemical compositions compared to bitumen, they may affect the binder structure and performance in a different way, particularly the long-term performance. Different bio-oils have different chemical compositions too. Thus, there are also some challenges associated with the use of bio-oils as bitumen extenders, such as compatibility issues, ageing effects, and durability concerns. Further research is needed to optimize the formulation and performance of bituminous binders extended with properly selected plant-based bio-oils for different applications and climatic conditions. Looking into the relevant literature to date, many previous studies have focused on examining the benefits of utilizing plant-based bio-oils during the initial construction phase of asphalt pavements, including the asphalt mixing process and initial properties of asphalt binders and mixtures [6-8]. Assessing their long-term durability under various traffic and climatic conditions is still necessary and continues to pose a challenge for capturing their benefits.

Focusing on the durability assessment, this study investigated a specific type of plant-based bio-oil for its use as an extender in bituminous binders. The investigated plant-based bio-oil is from a biorefinery refining bio-based by-products from the pulp and paper industry. It contains primarily high-boiling esters as well as free acids. In terms of material source, the bio-oil is essentially derived from the forest. The research of using this plant-based bio-oil in bituminous binders and asphalt mixtures has a long history. But until recently its practical application has been very limited. However, due to the current increasing demand for climate-smart materials and technical solutions, there has been a growing interest in exploring its use as an extender in bituminous binders for various applications. Recent advancements and research have led to increased attention and investigation into the potential benefits and performance of bituminous binders extended with this plant-based bio-oil. As a result, its practical applications are now being explored and evaluated to assess the effectiveness and feasibility. For this, the durability assessment is an indispensable aspect.

In this study, the durability of bio-extended bituminous binders (both unmodified and polymer-modified) containing the investigated plant-based bio-oil was evaluated using the approach of rheological characterisation after long-term ageing. By subjecting the binders to long-term ageing, which simulates the effects of prolonged exposure to environmental factors such as oxygen and heat, this study aimed to evaluate how the rheological properties of the bio-extended bituminous binders changed over time. A particular focus was placed on the changes of relaxation property after long-term ageing and the resistance to both fatigue cracking at intermediate temperatures and thermal cracking at low temperatures. It is expected that such a durability study will help in understanding the potential benefits and limitations of bio-extended bituminous binders containing the investigated plant-based bio-oil. The research output will provide researchers and practitioners new insights into the long-term performance of the bio-extended bituminous binders, thus informing their practical application in real-world conditions.

2. EXPERIMENTAL

2.1. Materials

Six bituminous binders were analysed in this study. They were of three different grades according to EN 12591:2009 and EN 14023:2010, namely two penetration grade 70/100 binders, two penetration grade 160/220 binders, and two polymer-modified bitumen (PMB) grade 40/100-75 binders. For each grade, there was a bio-extended binder with the investigated plant-based bio-oil (containing primarily high-boiling esters and free acids) and a commercially available reference binder for comparison purposes. All the binders are listed in Table 1 with their penetration (EN 1426:2015) and softening point (EN 1427:2015) values. Their performance grades (PG) were also determined according to ASTM D6373-21a and AASHTO M 320-22, as presented in Table 1.

Table 1. Bituminous binders analysed in this study

Binders	Bio 70/100	Ref 70/100	Bio 160/220	Ref 160/220	Bio PMB 40/100-75	Ref PMB 40/100-75
Penetration @ 25 °C, 0.1 mm	80	82	193	196	66	53
Softening point, °C	47.0	47.4	40.4	38.8	86.5	81.5
PG grade	58-22	64-22	52-28	52-28	76-22	82-16

2.2. Test methods

The six bituminous binders were aged in laboratory with the Rolling Thin Film Oven Test (RTFOT at 163 °C for 75 min according to EN 12607-1:2014) followed by Pressure Ageing Vessel (PAV) ageing at 100 °C and 2.1 MPa for 20 h according to EN 14769:2012. After the long-term ageing, the conditioned binders were analysed with various test methods for rheological characterisation. The dynamic shear modulus |G*| and phase angle δ of the binders were determined by a Dynamic Shear Rheometer (DSR) within the linear viscoelastic range at different temperatures and frequencies according to EN 14770:2012. As the aim of this study was to assess the durability of long-term aged binders, the focus of the DSR testing was placed on the intermediate temperature range (for evaluating the fatigue cracking resistance) and the low temperature range (for evaluating the thermal cracking resistance). The plate-plate geometry was used for the DSR testing. At intermediate temperatures, the plates of 8 mm diameter were used with 2 mm gap. At low temperatures, the plates of 4 mm diameter were used with 2 mm gap. In addition, the flexural creep stiffness of the long-term aged binders was determined by a Bending Beam Rheometer (BBR) at different low temperatures according to EN 14771:2012. The number of replicates for each test followed the requirements in the respective standard method.

3. RESULTS AND DISCUSSION

3.1. Intermediate-temperature fatigue cracking resistance

Fatigue cracking is a common failure mode of asphalt pavements after certain years of service. There are various technical parameters for regulating bituminous binders in the intermediate temperature range to minimise the fatigue cracking risk of asphalt mixtures. Table 2 presents the results of the investigated binders (after long-term ageing) for the currently most commonly used intermediate-temperature fatigue parameter |G*|·sin(δ) by ASTM D6373-21 and AASHTO M 320-22. As the PG grades of the binders were different in this study, by the current standards, their test temperatures for |G*|·sin(δ) were different, ranging from 16 °C to 37 °C. The test frequency (angular) was the same for all the binders, namely 10 rad/s. Table 2 shows that all binders passed the fatigue criterion of their respective PG grade (maximum 5000 kPa).

Table 2. Intermediate-temperature fatigue parameter |G*|·sin(δ) by ASTM D6373-21 and AASHTO M 320-22

Binders (after RTFOT+PAV)	Bio 70/100	Ref 70/100	Bio 160/220	Ref 160/220	Bio PMB 40/100-75	Ref PMB 40/100-75
Test temperature, °C	22	25	16	16	31	37
|G*|·sin(δ), kPa	3528	2280	3477	2930	1071	626

Despite its great popularity, the fatigue parameter |G*|·sin(δ) has been widely criticized [9-11]. It does not seem to correlate well with the fatigue performance of bituminous binders in the field. A recent study conducted in the United States (US) [12] investigated several alternative binder fatigue tests and parameters for specification purposes. As a conclusion, it recommended the Glover-Rowe parameter G'/tan(δ) and Christensen-Anderson R-value (rheologic index) after long-term ageing as the technical parameters for evaluating and regulating the binder fatigue performance. The study also proposed a change in test temperature for fatigue evaluation. Table 3 presents the results of the investigated binders for Glover-Rowe parameter G'/tan(δ) after RTFOT+PAV ageing at 10 rad/s. Their values at several different temperatures are listed, both at the proposed temperatures by the recent study [12] according to the binder PG grades and at 10 °C. The reason why the results at 10 °C are also presented is that the recently proposed temperatures for unmodified binders appeared to be even higher than their current standard temperatures (Table 2), although those proposed ones for PMB binders did become lower. Considering the specific climate conditions in Sweden, it is usually at 10 °C that the pavement fatigue performance is evaluated [13, 14]. Thus, 10 °C would be a reasonable level for comparing the different binders under the same condition. Table 3 shows that all binders also passed the new fatigue criterion of their respective PG grade as proposed by the recent study [12] (maximum 5000 kPa). At 10 °C, the G'/tan(δ) values of bio-extended binders after long-term ageing were at similar levels as their corresponding reference binders. Comparing pairwise, the differences within the same binder grade were in the range of only about ±10%. Over 22 °C, however, it seems that bio-extended binders have lower G'/tan(δ) values than the reference binders. This can be interpreted as a positive sign that bio-extended binders have slightly higher fatigue resistance than the reference binders in the temperature range over 22 °C.

Table 3. Glover-Rowe parameter G'/tan(δ) after RTFOT+PAV at 10 rad/s and different temperatures

Binders (after RTFOT+PAV)	Bio 70/100	Ref 70/100	Bio 160/220	Ref 160/220	Bio PMB 40/100-75	Ref PMB 40/100-75
G'/tan(δ) @ 10 °C, kPa	34081	35262	10721	9632	35742	40289
G'/tan(δ) @ 22 °C, kPa	N/A	N/A	518	765	N/A	N/A
G'/tan(δ) @ 25 °C, kPa	1347	2222	N/A	N/A	3237	N/A
G'/tan(δ) @ 27 °C, kPa	N/A	N/A	N/A	N/A	N/A	3429

The Christensen-Anderson R-value is one of the parameters in the Christensen-Anderson (CA) model for fitting binder master curves. It is defined as the difference between the logarithmic values of glassy (shear) modulus G_g and crossover (shear) modulus G_c. The R-value reflects the shape of the shear modulus master curve and is a good indicator of overall strain tolerance of the binder [12]. The glassy modulus G_g of bituminous binders is generally assumed as 1 GPa while the crossover modulus G_c is the dynamic shear modulus |G*| when the phase angle is 45° (namely G'=G''). At the crossover point, a binder shows equal elasticity as viscosity. It is considered as a sign to identify the broad transition of bituminous binders between the viscoelastic solid state and viscoelastic fluid state [15]. Thus, the related crossover parameters have also been investigated as potential binder performance indicators. Table 4 presents the results of the investigated binders for a few crossover parameters after long-term ageing. It is indicated that, at 10 rad/s, the bio-extended binders after RTFOT+PAV ageing have lower crossover temperature values than their respective reference binders. This means for bio-extended binders that they have a lower temperature limit up to which the elastic part of complex shear modulus G* (i.e., the elastic/storage shear modulus G') is dominant to the viscous part (i.e., the viscous/loss shear modulus G"). According to the time-temperature superposition principle, a higher crossover frequency at a specified reference temperature would have a similar effect, as demonstrated by the crossover angular frequency values in Table 4. Eventually, these differences lead to higher crossover modulus values of the bio-extended binders after long-term ageing as compared to the reference binders (as in Table 4).

Table 4. Crossover parameters (δ=45° and G'=G'') after RTFOT+PAV

Binders (after RTFOT+PAV)	Bio 70/100	Ref 70/100	Bio 160/220	Ref 160/220	Bio PMB 40/100-75	Ref PMB 40/100-75
Crossover temperature @ 10 rad/s, °C	18.6	24.6	12.6	15.2	33.6	40.3
Crossover angular frequency @ 10 °C, rad/s	0.287	0.116	2.593	0.659	N/A	N/A
Crossover angular frequency @ 40 °C, rad/s	N/A	N/A	N/A	N/A	97.64	7.993
Crossover shear modulus*, kPa	7332	3938	7482	3867	1388	600

*Average shear modulus at the indicated crossover temperature and crossover angular frequency.

Based on the crossover shear modulus values presented in Table 4, the Christensen-Anderson R-value can be calculated for each binder, as listed in Table 5 (the first row under headers). However, as can be noted in Table 4, the crossover modulus of the PMB binders was significantly lower than that of unmodified binders. This results in considerably higher R-value for PMBs as determined by the crossover modulus, which may affect their fatigue evaluation. To have an integrated approach for different binder types, according to Christensen and Tran [12], the Christensen-Anderson R-value should be calculated at a sufficiently high modulus level where the polymer modification effect is restricted. The researchers suggested two alternative methods for determining the R-value. First, at a high |G*| level (at least 10 MPa), Equation 1 can be used for determining the R-value.

$$R = \log(2) \cdot \frac{\log(|G^*| \cdot 10^{-9})}{\log\left(1 - \frac{\delta}{90}\right)} \quad (1)$$

In Equation 1, R is the Christensen-Anderson R-value (rheologic index); |G*| is the dynamic shear modulus in Pa; and δ is the phase angle in degrees (°) at the same temperature and frequency as |G*|. Second, at an even higher stiffness level, the BBR test results can be used for determining the R-value by Equation 2.

$$R = \log(2) \cdot \frac{\log(S/3000)}{\log(1-m)} \quad (2)$$

In Equation 2, S is the BBR creep stiffness at 60 s in MPa; and m is the BBR m-value at 60 s and at the same temperature as S. The calculation results of the investigated binders with the two alternative methods after long-term ageing are presented in Table 5 (the second and third rows under headers). The study by Christensen and Tran [12] claimed that binders with high R-values can result in rapid accumulation of fatigue damage in thin pavements at low temperatures while binders with low R-values can show poor fatigue performance in thick pavements. Thus, they proposed both a maximum limit and a minimum limit for the R-value criterion, namely a range from 1.5 to 2.5 as determined with the BBR test results (Equation 2). The results in Table 5 (the third row under headers) indicate that all binders passed this criterion. In addition, Table 5 shows that the bio-extended binders after RTFOT+PAV ageing have lower R-values than their respective reference binders. This is basically because of their higher crossover modulus as shown in Table 4.

Table 5. Christensen-Anderson R-value (rheologic index) after RTFOT+PAV determined with different methods

Binders (after RTFOT+PAV)	Bio 70/100	Ref 70/100	Bio 160/220	Ref 160/220	Bio PMB 40/100-75	Ref PMB 40/100-75
By crossover shear modulus	2.1	2.4	2.1	2.4	2.9	3.2
By DSR testing over 10 MPa	2.1	2.6	2.1	2.4	3.0	3.4
By BBR testing at -18 °C	1.8	1.9	1.9	2.1	2.2	2.4

Besides the binder fatigue parameters described above (mostly for PG grading), there are also some recent developments in Europe. For example, the latest revision of the European standard EN 14770 (final draft version FprEN 14770:2023 so far) proposes the determination of equi-shear modulus temperature and phase angle at 10 rad/s and at different |G*| levels using DSR. Among the proposed equi-shear modulus parameters, the temperature T3, at which |G*| is equal to 5 MPa after long-term ageing, is in the intermediate temperature range and holds the possibility to be a parameter for binder fatigue evaluation. Table 6 lists the results of the investigated binders for T3 and the phase angle at T3. Although no criterion has been established so far, they seem to have certain relationships with the above-described Glover-Rowe parameter G'/tan(δ) and R-value. Figure 1 presents the linear relationships between T3 and the Glover-Rowe parameter at 10 °C as well as between δ_{T3} and the R-value by DSR testing over 10 MPa. These relationships show that the bio-extended binders after RTFOT+PAV ageing follow the same interconversion rules between the indicated intermediate-temperature parameters as the reference binders do.

Table 6. Rheological parameters T3 and δ_{T3} (|G*|=5 MPa at 10 rad/s) after RTFOT+PAV

Binders (after RTFOT+PAV)	Bio 70/100	Ref 70/100	Bio 160/220	Ref 160/220	Bio PMB 40/100-75	Ref PMB 40/100-75
T(|G*|=5 MPa), °C	21.6	22.0	15.5	14.7	22.0	23.1
$\delta_{T(\|G^*\|=5\ MPa)}$, °	48.4	42.4	48.8	44.5	37.1	33.9

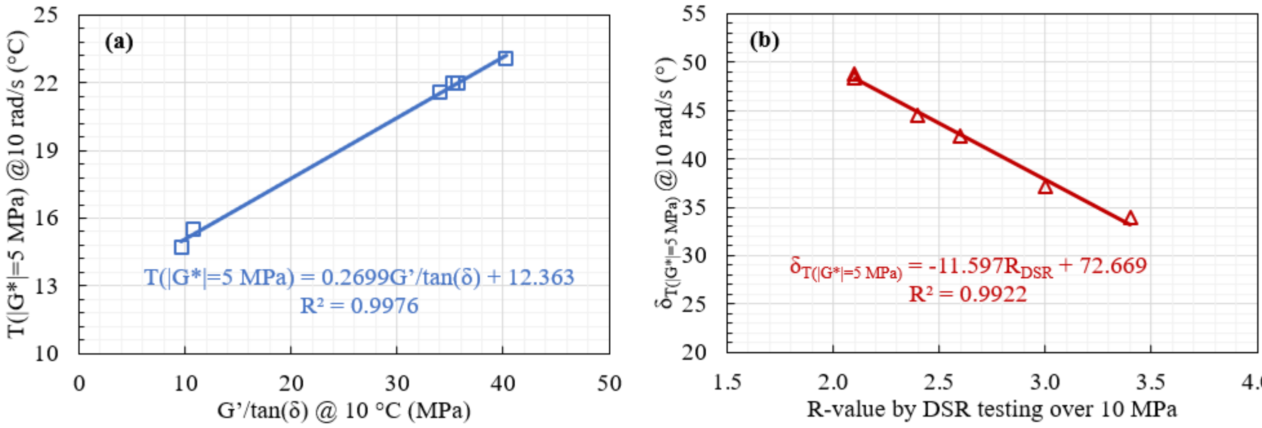

Figure 1: Relationship between different intermediate-temperature fatigue parameters after RTFOT+PAV – (a) T3 versus Glover-Rowe parameter G'/tan(δ) at 10 °C; (b) δ_{T3} versus R-value by DSR testing over 10 MPa

3.2. Low-temperature thermal cracking resistance

At low temperatures, asphalt mixtures undergo the thermal contraction process, which can result in the development of thermal stresses within the pavement. The thermal stresses that arise from this shrinkage can potentially cause damage to the pavement, typically thermal cracking. To minimise the risk of thermal cracking in asphalt mixtures, some test methods and technical parameters can be employed to ensure that the binder has suitable properties to withstand the stresses induced by low temperatures. This study employed the 4-mm DSR test and the BBR test after RTFOT+PAV ageing for evaluating the thermal cracking resistance of the investigated binders. Figure 2 presents the 4-mm DSR test results (dynamic shear modulus |G*| and phase angle δ) at 10 rad/s and different low temperatures. Although it is at a much higher frequency level than the actual thermal stress fluctuation in pavements, the results in Figure 2 may imply the relative levels of binder properties at much lower temperatures. It is indicated that, after long-term ageing and at very low temperatures, the bio-extended binders without polymer had to some extent higher |G*| than their respective reference binders. Meanwhile, the bio-extended PMB binder showed slightly lower |G*| than its reference binder. As for the phase angle at very low temperatures, the bio-extended 160/220 binder had significantly lower δ than the reference 160/220 bitumen after RTFOT+PAV ageing while the δ values of the other bio-extended binders were on similar levels as their corresponding reference binders. It is notable that most of these comparisons between bio-extended and reference binders led to differences that were not significantly larger than the precision of the test method (estimated repeatability: 15% for |G*| and 2° for δ). Thus, a further analysis of the 4-mm DSR results was needed to draw reliable conclusions.

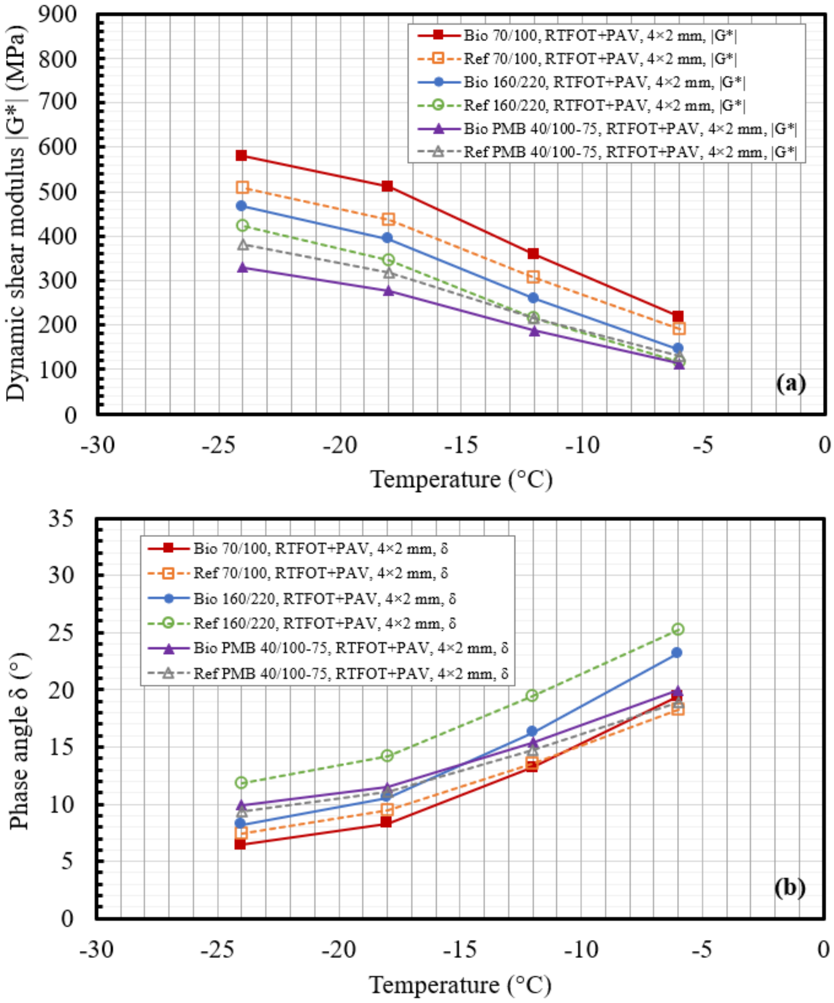

Figure 2: Test results at 10 rad/s with 4-mm DSR after RTFOT+PAV – (a) dynamic shear modulus |G*|; (b) phase angle δ

The Western Research Institute (WRI) method (revised approach at PG+10 °C) [16] was used to further analyse the 4-mm DSR test results and determine the low-temperature rheological properties of the investigated binders, including the shear stress relaxation modulus G(t) and its rate of change (slope) m_r at 60 s. With this method, the shear stress relaxation modulus G(t) of the investigated binders after long-term ageing could be obtained by approximate interconversion from the elastic/storage shear modulus G'(ω) by Equation 3 [17].

$$G(t) \approx G'(\omega)\big|_{\omega=\frac{2}{\pi t}} \qquad (3)$$

In Equation 3, G(t) is the shear stress relaxation modulus; G'(ω) is the elastic/storage shear modulus; ω is the angular frequency; and t is the loading time. The 4-mm DSR test data were at different frequencies (0.1-100 rad/s) and low temperatures (-6 °C, -12 °C, -18 °C, and -24 °C). The G' master curves were constructed by adopting the sigmoidal model for fitting and the William-Landel-Ferry (WLF) equation for shift factors. After this, the G(t) master curves could be obtained by the approximate interconversion (Equation 3). Figure 3 presents the obtained G(t) master curves for the investigated binders after RTFOT+PAV ageing at different reference temperatures. By fitting the G(t) master curve with a second order polynomial, the relaxation modulus and its slope at 60 s – G(60 s) and m_r(60 s) – could be determined at different reference temperatures. According to Farrar et al. [16], the G(60 s) level of 143 MPa corresponds to the BBR creep stiffness 300 MPa and the m_r(60 s) level of 0.280 corresponds to the BBR m-value 0.300 at 60 s and the same temperature as the master curve reference temperature. Thus, the critical temperatures T_{cG} at which G(60 s) is equal to 143 MPa and T_{cmr} at which m_r(60 s) is equal to 0.280 could be calculated. The difference between them (T_{cG}-T_{cmr}) is defined as the ΔT_c value determined by 4-mm DSR testing. All these analysis results of the investigated binders after long-term ageing are shown in Table 7.

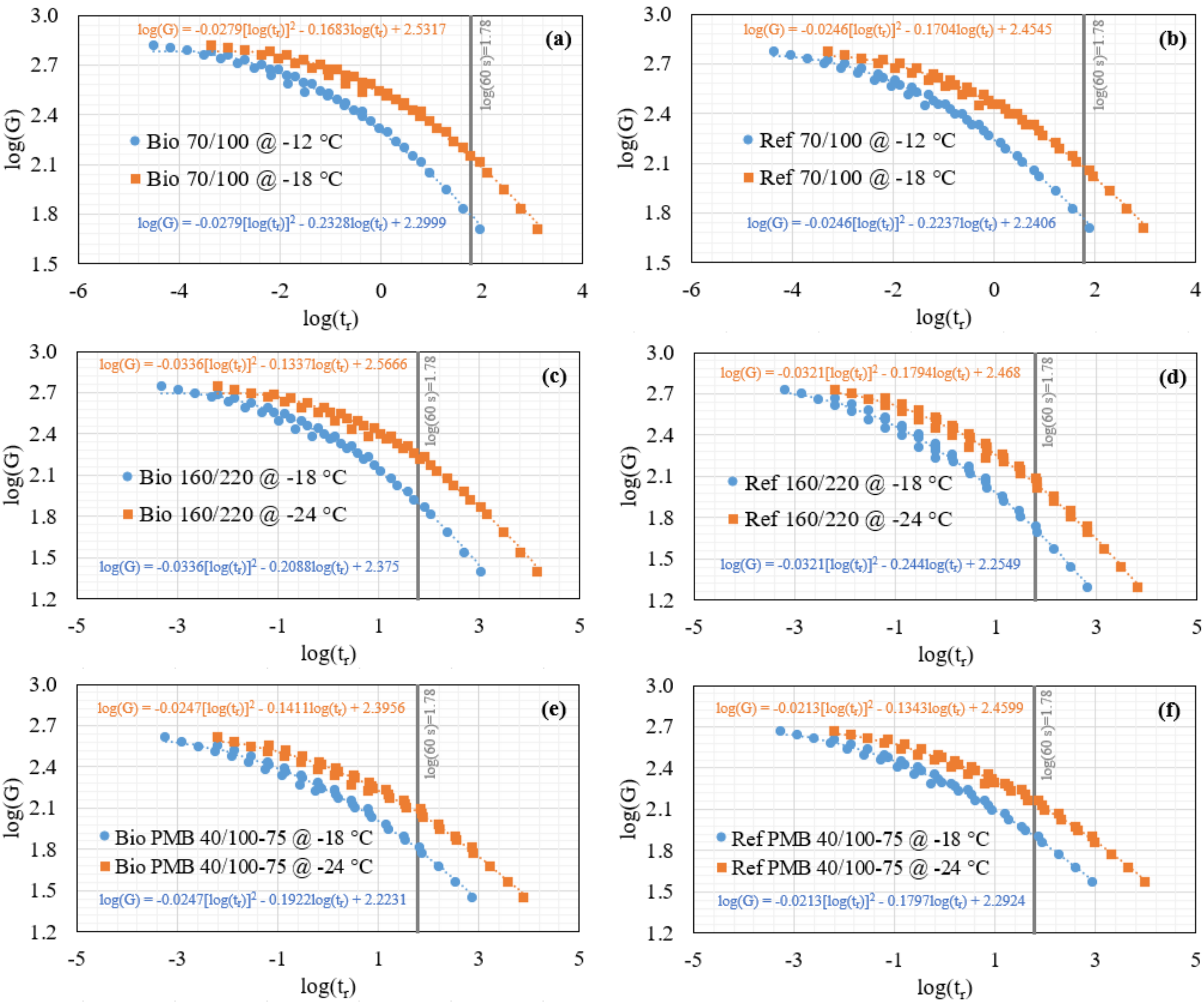

Figure 3: Shear stress relaxation modulus G(t) by 4-mm DSR testing – (a), (b), (c), (d), (e), (f) for the six different bituminous binders after RTFOT+PAV

Table 7. Analysis results of 4-mm DSR testing after RTFOT+PAV

Binders (after RTFOT+PAV)	Bio 70/100	Ref 70/100	Bio 160/220	Ref 160/220	Bio PMB 40/100-75	Ref PMB 40/100-75
G(60 s) at T_{ref} = -12 °C, MPa	62.8	58.2	N/A	N/A	N/A	N/A
m_r(60 s) at T_{ref} = -12 °C	0.332	0.311	N/A	N/A	N/A	N/A
G(60 s) at T_{ref} = -18 °C, MPa	139	119	79.0	52.4	63.6	80.4
m_r(60 s) at T_{ref} = -18 °C	0.268	0.258	0.328	0.358	0.280	0.255
G(60 s) at T_{ref} = -24 °C, MPa	N/A	N/A	167	112	117	142
m_r(60 s) at T_{ref} = -24 °C	N/A	N/A	0.253	0.294	0.229	0.210
T_{cG} @ G(60 s)=143 MPa, °C	-18.2	-19.6	-22.8	-26.0	-26.0	-24.0
T_{cmr} @ m_r(60 s)=0.280, °C	-16.8	-15.5	-21.9	-25.3	-18.0	-14.8
ΔT_c=T_{cG}-T_{cmr}, °C	-1.4	-4.1	-0.9	-0.7	-8.0	-9.3

It is indicated in Table 7 that all binders had negative ΔT_c values (< 0 °C) as determined by 4-mm DSR testing. The bio-extended binders without polymer both had their ΔT_c values higher than -2.0 °C, which is the minimum specification criterion limit (≥ -2.0 °C) adopted by some states in the US for binders after RTFOT and 20 h PAV ageing (some other states adopting ≥ -5.0 °C or ≥ -6.0 °C). The PMB binders had more negative ΔT_c (lower negative values) than the unmodified binders. This is a known phenomenon rising concerns about the applicability of ΔT_c for PMB binders [18]. Between the two investigated PMB binders, the bio-extended PMB had a higher ΔT_c than the reference PMB binder after long-term ageing. As for the critical temperature levels, Table 7 shows that the bio-extended 160/220 binder would not be able to withstand the same low temperatures as its corresponding reference bitumen. But the bio-extended PMB would be able to resist lower temperatures than the reference PMB. No significant difference could be observed in the critical temperatures of 70/100 binders.

As the ΔT_c parameter was originally proposed on the basis of BBR testing and some of the above-described analyses still need a further verification, BBR measurements were conducted on the investigated binders at different low temperatures. The BBR test measures the stiffness and creep behaviour of bituminous binders at low temperatures. It helps determine the low-temperature grade and the critical temperature at which the binder may undergo thermal cracking. Figure 4 presents the BBR measurement results of the investigated binders after long-term ageing. Each binder was tested at two different temperatures. By interpolation or extrapolation, the critical temperatures T_{cS} at which S is equal to 300 MPa at 60 s and T_{cm} at which the m-value is equal to 0.300 at 60 s could be calculated. The difference between them (T_{cS}-T_{cm}) is defined as the ΔT_c value determined by BBR testing. The calculation results of the investigated binders after long-term ageing are shown in Table 8.

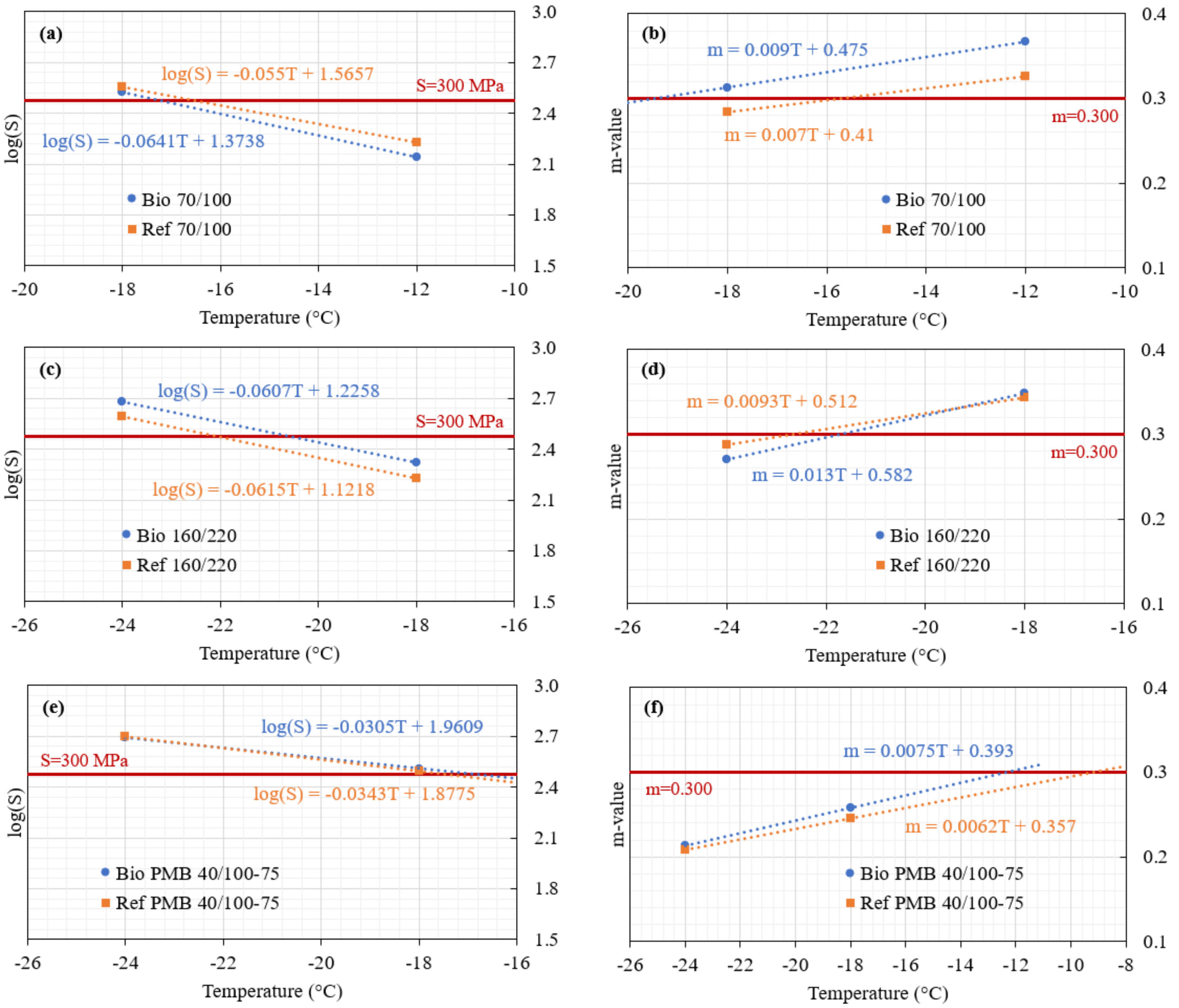

Figure 4: BBR test results at 60 s after RTFOT+PAV – (a) creep stiffness S of 70/100 binders; (b) m-value of 70/100 binders; (c) creep stiffness S of 160/220 binders; (d) m-value of 160/220 binders; (e) creep stiffness S of PMB binders; (f) m-value of PMB binders

Table 8. BBR critical temperatures at 60 s and ΔT_c after RTFOT+PAV

Binders (after RTFOT+PAV)	Bio 70/100	Ref 70/100	Bio 160/220	Ref 160/220	Bio PMB 40/100-75	Ref PMB 40/100-75
T_{cS} @ S=300 MPa, °C	-17.2	-16.6	-20.6	-22.0	-16.9	-17.5
T_{cm} @ m-value=0.300, °C	-19.4	-15.7	-21.7	-22.8	-12.4	-9.2
$\Delta T_c = T_{cS} - T_{cm}$, °C	2.2	-0.9	1.1	0.8	-4.5	-8.3

The results in Table 8 confirms that the bio-extended binders without polymer both passed the ΔT_c criterion (≥ -2.0 °C) after RTFOT+PAV ageing. The PMB binders also showed more negative ΔT_c (lower negative values) than the unmodified binders, as determined by BBR testing. The bio-extended PMB still had a higher ΔT_c than the reference PMB binder after long-term ageing. As for the critical temperature levels, Table 8 confirms that the bio-extended 160/220 binder would not be able to withstand the same low temperatures as its corresponding reference bitumen. But based on the BBR results, no significant difference could be observed in the critical temperatures for the 70/100 and PMB binders.

To have an overall analysis of the low-temperature test results, Figure 5 plots all the critical temperatures as well as ΔT_c criteria in the same space. It shows that all bio-extended binders without polymer are in the "acceptable" side of the plot. The investigated plant-based bio-oil did not affect the ΔT_c negatively and this is valid even for PMB. The bio-extended 160/220 binder shows significantly higher critical temperatures (less negative below 0 °C) than the reference 160/220 bitumen, which is unanimously supported by both the 4-mm DSR test results and the BBR test results. One possible reason for this is that the bio-extended 160/220 binder contains a higher amount of plant-based bio-oil than the other bio-extended binders and the effect of plant-based bio-oil on binder properties would be more significant and easier to capture by the testing.

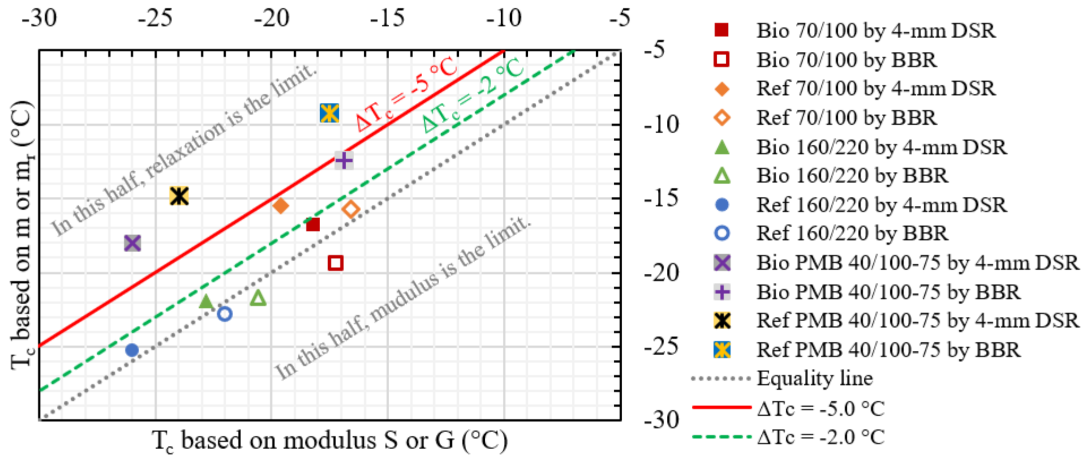

Figure 5: Comparison of critical temperatures and ΔT_c with different methods (all binders after RTFOT+PAV)

3.3. Overall durability assessment

The results described above show that the bio-extended binders passed all the specification criteria after RTFOT+PAV ageing that were discussed in this study, including:

- The intermediate-temperature fatigue criterion based on |G*|·sin(δ) by ASTM D6373-21 and AASHTO M 320-22 (maximum 5000 kPa at 10 rad/s),
- The new fatigue criterion based on Glover-Rowe parameter G'/tan(δ), as proposed by Christensen and Tran [12] (maximum 5000 kPa at 10 rad/s),
- The new fatigue criterion based on R-value as determined with the BBR test results (1.5-2.5) [12],
- For bio-extended binders without polymer, the ΔT_c criterion (≥ -2.0 °C) as the strictest requirement adopted by some states in the US, and
- For bio-extended PMB, a higher ΔT_c than the reference PMB binder.

Additionally, using the investigated plant-based bio-oil in bituminous binders did not affect the ΔT_c or the standard PG low-temperature grade negatively. In fact, the effect on ΔT_c was largely positive. This is valid even for PMB. However, despite the unchanged low-temperature grade, the bio-extended 160/220 binder did show significantly higher critical temperatures (less negative below 0 °C) than the reference 160/220 bitumen, which was unanimously supported by both the 4-mm DSR test results and the BBR test results. This might be related to the relatively high amount of plant-based bio-oil in the binder.

At 10 °C, the G'/tan(δ) value of the bio-extended 160/220 binder was marginally higher than the reference 160/220 bitumen while the other bio-extended binders had marginally lower values than their corresponding reference binders. Meanwhile, all bio-extended binders had lower G'/tan(δ) values than the reference binders over 22 °C. This can be interpreted as a positive sign that bio-extended binders have slightly higher fatigue resistance than the reference binders in the temperature range over 22 °C. But at lower temperatures, especially below 10 °C, this advantage may reduce or even cease. At very low negative temperatures, as mentioned above, bio-extended binders may show higher critical temperatures (less negative below 0 °C). This effect would be more significant for bio-extended binders with a higher amount of plant-based bio-oil and without polymer in the binder.

Furthermore, as suggested by Christensen and Tran [12], there is generally a good correlation between the R-value and ΔT_c of bituminous binders. To verify this correlation even for bio-extended binders, Figure 6(a) presents the linear relationship between ΔT_c of all the investigated binders and their R-values after long-term ageing. The ΔT_c values by both the BBR test and 4-mm DSR test were plotted against the R-values determined by DSR testing over 10 MPa. It shows that the bio-extended binders after RTFOT+PAV ageing follow the same correlation between the indicated parameters as the reference binders do. Since a linear relationship also exists between δ_{T3} and the R-value by DSR testing over 10 MPa, as shown in Figure 1(b), the ΔT_c values of bituminous binders (including bio-extended bituminous binders) can also be correlated to the δ_{T3} – the phase angle when |G*| is equal to 5 MPa at 10 rad/s after long-term ageing. Based on the obtained data in this study, the linear relationship between them is presented in Figure 6(b).

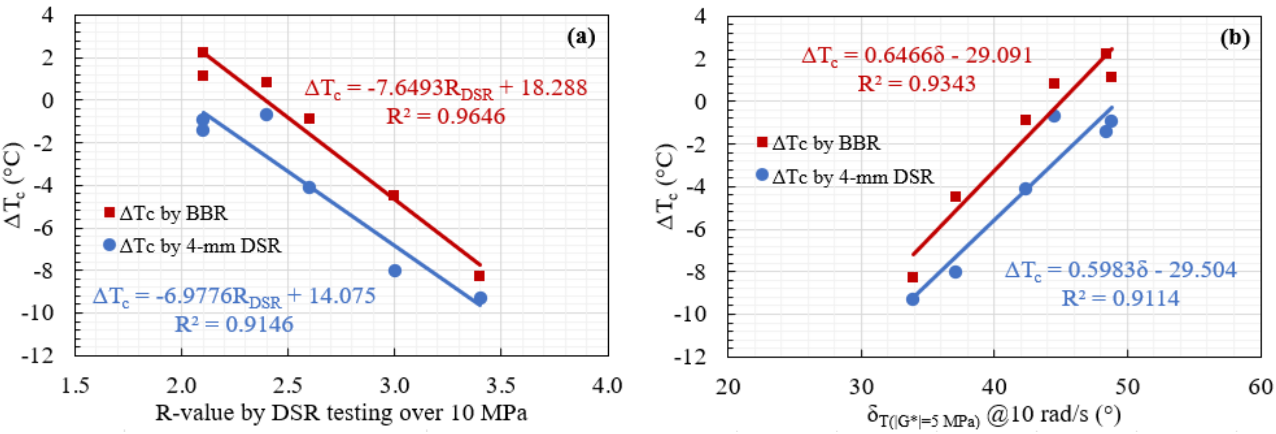

Figure 6: Relationship between different cracking indicators after RTFOT+PAV – (a) ΔT_c versus R-value by DSR testing over 10 MPa; (b) ΔT_c versus δ_{T3}

4. CONCLUSIONS

This study evaluated the durability of bio-extended bituminous binders (both unmodified and polymer-modified) containing a specific type of plant-based bio-oil that primarily consists of high-boiling esters and free acids. Rheological characterisation of binders after long-term ageing was conducted to assess the relaxation property changes and the resistance to both fatigue cracking at intermediate temperatures and thermal cracking at low temperatures. The results indicate that the bio-extended binders passed all the currently available relevant specification criteria after RTFOT+PAV ageing. The use of the investigated plant-based bio-oil in bituminous binders had largely a positive effect on the ΔT_c and did not affect the standard PG low-temperature grade significantly, indicating better thermal cracking resistance for the bio-extended binders at their respective low-temperature grade. The bio-extended binders after long-term ageing followed the same relationships between several durability-related performance indicators as the reference binders did.

Meanwhile, there were signs that bio-extended binders may have slightly higher fatigue resistance than conventional reference binders in the temperature range over 22 °C. But when the temperature decreases to the level below 10 °C, this advantage may reduce or even cease. At very low negative temperatures around the PG low-temperature grade, bio-extended binders may show higher critical temperatures (less negative below 0 °C) and would not be able to withstand the same low temperatures as the corresponding reference bitumen regarding the thermal cracking resistance. This effect would be more significant for bio-extended binders with a higher amount of plant-based bio-oil and without polymer in the binder. However, it is worth noting that this study conducted analyses only on the binder scale. A verification on the asphalt mixture scale will be necessary to assess if the above-mentioned effects of the investigated plant-based bio-oil on bituminous binders are significant enough to impact the asphalt mixture performance in practical applications.

Last but not least, it should be noted that different types of bio-oils have different chemical compositions, and they may have different effects as extenders in bituminous binders. This study investigated a specific type of plant-based bio-oil that is derived from bio-based by-products from the pulp and paper industry and contains primarily high-boiling esters and free acids. The conclusions drawn from this study are valid for this specific bio-oil and likely also for bio-oils with a similar chemical composition. However, for bio-oils with very different chemical compositions, additional test data would be necessary when generalising the related conclusions.

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