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Manufacturing terminal and field bitumen-tyre rubber blends: the importance of processing conditions

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Abstract

No-agitation Tyre Rubber Modified Bitumens (TR-MBs), also known as “terminal blends” or “field-blends”, are quite new technologies leading to bituminous binders similar to polymer modified bitumens in terms of both rheology and needed efforts during paving operations. Their manufacturing, as well as other TR-MBs, is strongly dependent on the selected processing variables as well as on the chosen materials. This study presents a literature review to provide an overview of the TR-MB technologies which focuses on the influence of processing conditions on the modification process, storage stability and overall properties of the final bitumen - tyre rubber blends.

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Keywords: tyre rubber; rubberised bitumen; processing conditions; wet process; terminal blend; field-blend

1. Introduction

The use of Tyre Rubber (TR) in asphalt mixes has been in use in the USA since at least the early 1960s by the Arizona Department of Transport. Since then the material has been used more widely in four states in the US and elsewhere, including South Africa, Europe and Australasia. There are two main processes of using rubber in asphalt, i.e. the dry and the wet process. Dry process consists in the production of asphalt mixture with the addition of a small ground rubber quantity from discarded tyres as a substitute for a part of the mineral aggregate, while the method of modifying bitumen with TR and other components, in order to obtain a new binder (TR-MB) which captures beneficial engineering characteristics of both base ingredients, is referred to be as the “wet process”. Nevertheless the term “wet process” refers to a whole family of technologies.

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In fact, the uses of TR as modifying agent in bitumen leads to blends with different compositions, method of production or preparation and with diverse physical and structural properties. The original wet process, invented by Charles McDonald, leads to a product with a series of benefits which are basically all linked to the binder’s increase in elasticity and viscosity at high temperatures that allow greater film thickness in paving mixes without excessive drain down or bleeding. A number of TR-MBs and mix specifications have been developed based on this original idea and each of them is related to a specific technology. Table 1 lists the most commonly used technologies/specifications in this area. Wet process is obtainable through two different processing systems: the original “McDonald system” that includes a high shear mixing vessel and separated storage agitated tanks to allow the digestion of TR into the bitumen through a continuous low shear stirring, and the “Continuous blending system”, that consists of a unit with agitators, in which the reaction occurs during the blending [1].

Depending on the adopted processing system, on the chosen processing conditions, and on the selected materials, the wet process leads to different technologies. Caltrans [2], makes an important distinction among the various blends related to the rotational viscosity of the resulting TR-MBs at high temperature. Based on this distinction. It is possible to divide the wet process into two families linked to two very different types of TR modification currently in use: the “wet process–high viscosity” and the “wet process–no agitation” also known as “terminal blend”. To promote clear understanding, definitions of the processes and technologies linked to TR-MBs are explained below.

Table 1 TR-MB technologies around the world

Technologies	Specifications	Country
Asphalt Rubber binder	ASTM D6114-97, 2009	USA
	Caltrans' Asphalt Rubber User Guide, 2006	
Bitumen Rubber binder	SABITA Manual 19, 2007	RSA
	AsAc Technical Guideline, 2007	
Crumb Rubber binder	Austroads, 2006	AUS & NZ

1.1. Wet process High-Viscosity

TR-MBs that maintain or exceed the minimum rotational viscosity threshold of 1,500 cPs at 177°C (or 190°C) over the interaction period should be described as “wet process–high viscosity” [2]. These materials require continuous agitation, with special equipment, to keep the TR particles uniformly distributed. They are based on the original wet process and may be manufactured with either McDonald or the continuous blending systems. Wet process-high viscosity binders typically require at least 15% TR to achieve the threshold viscosity. However, for some specifications the viscosity requirements are met also at lower TR content [2]. Wet process-high viscosity binders include not only asphalt rubber binder that meets the requirements of ASTM D6114, but also other technologies and specifications, such as bitumen rubber binder [3], used in South Africa, and Crumb Rubber Modified Binder [4], developed and commonly used in Australia (Table 2). Extensive literature [5], [6], Bahia & Davis, [7], clearly show the numerous successes obtained when using TR bituminous mixtures produced with the Wet Process. However, wet process is not a panacea. There are drawbacks to it, chiefly in terms of its workability [2], its recyclability [8], binder storage stability [9], [10] and the fumes it emits during the paving process [11]. Bitumen-rubber pavements are also more expensive than conventional asphalt ones [12] and can be difficult for paving contractors not accustomed to working with this product, which must be placed and compacted within a relatively narrow temperature window [13]. For these reasons, in the USA in the early 90s, this technology was left as dead, but when in 1991, the United States federal law named “Intermodal Surface Transportation Efficiency Act” (recently rescinded), mandated its widespread use, the TR-MB technology concept started to make a “quiet come back” [13]. Furthermore in 1992 the patent of the McDonald’s process expired and now the material is considered to be a part of the public domain. Since then, considerable research

has been done worldwide to improve the properties of TR-MB in order to try solving the issue related to its production and usage. In Europe, rubberised asphalt has been used since 1981 in Belgium, as well as in France, Austria, Netherlands, Italy, Germany and Sweden, and more intensively since 1999 in Portugal and Spain [14].

Table 2. Summary of the specification requirements for TR-MBs

Properties	ASTM 2002			Caltrans 2006 (USA)	SABITA 2007 (RSA)	Austroads 2007 (AUS)
	Type I	Type II	Type III			
Base bitumen requirements						
Penetration: dmm	85-100	120-150	200-300	120-150	60-100	85-100
Crumb rubber requirements						
Passing sieve: mm	2.36			2.36	1.18	2.36
Rubber content: %	≥15			18-22	18-24	15-18
Additives						
Extender oils: %	-			2.5 - 6	0 - 4	-
calcium carbonate /talc: %	0 - 4			-	0 - 4	-
Processing conditions						
Temp.: °C	177			190-220	180-220	195
Mixing speed: rpm	-			-	3000 rpm	-
Mixing time: min	45 + reaction			45-60	-	30-45

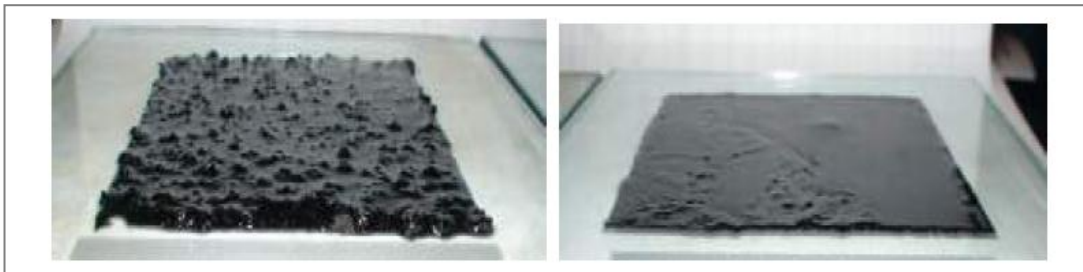


Figure 1. TR-MB from wet process- high viscosity (left) and wet process- no agitation (right)

1.2. Wet process NO-Agitation

The Wet process-no agitation technology was first used in Florida and Texas in the mid-1980s and till nowadays it seems to have been used only in some parts of the USA, namely California, Colorado, Louisiana, Arizona, and Nevada. The idea with TR-MB produced with the NO agitation-wet process is to produce a material in which the TR is fully digested, or better dissolved, into the bitumen without leaving visibly discrete particles. This leads to obtain TR-MBs that do not require constant agitation to keep discrete TR particles uniformly distributed in the hot bitumen.

The characteristic swelling process of the wet process-high viscosity is replaced by the depolymerisation/devulcanisation and the optimised dispersion of the TR into the bitumen by using high processing temperature (200-260°C) and high shear stress during the mixing (up to 8000 RPM), resulting in a smooth, homogeneous product. The term “terminal blend” is used when TR is digested into the bitumen at the refinery and is delivered to the plant in the same manner as conventional bitumen without rubber. However, such binders may be produced with similar procedures directly even on the field. To avoid misleads and restrictive terminology, in this case these binders are denominated “field blends” [15]. In this process, no modifications to the asphalt plant are required. These binders are typically modified with very fine TR particles that can be digested (broken down and melted in) relatively quickly and/or can be kept dispersed by normal circulation

within the storage tank rather than with agitation by special augers or paddles. Although such binders may develop a considerable level of rubber modification, rotational viscosity values rarely approach the minimum threshold of 1,500 cPs (1.5 Pa.s) at 177°C, that is necessary to significantly increase binder contents above those of conventional asphalt mixes without excessive drain down [2]. Wet process-no agitation TR-MBs seems to solve most of the limitations linked with the wet process. However, terminal and field blends are quite new technologies which are in constant development. Furthermore their manufacturing and properties are strongly dependent from the selected processing variables as well as from the chosen materials.

2. The importance of processing conditions

The nature of the mechanism by which the interaction between bitumen and TR takes place, has not been fully characterised. Traditionally it is reported that bitumen-rubber interaction is not chemical in nature [1] but other studies claim that the increase in binder viscosity cannot be accounted for only by existence of the TR particles swelling [7].

The reaction itself is made up of two simultaneous processes: partial digestion of the rubber into the bitumen on one hand and, on the other, adsorption of the aromatic oils available in this latter within the polymeric chains that are the main components of the rubber, both natural and synthetic, contained in the TR. The absorption of aromatic oils from the bitumen into the rubber’s polymer chains causes the rubber to swell and soften [16], [17]. TR particles are swollen (Fig. 2bII) by the absorption of the bitumen oily phase at high temperatures (160–220°C) into the polymer chains, which are the key components of the TR-MB to form a gel-like material. Therefore, during the reaction there is a contemporaneous reduction in the oily fraction and an increase of rubber particle sizes with a consequent reduction of the inter-particle distance (Fig. 2cII). This implies the formation of gel structures that produce a viscosity increase up to a factor of 10 [17], [1], [7].

Rubber reacts in a time-temperature dependent manner. If the temperature is too high or the time is too long, the swelling will continue to the point where it is replaced by depolymerisation/devulcanisation (Fig. 2bIII) which cause dispersion of the rubber into the bitumen (Fig. 2cIII). Depolymerisation starts releasing rubber components back to the liquid phase (Fig. 2bIII) causing a decrease in the stiffness (G^*) while the elastic properties (δ) continues to modify (Fig. 2aII). If temperature is high or time is long enough, depolymerisation will continue causing more destruction of the binder networking (Fig. 2cIV) and so δ modification is lost (Fig. 2aIII), [18].

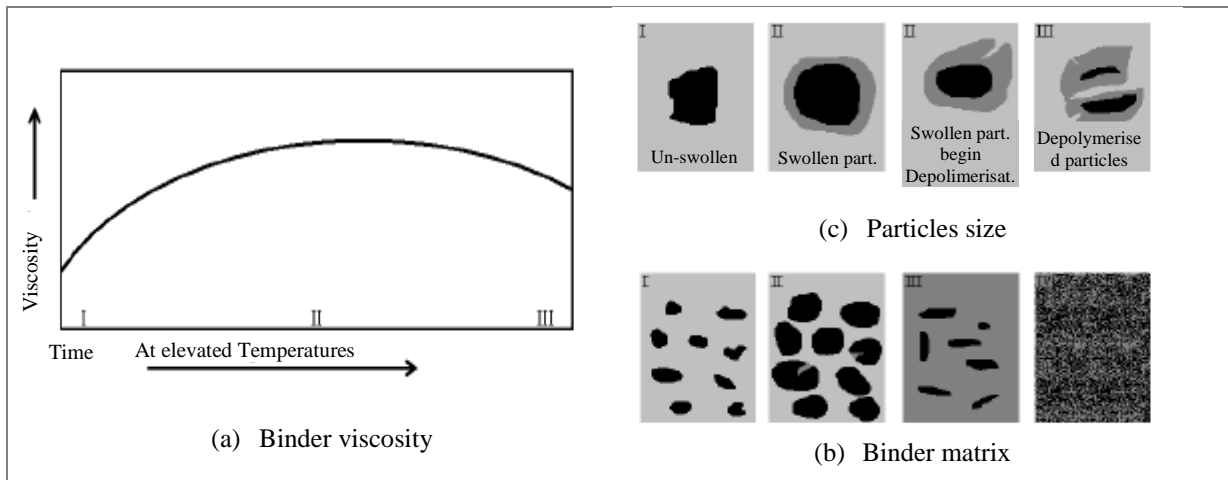


Figure 2 Bitumen-TR interaction phenomenon at elevated temperatures: change of properties over time.

The interaction between bitumen and rubber materials is material-specific and depends on a number of basic factors, including:

- Processing Variables: Temperature, Time and Device (applied shear stress)
- Base binder properties: Bitumen Source and eventual use of oil extenders
- Tyre Rubber properties: Source, processing methods, particle size and content

All these variables represent the processing conditions that need to be monitored in order to govern the modification process. This section aims to discuss each single factor in order to better explain their role in the modification process as well as the properties of the final blends

2.1. Processing variables

Due to the variety of materials utilised in TR paving materials, there are numerous issues relating to their selection and design of TR-MBs. Physical and engineering properties of modified binders are highly dependent on the unique interactions between the component materials: bitumen and TR. These interactions depend on their respective chemicals, but processing conditions such as temperature, interaction time, shear stress and pressure have a primary importance as well as in any mixing process [19].

Processing temperature

The final properties of TR-MBs are largely dependent on processing temperature. As reported by Navarro et al. 2004, the TR solubilised in the bitumen phase remains constant and equal to the initial value for processing temperatures comprised between 90 and 120°C. This happens because these conditions are not severe enough to break up the chemically cross-linked network. Thus, the observed rheological behavior would be only a consequence of the presence of TR particles swollen by light components of the maltenic fraction. Higher processing temperatures (160-180°C) lead to a partial depolymerisation/devulcalisation of the rubber network, increasing the amount of components that are incorporated to the bitumen phase (bitumen matrix) and, consequently, reducing both solid concentration and rubber particle size [20]. Various experiences [20], [4] showed that when 15% of TR is added, only a percentage ranging from 2 to 4 is dissolved or dispersed in the bitumen. As regards to the physical interaction process, Green and Tolonen [21] concluded that processing temperature has two effects. The first effect is on the rate of swelling of rubber particles. As the temperature increases, for example from 160 to 200°C, the rate of swelling increases.

The second effect is on the extent of swelling. As the temperature increases, the extent of swelling decreases. By increasing processing temperature, the dissolution/dispersion of TR into the bitumen is clearly enhanced due to the faster rates of breaking crosslink bonds.

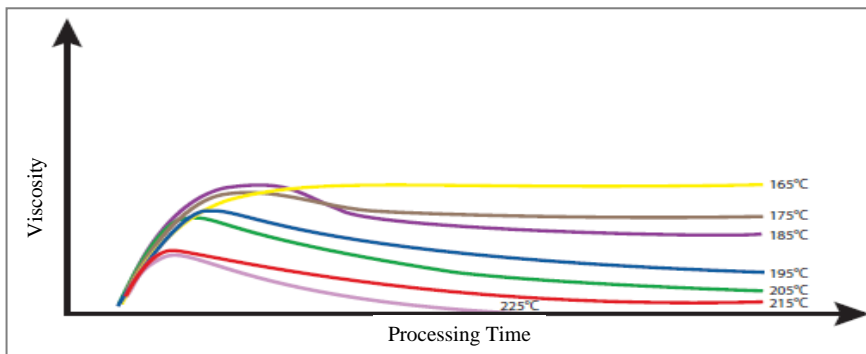


Figure 3 Typical changes in viscosity values from bitumen rubber at different temperatures over time (adapted from [3])

Lalwani et al. [22] concluded that because of the depolymerisation/devulcalisation of the rubber network the binder elasticity was drastically reduced by as much as three times when the temperature was increased from 200 to 300°C, while no significant differences occurred due to changing the temperature from 150 to 200°C. Although, a recent collection of studies on “high-curing” of TR-MBs [23] reports that a processing temperature below 232 °C (400°F) is significantly less efficient at curing the bitumen-rubber blends, while though more data are needed, there is evidence that curing temperatures at 260 °C (500 °F) would give a product with excellent settling properties and very high performance grade. However, high processing temperatures might lead to certain bitumen oxidation or primary aging [20].

Processing time

Rubber reacts in a time-temperature dependent manner, so the two processing conditions are strongly correlated (Fig. 2). The effect of interaction time on the developed properties is a function of the interaction temperature. Two main phases of the property development are considered: an initial phase or short term phase that usually last for 30 to 40 minutes but that varies with temperature and a second long-term phase that last for few hours. Most of the changes occur in the initial phase. Modification occurs in the first phase, properties stabilise in the second [18]. In general, the longer reaction time for digestion of the rubber-modified binder seemed to lead to an increase in the viscosity, which is related to the increase in the rubber mass through binder absorption. Oliver [17] indicated that increased digestion time (up to 2 hours over a temperature range of 180°C to 200°C) improved the rubberised binder properties. However, various sources demonstrated that prolonged digestion up to 24 hours was reported to have caused reduction in viscosity and elastic response, suggesting some undesirable rubber degradation.

Furthermore, rubber degradation takes place more rapidly at higher digestion temperatures [17], (Fig.3). In fact depolymerisation cracks the binder networking into lower molecular weight molecules. Depolymerisation starts very early at high interaction temperature and continues up to full destruction of the polymer network, if the binder is exposed to very high temperature for enough time (>210°C), [18], [24]. Recent studies on terminal blends show that 4-6 hours of curing a TR-MB at very high temperatures (230-260°C) significantly increase the settling properties of the blend by maintaining a high grade binder [21].

Processing Devices (applied shear stress)

As reported by Navarro et al. [20] the rheological behavior of TR-MBs is not influenced by the processing device and impeller geometry used for their manufacture at 180°C. Conversely, other authors have found a clear dependence of binder properties from the mixer speed and the applied shear rate. In fact, there are two type of mixing: (1) distributive mixing and (2) dispersive mixing (Fig. 4). Distributive mixing refers to uniform distribution of different components in space, and it does not require a high stress. Dispersive mixing refers to reduction of component size, and it occurs only when the stress in the melt exceeds the coherent strength of the component. In reality, distributive mixing involves some dispersive mixing and vice versa [19].

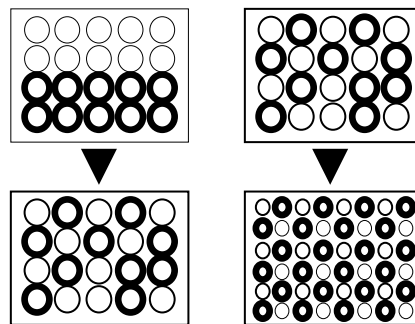


Figure 4 Distributive mixing (left) and Dispersive mixing (right)

Therefore, producing TR-MB with high shear stress, applied by using colloid mill equipped with disintegrating head at high speed (5000-8000 RPM), implies a dispersive mixing which reduces the particle size of coarse TR, allowing the interaction process to progress with greater speed [18], [23]. Agglomeration is another mechanism that can contribute to the increase of stiffness/viscosity during the modification process [25]. Shenoy [25] stated that high shear mixing provides the energy to break particle-particle bonds and helps reducing agglomeration. Once the polymer wets the particle, the bond fails to re-form, leading to better dispersion of the rubber within the filler in the polymer matrix [26]. High shear rate are fundamental to ensure proper dispersion of the rubber within the bitumen matrix and it has been demonstrated that this helps the storage stability of the final blend [25]. However, the processing device is secondary to the processing temperature; if the temperature is not high enough to produce TR depolymerisation/devulcanisation to a great extent, the influence of processing device is not significant [20].

2.2. Base binder properties

It is evident that the preparation of bitumen-rubber blends is strongly related to the nature and the overall properties of the raw components. However, the base bitumen composition is considered as a key factor influencing the final rheological properties of TR-MBs [18], [24]. In fact, both bitumen and TR are said to affect the properties of the modified bitumen and those of the mixes. However, the properties of the base binder typically have a greater influence on the rutting susceptibility and viscosity of the binder than the properties of the TR used in the matrix [27].

Bitumen source (physical and chemical properties)

Regarding the compatibility between base bitumen and the TR crumbs, the evolution of the bitumen composition during the curing process indicates that aromatic fractions of bitumen are the major cause of rubber swelling through a migration phenomenon. In fact, the miscibility with rubber of the oils used in the rubber processing industry diminishes in the order aromatic > naphthenic > paraffinic [28].

Viscoelastic properties measurements demonstrate that base bitumen composition should be selected carefully to achieve a good compromise between a sufficient rubber swelling and a reasonable viscosity of the final binder. [29]. Thus, in order to optimise the modification process, it is suggested to use bitumens that are particularly rich in aromatics and resins or, if necessary, to add aromatic oils in order to favor swelling of the TR crumbs. For this reason, all specifications in table 2 prescribe quite soft base bitumens, i.e. with high penetration grade.

Oil extenders

The production of TR-MBs can have an undesirable side effect, i.e. the absorption of aromatic oils from the bitumen into the rubber's polymer chains could require an increase in the binder content, which may lead to potential problems of flushing or bleeding, increasing the paving material's cost, and may cause tracking. In this case an external source of light fractions (extender oils) can be used to eliminate issues of compatibility. However, extender oils are expensive and typically increase emissions of aromatic and volatile compounds at high temperatures [2]. The type and amount of aromatic oil in the bitumen also plays a major role in determining the compatibility of bitumen-rubber blends [16].

2.3. Tyre rubber properties

Depending upon the type of TR, rubber processing method and rubber particle size used, the blending method is said to vary with the variation in the results of the end product. Furthermore, the viscosity is strongly related to the size of the particles and relative TR content the final blends [2].

Tyre Rubber Source

Generally it is assumed that the mixes made from automobile tyres differ from those made with truck tyres [21], [7]. It states that the difference in terms of the viscosity, ring and ball softening point and ductility, can in part be related to the chemical balance of rubber in the two tyre types (Fig. 5). The constituent of TR known to affect the blend behaviour is the natural rubber component. Glenn & Tolonen [21] indicate that whole truck tyres contain approximately 18 percent natural rubber compared with 9% for whole automobile tyres and 2% for automobile tyre treads. Therefore, when combinations of bitumen and TR materials cannot produce a satisfactory blend, it is possible to enhance the content of natural rubber to eliminate issues of compatibility.

Huff and Vallerga [30], indicate that bitumen-rubber prepared with vulcanised synthetic rubber (scrap tyres) indicated better weather and heat resistant properties when compared to the non-vulcanised rubber. The vulcanised rubber is said to form a bitumen-rubber sheet due to the swelling of rubber after absorbing the oils in bitumen. This is said to impart better resistance to fracture under traffic. The bitumen-rubber prepared with devulcanised rubber indicated better dispersion and dissolution in bitumen and better binder properties (adhesion and cohesion). However, these blends are reported to lack the toughness and resilience achieved with the vulcanised bitumen-rubber blends.



Figure 5 SEM micrographs of Ambient CRM (left) and Cryogenic CRM at 30x magnification (adapted from [27]).

Tyre Rubber processing

The method adopted to process the scrap tyres significantly affects the digestion of TR and the intermediate to high temperature properties of bitumen-rubber and its mixes [18], [24]. Oliver, [17] reports that an electron micrograph study on the rubber particles indicated that the rubber processing method affects the rubber size and shape of rubber particles (Fig. 5). The processing method, therefore, affects the surface area of the rubber particles, which in turn affects the rate of reaction and viscosity of the bitumen-rubber binder. A study conducted by Texas Transportation Institute theorised that the increased surface area per volume or weight of the fine particles enhances the ability of the particles to be swollen by, and thus bond with, the binder [23].

Tyre Rubber particle size and shape

TR particle size controls the swelling mechanism over time and affects the binder matrix. Buckley and Berger [31] showed that time required for swelling increases with the particle radius squared. Fine rubber swells faster and depolymerises faster, affecting the liquid phase more than the matrix of the binder. Fine particle sizes may require almost no time to react. Coarse rubber has more effect on the binder matrix but has less effect on the liquid phase than fine rubber. Liquid-phase modifications are more stable than matrix modifications [18], [24].

Another study stated that the coarser the particle size the higher the G^* value and the lower the δ under the same interaction conditions [25]. Moreover, different authors found a dependence of the particle aspect ratio (length/diameter) on the viscosity of TR-MB, so that higher aspect ratios lead to higher viscosities [32]. Settling properties are highly improved when smaller starting mesh sizes are used. Nevertheless, TR particle size has a minor influence on settling properties when high curing conditions are used, especially at high temperatures [23].

Tyre Rubber content

A bitumen modified with 15 percent TR can increase the high temperature viscosity of the blend by a factor of 10 or more. There is evidence to suggest that increasing TR concentrations, regardless of TR source or binder type, results in increasing viscosities, failure temperatures, and $G^*/\sin\delta$ values. This confirms that as TR is added to the binder, the mechanical properties and viscosity are increased; however, the extent of this increase is dependent on the type of binder and TR properties [27]. Higher TR content implies longer curing time and higher separation of the particles over storage time. However, high curing conditions reduce processing time and improve storage stability of the TR-MB [23].

3. Stabilisation of settling properties

The wet process-no agitation aims to produce bitumen-tyre rubber blends which do not occur in eventual phase separation of the TR from the binder during storage or transportation. As discussed in previous sections the improvement of the settling properties of rubberised binders is often linked to a high level of solubilisation of the TR within the bitumen matrix which occurs only at high temperatures and long processing time. However, at this stage the effect of the modification is significantly reduced. In fact, TR is shown to a number of studies to effectively modify the bitumen when it is still in the swollen state (Fig.1bII) and not completely devulcanised/depolymerised (Fig.1bIII). In order to maintain a convenient level of modification, recent studies have shown that is possible to improve the storage stability of TR-MBs by using high-curing conditions [23], by adding substances to operate a chemical stabilisation of the blends [33] and also by mixing rubberised bitumen blends in presence of a low percentage of polymers [26], with compatibiliser to activate the TR [10], or with sulphur to improve crosslinking [34]. Furthermore, Attia & Abdelrahman [26] suggest improving the stability of modified binders also by lowering the storage temperatures. In any case, the accurate selection of processing conditions covers the main role also in improving the settling properties of bitumen-TR blends.

4. Conclusions

Tyre Rubber Modified Binders (TR-MBs), produced through McDonald wet process and used worldwide (e.g. asphalt rubber), have been demonstrated to provide various benefits to pavements, however this technology is still struggling to be fully adopted in Europe, mainly because of their poor stability during high temperatures storage, which leads to high initial costs in modifying existing asphalt plants. The authors believe that “terminal blends” or “field-blends” are one of the key to boost the usage of rubberised pavement also in Europe.

This study highlights that both type of TR-MBs are extremely dependent on the processing conditions, particularly to what concerns the temperature and time of reaction, and their variation can lead to obtain a technology rather than another. With regards to the storage stability, even when other polymers, cross-linkers or other kind of additives are added into the blend, the processing conditions still play the main role.

TR-MBs must be properly designed and, where necessary, manufactured to comply with specifications and provide a quality product suitable for the expected climate and traffic conditions. Caltrans [2] as well as Sabita [3] and Austroads [4] are fully aware of the primary importance of governing modification process; for this reason all of them specify compulsory prior laboratory testing and a constant control of the binders properties during paving operations. Only an accurate selection of the processing variables and materials could lead to a superior binder design.

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