

Delft University of Technology

# The potential of Altraflex-2006 to improve low temperature behaviour of C-fix mixtures

M.Sc. Thesis Fábio Arce

December 2007



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#### **Mater's Thesis**

This is the study performed as a partial fulfilment of the requirements for the degree of Master of Science in Road and Railway Engineering at Delft University of Technology.

#### **Graduation Committee:**

Prof.dr.ir. A.A.A. Molenaar	Civil Engineering and Geosciences, Road and Railway Engineering, TU Delft;
Ir M.F.C. van de Ven	Civil Engineering and Geosciences, Road and Railway Engineering, TU Delft;
Dr.ir. A.L.A. Fraaij	Civil Engineering and Geosciences, Material Science and Sutainable Construction, TU Delft;
Ir. R.N.Khedoe	Civil Engineering and Geosciences, Road and Railway Engineering, TU Delft;
Drs. J.P.Wortelboer	Altavie;
G. van Emst	C-fix.

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# Abstract

Porous Asphalt Concrete (PAC) has been intensely used in the Netherlands as the wearing course aiming the reduction of noise emission, the decrease of splash and spray improving visibility during the rain, and diminishing the risk of aquaplaning. Due to its very open structure, with voids contents between 20 and 27%, the rainfall drains quickly improving driving safeties.

The drawback of PAC comes exactly from its very specific characteristic: the high void content. This high porosity not only permits the water to pass through the pavement layer, but also oxygen, salts, pollutants and sunlight increasing a lot the ageing rate of the binder. This leads to low ravelling resistance of the PAC mixture, which explains the short life span of a PAC wearing layer.

PAC produced with C-Fix is one of the solutions studied for the short life span presented in the wearing course. C-Fix has appeared in the market few years ago with an environmental friendly appealing and as a binder with good physical characteristics. Indeed, the binder has proven to be suitable for many applications not used before for a sort of asphalt binder.

Although some researchers were positive about the possible use of C-Fix in PAC mixture, there were important restrictions: the C-Fix binder has a brittle behaviour at low temperatures which could compromise its use in areas with cold winters such as the Netherlands. Also the relaxation of traffic and thermal loading are very low compared with B70/100. These restrictions to the use of C-Fix binder in PAC mixture at low temperature conditions fomented the idea of modifying it with addition of a polymer. The modifier (including SBS and other substances) is called Altraflex-2006. The polymer used in the modifier is a styrene-butadiene-styrene branched polymer.

The results on the tests done in the modified binder showed some improvements in the penetration values, extra ductility (higher strain reachable level), higher toughness for all the temperatures tested, lower complex shear modulus and extra relaxation. In general, the results could point to an improvement in the C-Fix binder properties regarding the brittleness. Less cracking sensibility could possibly be expected due to the addition of Altraflex-2006. Altraflex-2006 modified C-Fix would possibly be more suitable for PAC mixture than the pure C-Fix, although still high transition temperature is present, possibly leading to an earlier brittle behaviour when compared with bitumen.

Filler type investigation showed that Duras filler 15 seems to give an average better result in relaxation compared with Wigro 60K filler, although higher transition temperature was found. The toughness performance of the Duras 15 mastic was relatively comparable with Wigro 60K, with even better results at higher temperatures.

A considerable ageing was measured, even if C-Fix has better ageing properties than B70/100. Comparison with B70/100 in this research was not possible, but the addition of Altraflex-2006 could result in longer lifespan of the binder if comparison of the 1000 hours aged 6% Altraflex-2006 modified C-Fix binder is made with the pure unaged C-Fix master curves.

The results are promising but not enough yet to support the use of the Altraflex-2006 modified C-Fix as a binder for PAC mixture. The transition temperature is still reasonably high. The reduction of the phase angle in the 12% modified binder (and even higher in the 6%) does not reach higher frequencies in the DSR master curves that could possibly mean a less viscous behaviour at lower temperatures. Due to the low relaxation, also a residual stress is expected to develop in the mastic, even if this point has improved with the use of Duras 15 compared with Wigro 60K. Comparison with other types of polymers as well as PAC mixture tests using the modified binder are suggested to further assess the influence of Altraflex-2006 in the C-Fix binder.

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# 1. Introduction

C-Fix has been introduced in the market a few years ago with an environmental friendly appealing and as a binder with good physical characteristics. Indeed, the binder has proven to be suitable for many applications not suitable before for a sort of asphalt binder. Porous Asphalt Concrete (PAC) is one of the possible applications studied.

PAC is intensely used in the Netherlands as the wearing course aiming the reduction of noise emission, the decrease of splash and spray improving visibility during rain, and diminishing the risk of aquaplaning. Due to its very open structure, with voids contents between 20 and 27%, the rainfall drains quickly improving driving safety.

The drawback of PAC comes exactly from its very specific characteristic: the high void content. This high porosity not only permits water to pass through the pavement layer, but also oxygen, salts, pollutants, oils and sunlight increasing the rate of ageing of the binder. This leads to a low ravelling resistance of the PAC mixture, which explains the relative short and scattered life span of a PAC wearing layer.

#### 1.1 – Motivation of the research

Improving the ravelling resistance of the PAC wearing courses would bring a great improvement on the life span of PAC layers. This has been investigated by using a harder binder than the one prescribed in the Dutch RAW standard [6], the B70/100. C-Fix has been assessed as a possible binder for PAC mixture. One of the reasons for this is the higher viscosity resulting in less drainage of binder and mastic during production, transportation, layering and compaction, making the mixture as a whole less sensitive to weathering due to a more regular binder distribution. Furthermore higher bitumen content can be used. Also, mixtures with C-Fix have shown to have a higher tensile strength [3].

Khedoe [3] and Woldekidan [24] investigated the possibility of using C-Fix as the binder in PAC. Several tests have been performed in order to evaluate the C-Fix suitability in the PAC production.

Although both researchers were positive about the possible use of C-Fix in PAC mixture, there were important restrictions: "In Porous Asphalt surface layers in warm and humid conditions the C-Fix binder is a good alternative. But in the Dutch situation the application of C-Fix binder in a Porous Asphalt Concrete mixture is still questionable. The low temperatures still can cause problems for the C-Fix mixture. The material becomes too brittle. The stress in the material at low temperatures decays not that fast, this can cause high stresses in the PAC when subjected to traffic loading" [3].

These restrictions to the use of C-Fix binder in PAC mixture at low temperature conditions fomented the idea of modifying it with a polymer. The modifier (including SBS and other substances) is called Altraflex-2006. The polymer used in the modifier is a styrene-butadiene-styrene branched polymer.

### 1.2 – Research problem definition

This research aims to assess the improvements that could be achieved by the modification of C-Fix with the proposed modifier named Altraflex-2006 and to check the applicability of Altraflex-2006 modified C-Fix as a binder in PAC. Positive points by modifying C-Fix with Altraflex-2006 could be:

- lowering of the mixing temperature;
- improvement of the low temperature behaviour of C-Fix binder;

- relaxation properties could be improved, leading to less residual stresses due to traffic loads;
- adhesion properties could be improved;

#### 1.3 – Research plan

The test flow-charts are presented here for easier and clearer visualization of all the tests that were performed in this investigation. The research was basically divided in two phases. Tests involving the best dispersion of the modifier Altraflex-2006 into the binder C-Fix were performed in the first phase. The main question at this phase was the amount of Altraflex-2006 to be used in the C-Fix modification. First literature research was done aiming to have an overview of the polymer/bitumen modification proportions currently in use. Apart of the amount to be added, the practical feasibility of the mixture C-Fix and Altraflex-2006 had to be assessed. Microscopic investigations followed by conventional bitumen tests such as the determination of Penetration and Ring&Ball were carried out. In addition, DSR rheological tests were performed with pure and modified C-Fix. See Figure 1 for details in the variables of those tests.



Figure 1 - Phase 1 flow-chart tests.

In the second phase of the research, the main focus was in the filler type to be used together with the Altraflex-2006 modification in C-Fix. Two different filler types were investigated and also two different filler/binder ratios. Tests were performed with mastic to determine the best filler type and filler/binder ratio. In parallel, the same kind of tests with pure and 6% Altraflex-2006 modified C-Fix binder were also performed for possible comparison. Also, the fatigue behaviour of the C-Fix binder was checked as well as for the 6% Altraflex-2006 modified C-Fix. In addition to this, ageing resistance of the 6% modified binder and mastic was carried out. See Figure 2 for details of the tests and the variables investigated.



Figure 2 - Phase II flow-chart tests

#### **1.4 – Report structure**

Information and knowledge about C-Fix, Altraflex-2006, fillers and related materials that could help in understanding the assessed subjects are presented in chapter two of this report. Chapter three discusses the tests performed on the binder. Chapter four deals with the tests results and analyses, culminating then in chapter five with the conclusions and recommendations.

## 2. Definitions

In this chapter, the materials used in the research are defined, discussed, and evaluated. Comparison of C-Fix binder with some commonly used bitumens helps to understand difference between them. Some knowledge on polymer and polymer modified bitumen shall be presented as well in order to clarify what could be expected when modifying C-Fix with a SBS modification polymer. Also the properties and effects of different filler types will be discussed.

## 2.1 – C-Fix

### 2.1.1 – Origin and properties of C-Fix

C-Fix as well as bitumen come from different distillations from the same crude oil. Crude oil is a complex mixture of hydrocarbons differing in molecular weight and, consequently, in boiling range. The heaviest fraction taken from the crude oil distillation (fractional distillation) process is known as the long residue, a complex mixture of high molecular weight hydrocarbons, which requires further processing before it can be used as a feedstock for the manufacture of asphalt binders [1].

The long residue is further distilled at reduced pressure in a vacuum distillation column to produce gas oil, distillates and short residue. The Short residue is the feedstock used in the manufacture of over 20 different grades of bitumen. The viscosity of the short residue is a function of both the origin of the crude oil and the temperature and pressure used in the vacuum column during processing.

As a by-product of the refined crude oil, a very dense and carbon rich residue is produced. This is used to create a very low quality fuel used normally in intercontinental shipping vessels or thermo-electric power stations. Burning this fuel consequently results in a high CO<sup>2</sup> emission.

At Shell this by-product was used as feedstock to produce a binder for mineral aggregates. This carbon no longer is burned and brought to the atmosphere, but fixed in the binder, protecting and keeping the environment clean. Asphalt specimens that are produced appear to have a flexural strength in three-point bending test at room temperature of 4-9 MPa, which is similar to the flexural strength of cement concrete, 5-9 MPa. These values are not observed for asphalt mixtures with conventional bitumen at room temperature [1].

So, C-Fix is a thermoplastic binder for the construction industry that, mixed with aggregates, sand and filler materials, combines the qualities and advantages of asphalt (flexibility) with cement concrete (strength). A unique characteristic of using C-Fix in asphalt mixture is that its use reduces CO<sub>2</sub>-emissions significantly [1].

	Bitumen Pen 100	Bitumen Pen 10-20	C-Fix
Penetration 25°C [dmm]	100	10-20	5-6
Penetration 40°C [dmm]			15-28
Ring & Ball [°C]	42	60-78	84-88
Viscosities			
150°C	0,1		1,5
160°C			0,9
170°C			0,4

Table 1 - C-Fix properties compared with penetration grade bitumen [1].

Mixing temperature of C-Fix binder is 200°C.

The company and brand name C-Fix is derived from Carbon Fixation. C-Fix has been developed in response to the Kyoto Protocol. In a way, C-Fix serves "as  $CO_2$ -sink" ( $CO_2$ -fixation). Due to the fixation of carbon from crude oil in sustainable non-fuel applications, C-Fix contributes to the prevention of  $CO_2$ -emission.  $CO_2$  is one of the so-called greenhouse gases leading to global warming.



Figure 3 - Production of C-Fix [1]

#### 2.1.2 - Use of C-Fix as binder for paving mixtures.

C-Fix has been used in small scale, especially on overloaded road segments. Due to its high strength and rutting resistance, it has been mostly used in road constructions to prevent permanent deformation, wear, and chemical attack. Few examples are:

- exit lane of Rotterdam harbour;
- bus stop in Amersfoort (overloaded due to breaking);
- indoor and outdoor storage area of Heineken brewery (punctual high static loads);
- access to HEMA distribution centre in Utrecht (heavy traffic of freight trucks slow down in front of the gate);

Asphalt mixtures with C-Fix are very stiff at service temperature and only small deformation occurs when the pavement is subjected to a load with long loading time. This is the case in storage container areas where the pavement is subjected both to loads of long duration and to scrapping of containers. Comparison can be made in the pictures of Figure 4.



Figure 4 – Deformation due to punctual load in polymer modified bitumen (left) and C-Fix [1].

#### 2.2 - Bitumen

# 2.2.1 –Constitution and rheology of bitumen and related materials

Rheology is the time/temperature dependent response of materials which are subjected to an applied force. The rheological characteristics of a bitumen at a particular temperature are determined by both the constitution (chemical composition) and the structure (physical

arrangement) of the molecules in the material [2]. Changes in the constitution and/or the structure will change the rheological characteristics of a bitumen.

Basically bitumen is composed by 80% by weight of carbon; around 10% hydrogen; up to 6% sulphur; small amounts of oxygen and nitrogen; and trace amounts of metal such iron, nickel and vanadium [2].

Generally speaking, bitumen can be separated into two broad different chemical groups called asphaltenes and malthenes. The maltenes can be further separated into saturates, aromatics and resins.

Asphaltenes are n-heptane insoluble black or brown amorphous solids containing besides carbon and hydrogen some nitrogen, sulphur and oxygen. The asphaltene content influences the rheological properties of a bitumen. The higher the asphaltene content the harder the bitumen and the higher the viscosity. This will lead to a lower penetration and higher softening point. Asphaltenes constitute 5 to 25% of the bitumen.

Resins are soluble in n-heptane and like asphaltenes they are largely composed of hydrogen and carbon and contain small amount of oxygen, sulphates and nitrogen. They have a dark brown colour and its proportion to asphaltenes governs, to a degree, the solution (SOL) or the gelatinous (GEL) type of the bitumen.

Aromatics represent the major proportion of the dispersion medium for the asphaltenes. They constitute 40 to 65% of the total bitumen and are dark brown viscous liquids.

Saturates are non-polar viscous oils which are straw or white colour. Their components include both waxy and non-waxy saturates. This fraction forms 5 to 20% of the bitumen.

#### 2.2.2 - Bitumen structure

Bitumen is traditionally regarded as a colloidal system consisting of high molecular weight asphaltene micelles dispersed or dissolved in a lower molecular weight oily medium, the maltenes [3]. According to the proportions of the asphaltenes and maltenes (resins, aromatics, saturates) the bitumen structure can vary. It can be defined as a SOL-type if the presence of sufficient quantities of resins and aromatics of adequate solvating power keeps the asphaltenes separated. The resulting micelles have then a good mobility within the bitumen. A GEL-type bitumen occurs exactly when no sufficient quantities of resins/aromatics are present or if they don't have sufficient solvent power. In this way, the asphaltenes can associate together, leading to an irregular open packed net structure. Figure 5 shows the difference between GEL and SOL-type bitumen.



Figure 5 - SOL-type bitumen (left) and GEL-type bitumen [2]

Depending on the proportions of asphaltenes and maltenes, the rheology of the binder in question can change. For instance when keeping the asphaltenes content constant it has been demonstrated that:

- there is marginal reduction in shear susceptibility if the aromatics content is increased keeping constant the ratio saturates/resins;
- a softer bitumen is produced if the ratio resins/aromatics is kept constant and the amount of saturates increases;
- an increasing resins content hardens the bitumen, reduces the penetration index and shear susceptibility but increases the viscosity [2].

Also the change in the asphaltene content changes the properties of the bitumen. The greater its content, the higher the viscosity at a constant temperature. The shape and the aggregation of the asphaltenes also have an influence on the viscosity of the bitumen. C-Fix, which has a high asphaltene content, behaves similar to a GEL-type bitumen [3].

A bitumen with a penetration grade of 100 has around 6% of asphaltenes, while C-Fix has from 30 up to 35% [3]. C-Fix having a gel-type structure and a high asphaltene content is related to a so called hard bitumen.

### 2.3 - Polymer

#### 2.3.1 – Polymer definition and properties

#### 2.3.1.1 – Brief history and origin

Polymers have existed in natural form since life began such as proteins and polysaccharides playing an important role in plant and animal life. From the earliest times, man has exploited naturally-occurring polymers as source for clothing, decoration, shelter, tools, weapons, writing materials, etc. The origin of today's polymer industry is commonly accepted as being in the nineteenth century when important discoveries were made concerning the modification of some natural polymers [4].

In the first half of nineteenth century, Thomas Hancock discovered that when masticated (subjected repeatedly to high shear forces), natural rubbers becomes more fluid making it easier to blend with additives and to mould. A few years later, Charles Goodyear found that the elastic properties of natural rubbers could be improved eliminating its tackiness by heating with sulphur. At the beginning of the nineteenth century's second half Nelson Goodyear (Charles's brother) patented the vulcanization of natural rubber with large amounts of sulphur to produce a hard material more commonly known as hard rubber, ebonite or vulcanite.

At the same period cellulose nitrate was found to be a hard elastic material which was soluble and could be moulded into different shapes by application of heat and pressure. After several improvements, until the end of the nineteenth century these and other polymers were semisynthetics as they were produced from natural polymers. Bakelite phenol-formaldehyde resins, created by Leo Baekeland, are the first fully-synthetic polymers to be commercialized in 1910. Methyl rubber is the first synthetic rubber to be produced from 2,3-dimethylbutadiene during World War I [4].

#### 2.3.1.2 – Definition of polymer

Polymer is a substance composed of molecules which have long sequences of one or more species of atoms or groups of atoms linked to each other by primary, usually covalent, bonds [4]. Macromolecules are formed by linking together in a chain monomer molecules through chemical reactions, known as polymerization. A monomer molecule is a small molecule that may become chemically bonded to other monomers to form a polymer. The long chain nature formed in polymerization gives the polymers their characteristic properties.

Polymers may have linear and non-linear skeletal structures. The linear polymers are formed by joining chemically end-on-end the basic building blocks (i.e. the monomer) and may be represented by a chain with two ends. These linear systems are thermoplastic, or in other words, when heated they melt and when cooled they solidify and harden. Over 80% of plastics are thermoplastic in nature such as polyethylene (PE), polypropylene (PP), polyvinyl chloride (PVC) and polystyrene (PS). They may be homopolymer when the long chains are made of the same monomers. If two or three types of monomers are used, they are called copolymers such as styrene-butadiene rubber copolymer [5].

The non-linear or network polymers are based on a different type of monomer which can be chemically joined up not only at each end but elsewhere too. These monomers can form covalently bonded three-dimensional networks. If the node points or links are close together, a stiff material is formed, otherwise wide separated the network may be flexible [5].



Figure 6 - Basic linear chain polymer [5].



Figure 7 - Network polymer - left: short chain; right: long chain [5].

### 2.3.2 – Use of polymer to modify bitumen

One of the prime roles of adding polymer to a bitumen is to increase the pavement's resistance to permanent deformation at high temperatures, without changing its properties at other temperatures. This is achieved either by stiffening the bitumen so that the total viscoelastic response of the asphalt is reduced, or by increasing the elastic component of the bitumen via reducing the viscous component. Both ways result in a reduction of permanent deformation [2].

There are many types of polymer being used in bitumen lately aiming the goals mentioned before. Some of their requirements to be effective economically and practically are:

- be readily available;
- resist degradation at asphalt mixing temperatures;
- blend with bitumen;
- be cost effective;
- be chemically and physically stable during storage, application and in service;
- be able to be processed by conventional equipment [2].

Some types of polymers are given in the next Table 2, with the properties they are expected to improve in the bitumen.

Modifier	Permanent deformation	Thermal cracking	Fatigue cracking	Moisture damage	Ageing
Elastomers <sup>[80-82]</sup>	1	1	1		1
Plastomers <sup>[83,84]</sup>	1				
Tyre rubber <sup>[85]</sup>		1	1		
Carbon black <sup>[86,87]</sup>	1				1
Lime <sup>[88,89]</sup>				1	1
Sulphur <sup>[90,91]</sup>	1				
Chemical modifiers	1				
Antioxidants					1
Adhesion improvers				1	~
Hydrated lime <sup>[92]</sup>				1	1

Table 2 – Expected improvement of binder properties for different polymer types addition [2].

C-Fix has a high resistance to permanent deformation due to its higher Young's Modulus when compared with normal bitumen. Its higher penetration index (PI) also indicates a high resistance to permanent deformation. The drawback of C-Fix is its crack sensitivity and brittle behaviour at low temperatures as stated by Khedoe [3]. These issues may bring some difficulties in applying C-Fix binder in areas where winter might be mild to severe. The brittle behaviour of this binder could make the lifespan of the pavement much lower due to crack formation and ravelling.

The polymer modification proposed in this research aims to improve its brittle behaviour at low temperatures and still keep the good resistance to permanent deformation of the C-Fix binder. The polymer in question is a SBS (Styrene-Butadiene-Styrene) thermoplastic elastomer. For this reason, further explanation on the properties of a elastomer shall be presented.

### 2.3.3 – Elastomer polymers

Thermoplastic elastomers derive their strength and elasticity from physical cross-linking of the molecules into a three-dimensional network. This is achieved by the agglomeration of the polystyrene end-blocks into separate domains, providing the physical cross-links (mid-blocks) for a three-dimensional polybutadiene rubbery matrix. The end-blocks stiffen the polymer and the mid-blocks give the material its exceptional elasticity. Figure 8 shows this distribution. At high temperatures, the polystyrene softens and will even dissociate under stress, allowing easy processing. Later cooling will re-associate them and elasticity is restored [2].



Figure 8 - Distribution of polystyrene end-blocks and the cross-links polybutadiene mid-blocks in a SBS polymer [2].

When the polymer is added to the hot binder, the binder immediately starts to penetrate the polymer particles causing the styrene domains of the polymer to become solvated and swollen. Medium to high shear mixers are required to adequately disperse thermoplastic elastomers into the bitumen.

As discussed earlier, bitumen can be subdivided into groups of molecules that can have common structures which are: maltenes (saturates, aromatics and resins) and asphaltenes. Saturates and aromatics can be viewed as carriers for the polar aromatics (resins and asphaltenes). The polar aromatics are responsible for the viscoelastic properties of the bitumen at ambient temperature. The addition of thermoplastic elastomers with a molecular weight similar to or higher than that of the asphaltenes disturbs the phase equilibrium. The polymer and the asphaltene 'compete' for the solvency power of the maltenes phase and if insufficient maltenes are available, phase separation may occur [2].

Phase separation, or storage instability will not be further investigated as the polymer used in this research is proposed to be mixed with the binder in a new manner. The polymer is produced as dry granulates which can be added directly in the pug-mill. This will bring great flexibility in the use of polymer modified binders. The storage stability is no longer a problem when polymer modified binders are produced according to the correct amount needed.

## 2.3.4 - Altraflex-2006

Altraflex-2006 is an oily solution containing basically 40% of high molecular weight SBS polymer. It includes in its formula some active filler calcium hydroxide which can help in absorbing free radicals, so that the oxidation process is slowed down, improving in this way ageing resistance. It is expected to improve the permanent deformation as well as fatigue cracking due to its thermoplastic elastomer characteristics. The adhesion is improved which at the same time improves water resistance.

So Altraflex-2006 is a product that consists of more than one component, which are:

- for the polymer modification: a SBS elastomer;

- for ageing-durability: a material that can absorb free radicals so that the oxidation process is slowed down;

- an adhesion improving component on stone, that ensures at the same time better water resistance.

The philosophy behind these additives is that it is ensured that oxygen and UV have as little as possible influence on the interaction between the mortar and the stone area. As a thermoplastic elastomer, Altraflex-2006 is expected to reduce the brittle behaviour of C-Fix under low temperatures. Finally Altraflex-2006 is easy to use since it can be added directly in the pug-mill.

Altraflex-2006 in this thesis report may only be referred to as "polymer" as well.

### 2.4 – Filler

The use of filler in the asphalt mixture has shown to improve its properties concerning drainage of binder and improved adhesion of the mastic to the aggregate preventing stripping. It is also expected to increase mastic's viscosity improving permanent deformation resistance [2].

Therefore also the influence of the filler is investigated. According to the Dutch RAW standard [6] a so called "middle type" filler should be used for PAC, with a certain amount of calcium hydroxide. This component is expected to act mainly as an anti-stripping agent, improving the bond between the aggregate and binder.

Due to the already higher viscosity and higher strength of the C-Fix binder when compared with bitumen, use of the "middle type" filler could be questioned. In this sense, the calcium hydroxide content used in the mixture could be decreased by the use of a less active filler (or weak filler) composed only of limestone for instance. The calcium hydroxide could increase the already high strength of the C-Fix binder, worsening the brittle behaviour of the mastic.

The filler types used in this research are both produced by Ankerpoort B.V. Their properties are described in Table 3.

	Wigro 60K	Duras Filler 15	
Туре	Medium type filler with hydroxide	Very weak	
Producer	Ankerpoort Winterswijk	Ankerpoort Maastricht	
Components	Limestone 65 – 75%	Limestone 100%	
	Calcium hydroxide 25 – 35%		
Gradation			
• 2 mm	100 % (m/m)	100 % (m/m)	
<ul> <li>0,125 mm</li> </ul>	87 - 97 % (m/m)	90 - 100 % (m/m)	
<ul> <li>0,063 mm</li> </ul>	80 - 90 % (m/m)	76 - 86 % (m/m)	
Density	2,48 – 2,68 [Ton/m <sup>3</sup> ]	2,60 – 2,80 [Ton/m <sup>3</sup> ]	
Water sensitivity	≤ 10% (m/m)	≤ 10% (m/m)	
Specific surface	4.000 – 6000 [cm <sup>2</sup> /gr]	5.330 [cm <sup>2</sup> /gr]	
Bitumen number	56 - 62	28 – 34	

Table 3 - Filler properties.

The granular shape and gradation of the filler exerts considerable influence on the final density of the stone/sand mixtures in the asphalt. The finer the filler, the more it absorbs bitumen in the mixture, influencing the final coating of aggregates. The gradation curves of the two fillers are presented in Figure 9.

**GRADATION OF FILLERS** 



Figure 9 - Wigro 60K and Duras 15 gradation curves.

## 3. Test background

In this chapter, the tests that were performed in this research are presented. For each of the tests, the theory behind the test shall be given, together with some extra information, when necessary. First, information on the viscoelastic models are presented, which are extremely useful on the tests results analysis and interpretation.

#### 3.2 – Viscoelastic model

It is a well known fact that bituminous mixes show a viscoelastic and viscoplastic behaviour [7]. To understand how the material responds to an applied load, sets of mechanical elements such as springs and dashpots are used to form models. Such models include the Maxwell model, Kelvin (Voigt) model and Burger's model. Figure 10 shows the response on the mechanical elements to an applied load.



Figure 10 - Response of the mechanical elements for an applied load [24].

#### 3.2.1 – The Maxwell model

This model is a combination of a spring and dashpot in series. The Maxwell model is most suitable for cases in which a constant strain is applied and the stress in monitored (stress relaxation). It is capable of capturing permanent deformation due to the presence of a dashpot connected to a spring in series. Any strain accumulated within the viscous component of that unit is entirely non-recoverable. The Maxwell model is commonly described using its relaxation time [24]. See Figure 11 for details.

$$\frac{d\sigma}{dt} = E \cdot \frac{d\varepsilon_1}{dt}$$

$$\sigma = \eta \cdot \frac{d\varepsilon_2}{dt}$$

$$\varepsilon = \varepsilon_1 + \varepsilon_2$$

$$\sigma = \sigma_1 = \sigma_2$$

$$\frac{d\varepsilon}{dt} = \frac{1}{E} \cdot \frac{d\sigma}{dt} + \frac{\sigma}{\eta}, \text{ where } \eta = \tau \cdot E$$

#### Figure 11 - Maxwell model [7].

Where:

- E = Elastic modulus [Pa];
- $\sigma = \text{stress [Pa]};$
- $\varepsilon = \text{strain} [\%];$
- $\eta$  = viscosity [Pa.s];
- $\tau$  = the relaxation time for a Maxwell model [s];

 $\frac{d\varepsilon}{dt} = \text{strain rate.}$ 

I

#### 3.2.2 – The Kelvin model

The Kelvin (Voigt) model is the simplest representation of the behaviour of viscoelastic materials. It is composed of a linear spring and a dashpot in parallel and is most suitable for cases in which a creep load is applied and the strain is monitored (strain retardation). The Kelvin model is commonly described using its retardation time [24]. Figure 12 presents the details.

E  

$$\sigma_{1} = E.\varepsilon$$

$$\sigma_{2} = \eta \cdot \frac{d\varepsilon}{dt}$$

$$\varepsilon = \varepsilon_{1} = \varepsilon_{2}$$

$$\sigma = \sigma_{1} + \sigma_{2}$$

$$\sigma_{1} = E.\varepsilon$$

$$\sigma_{1} = E.\varepsilon$$

$$\sigma_{1} = E.\varepsilon$$

$$\sigma_{2} = \eta \cdot \frac{d\varepsilon}{dt}$$

$$\sigma_{1} = \frac{\sigma}{\eta} + \frac{E.\varepsilon}{\eta}$$
, where  $\eta = \tau \cdot E$ 



Where: E = Elastic modulus [Pa];  $\sigma$  = stress [Pa];  $\varepsilon$  = strain [%];  $\eta$  = viscosity [Pa.s];  $\tau$  = the relaxation time for a Kelvin model [s];  $\frac{d\varepsilon}{dt}$  = strain rate.

#### 3.2.3 – The Burgers model

The Burgers' model is composed of the two models previously explained connect in series, namely the Maxwell and the Kelvin models. It is widely used to model complex viscoelastic behaviour such as that one of bitumen or C-Fix binder and mastics as they exhibit signs of both stress relaxation and strain retardation [24]. See Figure 13.



#### Figure 13 - The Burger model [7].

The principles of the Burgers' model and the viscoelastic response are used in the interpretation of the different tests, such as tests done with the dynamic mechanical analyzer, dynamic shear rheometer and relaxation tests.

#### 3.3 - Microscopy investigation

Multiphase systems like binder polymer blends have some requirements on the discontinuous phase to maintain their structure over a long life time. To assess the homogeneity of a blend, samples should be checked using the fluorescence microscopy under UV light [8]. As most of the polymers produce a yellow fluorescence and the binder remains dark, a clear contrast is expected to be seen between the two phases. Particle size, shape and distribution can be assessed.

In order to have the samples well visualized, special preparations have to be done before bringing them under the microscope. They have to be prepared, and cooled down to room temperature. After that, they have to be placed in the deep freezer for at least 3 hours at -20°C. The frozen samples have to be broken in small flat pieces in order to get good focus in the microscope. The samples can also be cut with a sharp tool, but this procedure can deform the polymer distribution in the inspected surface possibly resulting in false distribution patterns. To avoid this, the samples are better broken at low temperature when still very brittle.

The inspection can be done through a protection glass or directly on the surface. The magnifications suitable are of 25 to 500 for binder inspections. At least three independently prepared surfaces have to be inspected for each polymer content, according to NEN-EN 13632 [8].

The distribution of the polymer phase or binder can be assessed by measuring the particle size distribution or by comparison with standard pictures provided in the NEN-EN 13632 [8]. Compatibility, stability and compliance can all be assessed by fluorescence microscopy. It has to be mentioned that this does not give the final quality assessment or indication of the binder performance. It provides important information for understanding the system and parameters which might have an influence on the final quality.

According to the NEN-13632, the microscope used should have some features in order to make this fluorescence investigation:

- Light source:	High pressure Xenon lamp, 75W;
- Excitation filter:	BP 420/490 (transmit wavelengths from 420nm-490nm);
- Beam splitter filter:	RKP 510 (reflects wavelengths shorter than 510nm and transmits light of longer wavelengths):
- Barrier filter:	LP 515 (transmits wavelengths longer than 515nm) [8].

### 3.4 – Rheology

Rheological properties of the binder are investigated through a number of conventional binder tests. Rheology is the study of the deformation and flow of a material when subjected to an applied stress. Conventional binder tests such penetration and softening point plus Dynamic Shear Rheometer (DSR) test and Dynamic Mechanical Analysis (DMA) test will be commented.

## 3.4.1 – Penetration

The consistency of a binder is measured by the penetration test NEN-EN 1426 [9]. This consistency is expressed in tenths of a millimetre that a standard needle will penetrate vertically into a sample of the material.

The principle of this test is to measure the penetration of a standard needle into a conditioned test sample. For penetrations up to  $500 \times 0.1$  mm the test conditions are: temperature of  $25^{\circ}$ C, an applied load of 100 g and a duration of loading of 5 s. For penetrations above  $500 \times 0.1$  mm the temperature can be reduced to  $15^{\circ}$ C. It is not mentioned in the standard the temperature for very low penetrations measured at  $25^{\circ}$ C. But it is already common practice to perform those tests at 40°C and 60°C for very hard binder such as C-Fix. Figure 14 shows a representation of the penetration test.



Figure 14 - Penetration scheme [10].

#### 3.4.2 – Softening point

Softening point or Ring & Ball (R&B) temperature is the temperature at which the material under standardized test conditions reaches a specific consistency [11].



Figure 15 - Softening point (Ring & Ball) scheme [10].

Two horizontal discs are filled with binder, heated at a controlled temperature rate in a liquid bath while each supports a steel ball. See Figure 15 and Figure 16. The softening point is reported as the mean of the temperatures at which the two discs soften enough to allow each ball, enveloped in binder, to fall a distance of 25,00mm  $\pm$  0,4mm as long as the difference between both temperatures does not exceed 2 seconds (for modified bitumen and softening points above 80°C).



Figure 16 - Ring & Ball test photo series.

The softening point and penetration test are very useful for bitumen classification. They are simple and fast tests which can characterise well different grades of bitumens, establishing uniformity on delivery or sources of supply.

#### 3.4.3 - Dynamic shear rheometer (DSR)

The Dynamic Shear Rheometer is used to characterize the viscous and elastic behaviour of asphalt binders [12]. The resistance to shear is expressed in terms of the shear modulus,  $G^*$ , which is defined as:

$$G^* = \frac{\tau_{\max}}{\gamma_{\max}}$$

Where:  $G^*$  = Complex Shear Modulus [Pa]  $\tau_{max}$  = Maximum shear stress [Pa]  $\gamma_{max}$  = Maximum shear strain [-]

The elastic modulus and shear modulus are related by the equation:  $E = 2.(1 + \mu).G^*$ ,  $\mu$  is Poisson's ratio, and depends on the compressibility of the material. It may be assumed to be 0,5 for almost incompressible pure bitumens, having the above equation simplified as  $E \approx 3.G^*$ .

Used to define rheological properties of binders, the DSR procedure involves determining the Complex Shear Modulus (G\*) and the Phase Angle ( $\delta$ ) over a range of test frequencies and test temperatures when tested in oscillatory shear [12]. A known oscillatory shear stress/strain is applied to the temperature controlled test geometry, in which the binder test specimen is held. The Complex Shear Modulus is measured, which is the total response of the binder under the applied stress.

The Complex Shear Modulus can be decomposed in the in-phase and out-of-phase components which are storage modulus (G') and loss modulus (G''). The storage modulus gives information on the elastic responses of the material while the loss modulus is associated with the viscous responses.

The Complex Shear Modulus can be calculated

$$|G^*| = \sqrt{(G')^2 + (G'')^2}$$
$$\delta = \frac{G''}{G'}$$

Where:  $G^*$  = Complex Shear Modulus [Pa]

G' = Storage modulus G'' = Loss modulus  $\delta$  = Phase angle

The complex shear modulus and the phase angle define the material resistance to shear deformation in the linear viscoelastic range and are related to rutting and fatigue resistance of the asphalt mixture. Long duration or static loads or very low frequency loading will show the viscous characteristics of the binder. At short loading times or high frequencies the elastic properties will dominate. The influence of temperature is similar: higher temperatures will reveal viscous behaviour and elastic characteristics dominate at lower temperatures.

The test set-up consists of two parallel metal plates, acclimatised chamber, loading device and control/data acquisition system. Two plate sizes are normally used namely 8 and 25 mm. The 8mm plates are suitable for lower temperature ranges and materials with higher complex modulus (G\* between 100 kPa to 10.000 kPa). The 25 mm plates are suitable for tests at higher temperatures and materials with lower complex modulus (G\* between 1 kPa and 100 kPa). One of the plates has oscillating movements with respect to the other plate at pre-defined frequencies and rotational displacement amplitudes. Purely elastic materials have 0° as phase angle. In the contrary, purely viscous materials have 90°. Details of the oscillating plates and the stress-strain DSR test output are shown in the Figure 17.



Figure 17 - DSR plates set-up [3] and output measurements [2].

The (loading) time/frequency and temperature are interrelated by the so called time-temperature superposition principle which states that the temperature will not change their frequency or time dependent behaviour, but will merely shift the frequency/time scale. This principle is used to build the master curves of complex shear modulus as well as the phase angle and is further detailed in the next paragraph.

#### 3.4.3.1 – Time-temperature superposition principle.

As said before, the master curve of materials such as bituminous binders or polymers can be built based on the time-temperature superposition. This is mainly done because it is not practical to perform test over the entire temperature and loading time domain in order to determine the binder's dependency on these factors [7].

It is thought that there is a general equivalence between time and temperature. For instance, a polymer or binder which has rubbery characteristics under a given set of testing conditions can be induced to show glassy behaviour by either reducing the temperature or increasing the testing rate of frequency. So, it was found empirically that curves from DSR tests performed at different temperatures could be superposed by keeping one on them fixed and shifting all the others by different amounts horizontally, parallel to the logarithmic frequency axis. If an arbitrary reference temperature  $T_s$  is taken to fix one curve and if  $\omega_s$  is the frequency of a point on the curve at  $T_s$  with particular shear modulus and  $\omega$  is the frequency of a point with the same shear modulus on

a curve for a different temperature, then the amount of shift required to superpose the two curves is a displacement of (  $\log \omega_s - \log \omega$ ) along the log frequency axis, displacement which is called the shift factor  $a_{\tau}$  [4].

The Williams, Landel and Ferry equation, or shortly called WLF equation which is used to build the master curve was originally developed empirically and holds extremely well for a wide range of polymers and binders:

$$a_{T} = \frac{-C_{1} \cdot (T - T_{s})}{C_{2} + (T - T_{s})},$$

Where:

 $a_T$  = shift factor;

 $C_1$ ,  $C_2$  = material dependant constants;

 $T_{s}$  = reference temperature [°C];

T = test temperature [°C].

#### 3.4.4 - Dynamic mechanical analysis test (DMA)

Dynamic Mechanical Analysis can be used to locate the glass transition region and to determine the glass transition temperature of amorphous and semi-crystalline materials [13]. The analyzer monitors changes in the viscoelastic properties of a material as a function of temperature and frequency. In ideal cases, the temperature at which a big drop in the storage modulus occurs marks the glass transition temperature ( $T_g$ ). Different phases on an idealized DMA scan are displayed in the Figure 18.



Figure 18 - Idealized DMA scan [14].

To understand the principle of this test, knowledge on the molecules disposition and move-ability are important. Polymers and asphalt binders have roughly the same behaviour when subjected to a temperature change. They exhibit glass properties at low temperatures, but more rubbery at higher temperatures. As mentioned before, polymers are formed of linear or network molecular chains showing no structural order, or in other words, they are amorphous systems.

In this discussion, the temperature rise is assumed to be slow and uniform in the polymer specimen. If the sample is really cold, no chains or parts of the molecule can move, so the material is frozen into a rigid glassy state. On slow heating, side groups have sufficient thermal energy to rotate, with a slight drop in the modulus (so called  $\beta$  transition temperature or  $T_{\beta}$ ). On further heating, complete blocks or segments of the material chain become free to move as local entities, perhaps in a crankshaft motion. This is the glass-rubber transition temperature, or  $T_g$ . The modulus now drops by several orders of magnitude, as seen in Figure 19 [5].



Figure 19 - Molecule mobility [5].

A sharp transition temperature is not always present. As the temperature approaches  $T_g$ , some smaller segments become free to move and this causes a slight drift downwards in the modulus before the catastrophic drop of three orders of magnitude [5].

Specimens with normally rectangular shape are tested at a heating rate of  $1^{\circ}$ C/min and a sinusoidal strain with frequency of 1 Hz. The maximum amplitude should be within the linear viscoelastic range of the material. Strains of less than 1% are recommended and should not exceed 5%. The set up used in the DMA was the dual cantilever clamps system. A rectangular specimen with dimensions of around 60mm x 13mm x 3.5 mm (LxWxT) is clamped in the dual cantilever as displayed in Figure 20. The sinusoidal deformation is induced in the middle of the rectangular specimen.



Figure 20 - Dual cantilever set up in the DMA test [18].

A tangent to the storage modulus curve below the transition temperature and at the inflection point is approximately constructed to aid the glass transition temperature determination. The temperature at which these tangent lines intersect is reported as the glass transition temperature,  $T_g$  as illustrated in Figure 21.



Figure 21 - Glass transition temperature according to the storage modulus curve [13].

#### 3.5 – Direct tensile test (DTT)

This test is used to determine the stress at failure and the strain at failure of asphalt binders by applying direct tension with a constant rate of elongation [15]. This test method is suitable for brittle and brittle-ductile failure resulting in a fracture of the test specimen. Ductile failure where the specimen only stretches is not applicable. The temperature ranges used in this test should be also in accordance with the brittle or brittle-ductile failure. This test is designed to measure the strength of the asphalt binder at critical cracking temperatures.

Test specimens are prepared by pouring hot asphalt binder into a suitable mould. Two aluminium or plastic end tabs are used to bond the asphalt binder during the test and transfer the tensile load from the test machine to the asphalt binder.

A displacement transducer is used to measure the elongation of the test specimen as it is pulled in tension at a constant rate of 1 mm/min. The load developed during the test is monitored and the tensile strain and stress in the test specimen when the load reaches a maximum are reported as the failure strain and failure stress, respectively. Specimen and data output details are shown in the Figure 22.



Figure 22 - Direct tensile test principle [19].

Failure stress is the point at which the tensile load reaches a maximum value as the test specimen is pulled at a constant rate of elongation. Failure strain is the tensile strain corresponding to the failure stress:

$$\sigma_f = \frac{P_f}{A}$$
$$\varepsilon_f = 100.\frac{\delta_f}{L_e}$$

Where:

 $\sigma_{f}$  = failure stress [MPa];

- $P_f$  = failure load [N];
- A = initial cross section [m<sup>2</sup>];
- $\varepsilon_{f}$  = failure strain [%];
- $\delta_{f}$  = elongation at failure [mm];
- $L_e$  = effective gage length [mm].

#### 3.6 - Relaxation test

The relaxation test is performed using the same test set-up as for the Direct Tensile Test. The specimens are prepared in the same way. A small displacement is imposed on the specimen in the longitudinal direction and held at the specific strain level, resulting in a tensile stress. The tensile stress dissipation is measured, in this so-called displacement controlled test (see Figure 23). Different temperature may be used for the test, depending on the properties being investigated.

To be able to compare the relaxation of different binders and mastics at different temperatures, the relaxation is displayed as a percentage of the maximum stress measured during the test.

$$R = 100. \frac{\sigma_{res}}{\sigma_{max}}$$

Where:

 $\begin{aligned} R &= \text{relaxation [\%];} \\ \sigma_{res} &= \text{residual stress [MPa];} \\ \sigma_{\text{max}} &= \text{maximum stress [MPa].} \end{aligned}$ 



Figure 23 - Illustration of the relaxation test in displacement controlled test [3].

#### 3.7 – Adhesion zone fatigue test (Stone columns test)

The primary function of a binder is to act as an adhesive. The need to ensure adhesion between the aggregate and the binder is very important. The adhesion of binders to most types of dry and clean aggregate is fairly good to excellent. However, aggregate is easily wetted by water, the presence of which can result in unexpected problems. Also the aggregate mineralogical composition, chemical composition, shape, structure, residual valency and texture of the surface are of importance. In the mixture itself, the void content, permeability, binder content, bitumen film thickness and filler type play an important role on adhesion [2].

The properties of the binder are also important in the acquisition and subsequent retention of the binder/aggregate bond. Properties such as bitumen viscosity during coating and in service, polarity and constitution influence adhesion characteristics. The aggregate nature is indeed by far
the most important factor in binder/aggregate adhesion [2]. Despite this fact, some binder properties play an important role also, which claims the necessity of assessing the binder adhesion characteristics.

The aim of the stone columns fatigue test is to assess one of the causes, among many, of adhesion failure between the mastic and aggregates in the asphalt mixtures. To fully understand this failure possibility, the failure mechanisms should be known.

There are basically two failure mechanisms between binder and aggregate. It is said to be adhesion failure when water can get between the binder film and the aggregates, breaking the adhesive bonds between the aggregate and binder. Stripping of the binder from the aggregate may occur possibly due to higher affinity of aggregates for water than for binder. The failure then occurs due to a lack of adhesion between binder and aggregate [20].

In contrary, when water affects the binder itself by chemically and/or physically interacting with it, reduction in cohesion within the binder may result. This is said to be cohesion failure, which can lead to a severe reduction in integrity and strength of the mixture. Cohesion failure may be divided in two types of mechanisms. The first type is associated with a softening of the binder in the presence of water, causing failure within the binder film. The other type is associated with the softening of the bitumen, which weakens the bond between the binder and the aggregate, causing separation of the film from the aggregate surface. In this case, cohesion failure may be due to first cohesion loss followed by adhesion loss [20].

Whether a cohesive or adhesive failure occurs depends on the nature of the binder and the relative thickness of the binder around the coarse and fine aggregates. If the binder has low resistance to fatigue, a rupture may occur exactly between the mastic (binder, sand and filler) and coarser aggregates. There is a thickness of the binder coating the aggregate where no filler/sand in present, which is supposed to be the link between the mastic and the aggregate. There would be no reason to use an extremely high strength binder such as C-Fix, if there would not be guaranty that this same binder would have a good adhesion property to link the mastic to the coarse aggregates.

In the stone columns test, the thickness where no filler/sand is available is assumed to be 15  $\mu$ m. Based on this assumption, specimens Ø 3,9 mm drilled out of stones in the shape of "columns" are glued together via a 15  $\mu$ m binder thickness (see Figure 24).



Figure 24 - Stone columns specimen [21]

Care should be taken when preparing those specimens. The cores should be drilled from stone slices with the most parallel surfaces possible. As the roughness was also a concern, the stone slices were sand blasted first before drilling the specimens. The cores were then boiled in demineralised water to have the grease and dust removed.

For the assembly of these specimens, it is important that the two stone columns are precisely centred and the two contact surfaces exactly parallel. Use of a rheometer with specially designed clamps is a solution, as it can position the drive shaft within an accuracy of 1  $\mu$ m. See Figure 25 for details.



Figure 25 – Stone column specimen preparation [21].

After assembling the specimens, they can be kept in the refrigerator until the moment of the test. To determine the fatigue resistance of the adhesion layer, a oscillating torque is applied to the stone-binder sample, inducing a sinusoidal angular rotation of the upper stone and shear strain in the binder interface.

Due to the thin layer of viscoelastic material the specimen becomes very stiff. For correct mounting, the two clamps of the measurement system must be exactly. This would avoid stresses originated due to imperfections in the alignment in the binder interface layer as the clamps are tightened. For this reason, after the sample is mounted into the upper (free rotating) clamp, the lower part of the sample is glued to the fixed base of the rheometer using a quick drying strong adhesive (HBG X60) - see Figure 26.



Figure 26 – Stone binder sample mounted in a DSR test set-up [21].

Tests showed that this adhesive is much stronger than the interface layer. Both auto gap temperature compensation and (zero) normal force control are essential DSR equipment features in order to avoid unwanted loading of the thin interface layer.

Short-term and long-term fatigue test ranges have to be determined. For this reason, different torque ranges are applied in the mounted specimen. Depending on the kind of binder being tested, a different range in frequency is also suitable. The 15  $\mu$ m binder thickness should give an insight on the fatigue properties in the adhesion zone existent between the mortar and the aggregate. The results can be presented in number of cycles related to torque level prior to failure. Time versus torque level or frequency used in the test can be another option.

### 3.8 – Ageing procedure adopted

There have been a number of attempts to simulate the long-term ageing of binder in situ, which seems to be extremely difficult because of the number of variables that affect binder ageing such as void content, mixture type, aggregate type, etc. Specially when dealing with porous asphalt where the void content is extremely elevated. The US Superpave specification uses the rolling thin-film oven test (RTFOT) to simulate initial ageing followed by ageing over 20 hours at elevated temperature (90, 100 or 110°) and a pressure 2070 kPa in a pressure ageing vessel.

Since the method previously described is still being validated, other methods are currently being studied and checked for possible validation. "The effect of ageing on binder properties of porous asphalt concrete" is a PhD study currently being carried by E.T.Hagos [27]. In his research, a protocol has been developed to simulate the binder ageing in the Porous Asphalt Concrete (PAC).

In this PhD research, a laboratory accelerated weathering instrument is used for ageing the binder in PAC slabs. This weathering method implies the use of an instrument that is capable to reproduce the average weather conditions, regarding sunlight, humidity, temperature, seasons, etc. Explanations on these variables will be presented in order to clarify the ageing concept.

### 3.8.1 – Weather factors

There are four main factors on weathering, which are: solar radiation (light energy), oxygen, temperature and moisture. For each of these factors, not only the amount is important, but also for instance types of solar radiation, different phases of moisture or temperature cycling have a significant effect on materials being aged. These factors, together with secondary effects such as pollutants, biological phenomena, and acid rain act to cause "weathering" [22].

Radiant energy that comes from the sun is made up of photons that travel through space as waves. Their energy is proportional to their frequency, velocity of light and wavelength. The solar radiation that reaches the earth's surface consists of wavelengths between 295 and 3.000 nm. The three main wavelength ranges are presented in the Table 4.

Range name	Wavelength	% of total solar
	range	
Ultraviolet (UV)	295 – 400 nm	6,8
Visible (VIS)	400 – 800 nm	55,4
Infrared (IR)	800 – 2450 nm	37,8

Table 4 - Solar sunlight separation [22].

Irradiance is the radiant flux incident on a surface per unit area or the rate of incident flux per area, commonly expressed in  $W/m^2$ . Normally it should be mentioned the spectral range the irradiance measurements were taken, for instance 295-400 nm – total solar. Radiant exposure is the time integral of (spectral) irradiance, expressed in J/m<sup>2</sup>. It can also be understood as the cumulative sunlight exposure at any specific time. Spectral irradiance is the irradiance, but classified per wavelength, normally stated in  $W/m^2/nm$ . It can also be explained as the quality of the irradiance [22].

The temperature of materials exposed to solar has an influence on the effect of the radiation. Photochemical reactions are usually accelerated at elevated temperatures. Temperature determines also the rate of subsequent reaction steps, or secondary reactions. A general rule of thumb assumes that reaction rates double with each 10°C rise in material temperature [22].

A material exposed to sunlight has its temperature determined by different factors. The specimen surface temperature is a function of ambient temperature, specimen solar absorptivity, solar irradiance and surface conductance. Therefore, in the presence of sunlight, the surface temperature of an object is usually considerably higher than the temperature of the air.

Concerning the temperature settings, the higher the test temperature, the higher the ageing rate, which makes the test duration shorter. Care should be taken not to damage or burn the material tested. The surface temperature should be set accordingly to the air temperature, always higher, as explained in the previous paragraph.

Water is one of the substances in our environment that is everywhere, whether in the form of humidity, rain, dew, snow, or hail. There are two ways in which water can affect materials:

absorption and desorption [22]. Absorption leads to volume expansion, which results in stresses on the dry sub-surface layers. Desorption follows from a dry-out period, leading to volume contraction. As the hydrated inner layers resist this contraction, surface cracking occurs. Freezethaw cycle is another physical effect of water. As water expands when it freezes, it causes stresses and probably failure.

# 3.8.2 – Local climate

It is not practical to determine the weathering characteristics of materials in all the world's climate. The major marketing area of the material should be taken into consideration when selecting suitable climates and sites for weathering tests. In the Netherlands, Lochem has been chosen as a representative location. Hoek van Holland is also such a location.

From these two locations, data are available as showed in the Table 5.

Location	Latitude	Longitude	Elevation (m)	Average ambient temp (°C)	Average ambient RH (%)	Annual mean rainfall (mm)	Annual mean total radiant exposure (MJ/m <sup>2</sup> )
Lochem	52° 30′ N	6° 30′ E	35	9	83	715	3700
Hoek van Holland	51° 57′ N	4° 10′ E	6	10	87	800	3800

Table 5 - Climate data in the Netherlands [22].

## 3.8.3 – Laboratory weathering

There are two ways of ageing materials: natural, by exposing them to direct sunlight and other elements of weather; and in the laboratory. In the laboratory, there are several ways used to mimic the outside weather conditions.

Due to the necessity to speed up evaluations the ageing resistance of materials than it can be obtained by outdoor exposure tests (long natural weathering), devices with artificial light sources are generally use to accelerate the degradation. These light sources are filtered long arc xenon, fluorescent metal halide lamps and carbon arc lamps.

The acceleration occurs for several reasons, among them are: the tests can run continuously at naturally occurring or higher irradiance than solar radiation, uninterrupted by the natural day/night cycle, seasonal variations and weather conditions. Temperatures, thermal cycles, humidity, and water exposure also can be manipulated to reach a maximum. Specimens can be exposed to spectral energies at or beyond the limits of their intended service exposure. In addition, a fundamental benefit of a laboratory test is the reproducibility and repeatability.

The xenon long arc, when properly filtered, simulates UV and visible solar radiation more closely than any other artificial light source. Xenon arc is a precision gas discharge lamp in a sealed quartz tube. The spectral power distribution is altered through filtering to simulate solar radiation. It is widely preferred as a light source when the material to be tested will be exposed to natural sunlight.

For those reasons, a xenon long arc instrument produced by ATLAS (the weatherometer Suntest XXL – see Figure 27), was used in the ageing protocol for this research. In a previous research [27], a long ageing protocol was made for ageing porous asphalt mixture. The PAC specimens were aged for 1.000 hours , subjected to UV (60 W/m<sup>2</sup> in the range of 300-400 nm), 70% relative humidity, a room temperature of 40 °C resulting in a surface temperature of 70 °C. These parameters were decided based on the Dutch weather conditions, giving an ageing expectancy of about 10 to 12 years. Comparison with real in-situ core drilled 10 years aged PAC mixture showed that the ageing obtained in the laboratory was lower: 3 to 4 years [27].



Figure 27 - Weatherometer Suntest XXL - ATLAS [21].

# 4. Test results and analysis

In this chapter, the results of the tests will be presented, discussed and evaluated. Comparisons between the behaviour of C-Fix and normal bitumen will be made. In the research phase 1, the best dispersion of Altraflex-2006 was the main goal. Microscopy investigation, rheological and conventional binder tests were performed. The filler type and ratio investigation came in the second phase of the research.

## 4.1 – Microscopic investigation – phase 1

In the initial phase in this research, the optimum polymer content was investigated. To start the research, some literature review was done to check which amounts of polymer are already normally in use. Most of information found stated 6% as being a high polymer content and 3% a low polymer content [16]. As Altraflex-2006 is composed of around 50% SBS polymer, the maximum Altraflex-2006 amount to be added in the C-Fix modification was 12% by weight of binder. As a second amount of Altraflex-2006 for investigation, 6% was taken as the low amount.

The mixing temperature used was 200°C for the C-Fix. The main concern, quickly noticed after initial mixing trials, was the drop in temperature in the binder when adding Altraflex-2006 at room temperature. Indeed, the temperature dropped significantly after adding the polymer. This fact may not happen when applying Altraflex-2006 as it is meant to be used: directly added in the pug-mill as proposed by its manufacturer. When added in the pug-mill, the high temperature of the aggregates as well of the bitumen would prevent the big temperature drop encountered in the laboratory.

In reality the temperature drop can be expected to be much lower because in that case the Altraflex-2006 (amount of could material) is only around 0,36% of the total heated material being mixed in the pug-mill. While when mixing Altraflex-2006 with C-Fix in the laboratory, the cold material (Altraflex-2006) is 6% of the total hot C-Fix binder.

Indeed a big temperature drop was noticed, especially when adding 12% of Altraflex-2006. This led to temperature drops from 200°C to around 165°C, making the mixing procedure much more difficult and complicated due to the high viscosity of C-Fix at such a low temperature. Several times mixes needed to be repeated because polymer grains were still present at the end of the mixing procedure. At the end of the mixing procedures, the material should look homogeneous and with a continuous phase (bare eyes) for both 6 and 12% modified C-Fix.

The mixing procedure was carried out with a heavy duty laboratory high shear mixer Silverson model L4R. The rotation used was around 6.000 rpm at 200°C for 10 minutes for the two Altraflex-2006 amounts studied. In the preparation of the samples, 300 grams of C-Fix was heated to fluid condition (200°C) and poured into 900 ml cylindrical flask. Under controlled temperature and with the high shear mixer, a pre-weighted amount of Altraflex-2006 was added gradually to the binder. Mixing was performed then for 10 minutes. After mixing, samples were poured in aluminium trays to cool down to room temperature. The trays were labelled and stored in the freezer at -5°C to be used later.

Small pieces of the modified binder were broken later to be frozen up to -20°C. After being in the freezer for at least 3 hours (in this case most of the time at least one day), the samples were broken in small flat pieces with appropriate sharp tools. Flat surfaces were checked in the microscope. The method used was incident light microscopy and magnifications of 12,5 and 40 times could be performed in the microscope using UV light. As extra information, pure C-Fix and Altraflex-2006 were also assessed in the microscope. Examples of the results are given in Figure 28.



Figure 28 - UV microscope pictures

In Figure 28 (a), pure Altraflex-2006 is shown. The SBS polymer used in the Altraflex-2006 composition shows yellow fluorescence light when exposed at UV light. Figure 28 (b) shows Pure C-Fix. The C-Fix binder unexpectedly showed slightly little fluorescence as well as Altraflex-2006 under UV light.

The C-Fix modifications are showed in Figure 28 (c) -6% Altraflex-2006 modified C-Fix and Figure 28 (d) -12% Altraflex-2006 modified C-Fix. The Altraflex-2006 is dispersed well in C-Fix. There were no spots focused where it showed bigger droplets of polymer only. As mentioned before, the 12% Altraflex-2006 modified C-Fix was more difficult to mix, demanding initial mixing temperatures a little higher than for 6%. This would prevent the big drop in temperature during the mixing process. With extra care, the 12% Altraflex-2006 modified C-Fix showed enough good dispersion, providing good samples for the microscope investigation.

### 4.1.1 – Test results analysis

The morphology or compatibility is the result of the mutual effect of polymer and bitumen, and consequently is influenced by bitumen composition and polymer nature and content [16]. In general, at low polymer contents, the small polymer spheres swollen by bitumen compatible fractions (e.g. aromatics) are spread homogeneously in a continuous bitumen phase. Better dispersion of the polymers is also expected in bitumens with higher aromatic and lower asphaltene contents, according to Isacsson [16].

C-Fix, with its high asphaltene content is expected to have little solvency of polymers. With 30 to 35% asphaltene content C-Fix has, proportionally to normal grade bitumens, less saturates and aromatics which are the more compatible fractions to disperse the polymer added. This could explain the more complicated mixing procedure for the higher Altraflex-2006 content in this research. Also the Altraflex-2006 grains size seemed to be an inconvenient during mixing. Some polymer grains were, although partially melted, still present in smaller sizes than the original sizes added to the binder. Probably smaller sizes would make it easier to disperse.

According to NEN-EN 13632 [8], for 6% modified binder the distribution of the polymer can be classified as a continuous binder phase, homogeneous, small polymer particles size and roundish shape. For 12%, the homogeneity distorts more to what can be classified as medium size.

# 4.2 – Rheology – research phase 1

## 4.2.1 – Conventional binder tests

Conventional binder tests such as softening point and penetration were carried out to complement the microscopy investigation. The softening point and penetration determination were performed according to NEN-EN 1427 [11] and NEN-EN 1426 [9] respectively. For comparison, results obtained for bitumen grade 70/100 are also presented [3]. The results are shown in Table 6.

	Bitumen 70/100 [3]	C-Fix - C5	C-Fix + 6% Altraflex-2006	C-Fix +12% Altraflex-2006
Penetration 25°C [0,1mm]	64	5	10	10
Penetration 40°C [0,1mm]	301	15	20	22
Penetration 60°C [0,1mm]	-	38	47	49
Softening point [°C]	45,6	84	97	106
Penetration Index	-1,8	0,5	2,9	3,8
Density [Ton/m <sup>3</sup> ]	1030	1090	-	-

 Table 6 - Conventional test results for Pure and Altraflex-2006 modified C-Fix.

As penetration measurements at standard temperature (25°C) were very low, also measurements at 40 and 60°C were done. For more precision on the decision on the amount of Altraflex-2006 to be used in the second phase of the research, penetration tests were also performed on mixes with 2 and 4%. The results are presented in the Figure 29. There is an increase in the penetration with an increase of the modifier content with a more stable plateau between 6% and 12%. Higher amounts of modifier could give lower penetration values suggested by Gayle et al [17].

The softening point increased steadily with the increase in the modifier content. According to Whiteoak [2] the temperatures required to achieve a viscosity of 0,2 Pa s (viscosity at mixing temperature) can be crudely estimated by simply adding 110°C to the softening point. The expectation of lowering the mixing temperature based on this fact was not reached. The mixing temperature of the modified C-Fix shall be the same as that of pure C-Fix.



Modifier content [%] vs. Penetration [dmm]



Regarding the Penetration Index (PI) in Table 6, large improvement in deformation resistance (also called rutting) of the mixture produced with C-Fix binder can be expected compared with 70/100 bitumen. The Penetration is calculated based on the linearity of the penetration values for different temperatures and also the softening point temperature when plotted in the Bitumen Test Data Chart (Shell). This is clearly not the case for C-Fix as presented in the Figure 30. So, PI numbers in Table 6 are more a reference number.



Figure 30 - Penetration and softening point plotted in the Bitumen Test Data Chart.

Table 6 shows the higher PI of C-Fix compared to 70/100 bitumen. Since researches have proven that higher PI bitumens (here it should be read "binder") result in lower rut depth occurrences [2], increasing the PI of bitumen significantly improves the resistance to deformation. In this aspect, the use of modified C-Fix could strongly improve to the life span of the pavement regarding permanent deformation.

#### 4.2.1.1 - Test results analysis

According to the results above presented, C-Fix is indeed a hard binder. The high Penetration Index, in addition to the very low penetration values and high softening point corroborates this theory. When compared with bitumen, C-Fix has extremely low penetration and much higher softening point. Modification with Altraflex-2006 has resulted in higher penetration values.

### 4.2.2 – Dynamic shear rheometer test (DSR)

The dynamic shear rheometer was used to characterize the viscous and viscoelastic behaviour of the Pure, 6 and 12% Altraflex-2006 modified C-Fix. DSR tests were carried out according to the prNEN-14770 [12] using a Dynamic Shear Rheometer – AR200ex (TA Instruments). Parallel plates with a 2 mm gap, Ø 8 mm were used in a temperature range of -10°C to 60°C and a frequency sweep from 0,1 to 400 rad/s. The effect of thermal expansion of the instrument is deducted during the increase of temperature, keeping the normal force at zero by slightly increasing the gap.

The (loading) time/frequency and temperature are interrelated by the so called timetemperature superposition principle which states that the temperature will not change their frequency or time dependent behaviour, but will merely shift the frequency/time scale. This principle was used to build the master curves presented in the Figure 31 and Figure 32.



Figure 31 - Complex Shear Modulus (G\*) master curves at 20°C.

The Complex Shear Modulus (G\*) and Phase angle ( $\delta$ ) are plotted versus the frequency. There is a decrease of the complex shear modulus for both modified binders (6 and 12%). The 6% modified binder has an almost constant parallel complex shear modulus line, just shifted to lower values. The lower values of complex shear modulus indicates a higher flexibility of the modified binder which could indicate a less brittle behaviour at low temperatures. Comparison of the master curves for G\* of pure and modified C-Fix has shown that there is a bigger change in the properties of the materials at low frequencies (long loading time – high temperatures).



Figure 32 - Phase Angle (δ) master curves at 20°C.

Also the phase angle has showed a bigger change at low frequencies or high temperatures. Figure 32 shows that in the low frequency range the phase angle for the 12% modified binder is lower than that of pure binder. This characteristic shows that the modified C-Fix is less viscous under long loading time, improving its permanent deformation resistance due to more elastic behaviour. For the 6% modified binder, the change is less pronounced, but still more elasticity is present at low frequencies.

#### 4.2.2.1 - Test results analysis

Ideally modifying binders with polymers should result in a lower viscosity (more flexibility) at low service temperatures reducing the risk of low temperature cracking, and higher viscosity at elevated temperatures leading to a reduced formation of permanent deformation.

For C-Fix binder, the ideal situation would be to obtain a lower phase angle in the lower frequency range. For the complex shear modulus, lower values at higher frequencies would give the material less brittle behaviour at low temperatures. As this binder has a high stiffness, even a lower stiffness due to polymer modification at lower frequencies would not result in much decrease in permanent deformation resistance at high temperatures.

The modification of C-Fix assessed with DSR showed that both percentages used (6 and 12%) resulted in a decrease in the complex shear modulus (G\*) giving a less stiff material. This change in itself would be good for the C-Fix binder as it has a high modulus of elasticity, with a brittle behaviour at low temperatures. With a less stiff material, improvement on the brittle behaviour could be expected.

The phase angle ( $\delta$ ) has a more remarkable change for the 12% modification sample, presenting lower phase angle at low frequency (high temperature/long loading time), indicating a less viscous behaviour. For the 6% modified C-Fix, only higher phase angles were measured in the frequency range of interest (table in the Figure 32). It would have been better if the range of lower phase angle for the modified C-Fix (both percentages of Altraflex-2006) would reach higher frequencies, when compared with the unmodified binder. This could improve the C-Fix brittle behaviour more effectively.

### 4.3 – Rheology – research phase 2

The DRS master curves clearly showed reduced complex shear modulus at high frequencies (low temperature) for the modified binder. The microscopic investigation and the conventional binder tests together with the DSR results supported the decisions made for

further steps in this research. The microscopic pictures of the 12% modified binder showed bigger polymer particles, which could imply an inhomogeneous modification. During mixing, some difficulties in dissolving high contents of Altraflex-2006 in the binder were encountered. Some grains were still visible at the end of the mixing procedure. In the DSR tests, not much difference between 6 and 12% was visible regarding the complex shear modulus. The higher the polymer content, the higher the softening point of the modified binder, but the softening point is already very high. For these reasons 6% Altraflex-2006 modified C-Fix was chosen for further investigation in the phase 2 in this research.

#### Filler

For the 6% modified binder, different filler types and different filler/binder ratios were studied. The filler Wigro 60K was chosen based on requirements in NEN-EN 13043 [25]. Wigro 60K has the minimum calcium hydroxide content required for medium type and active fillers, to be used in porous asphalt. The filler Duras 15 was chosen because it is a kind of inert or weak filler. The C-Fix binder has higher stiffness in itself with higher modulus of elasticity when compared with bitumen and lower penetration values. When modified, a higher Penetration Index (PI) was calculated, implying higher resistance to permanent deformation. A weak filler also absorbs less maltenes from the C-Fix Binder in contrast with Wigro 60K filler, not decreasing the viscoelastic properties. These reasons supported the idea that the binder is already "strong" enough to withstand the loading in a pavement, not needing strong fillers to improve its stiffness. In a worst case scenario, an active filler could make the binder even more brittle.

Different filler binder ratios were used to check the possibility of decreasing the amount of filler which is, according to the RAW Standard [6], needed in the asphalt. In the porous asphalt gradation in this same standard, the amount of filler prescribed by weight is the same as the prescribed amount of binder. For normal bitumen it is reasonable to do so, but for a hard binder such as C-Fix it could make it even more brittle. Therefore it was decided to investigate lower filler contents. Filler/binder ratios of 1,0 and 0,5 were used in this investigation for both filler types.

The influence of filler was assessed by means of dynamic mechanical analysis (DMA) and direct tension tests (DTT). These tests were also performed on pure and 6% Altraflex-2006 modified C-Fix.

### 4.3.3 – Dynamic mechanical analysis (DMA)

DMA tests were performed according to the ASTM E1640 [13]. This test was done for the pure binder, 6% modified binder and mastics which had the following composition:

- 6% modified binder plus Wigro 60K filler/binder ratio 1,0;
- 6% modified binder plus Wigro 60K filler/binder ratio 0,5;
- 6% modified binder plus Duras 15 filler/binder ratio 1,0;
- 6% modified binder plus Duras 15 filler/binder ratio 0,5.

DMA tests with constant frequency (1 Hz) and temperature increase of 1°C/min (ranges varying according to the material from -50°C to 20°C) and a displacement amplitude (varying according to the material from 5  $\mu$ m to 15  $\mu$ m) were carried out using a Rheolyst Rheometer serie AR1000 (TA Instruments). Specimens with 59x12,75x3 mm (LxWxT) were prepared in silicon moulds and tested accordingly. Depending on the material tested such as the pure binder, the temperature and displacement amplitude should be reduced. The brittleness of the material at low temperature made the pure binder samples more susceptible to failure when tested at extremely low temperatures (-40 to -50°C) and higher displacement amplitudes (10-15  $\mu$ m). The 6% modified binder behaved comparably in the same manner as the pure binder. For the mastics, higher amplitudes and lower temperatures were suitable.

The results are presented in the Figure 33 to Figure 35. The 6% modified C-Fix binder appeared to have a lower transition temperature than the pure binder. It shows a possible improvement in the low temperature behaviour of C-Fix. The transition temperature dropped from around 0°C for pure binder to -8°C for the Altraflex-2006 modified binder. This indicates a lower temperature at which the material starts to behave more as a solid than as a viscoelastic material. This lower transition temperature would make it possible to use modified C-Fix at lower temperature ranges. This would possibly make the modified C-Fix better suitable for asphalt mixtures in regions with mild winters than the pure binder.

When comparing with other research transition temperature measurements, the values were considerably different. By Khedoe [3] (see table in the detail at Figure 33), the transition temperature found for pure C-Fix (or C-5) was 12,6°C compared to the zero degree found in this research. Also no influence of the filler/binder (f/b) ratio was present for the mastic produced with Wigro 60K, while here a considerable difference in transition temperatures was found for the same filler and different f/b ratios.



Figure 33 - Transition temperature for pure C-Fix, 6% Altraflex-2006 modified C-Fix and the mastics.

For the mastics tested, the medium type filler Wigro 60K with calcium hydroxide gave lower transition temperature than the weak filler Duras 15. The higher the filler content the lower the transition temperature for the Wigro 60K filler. For the Duras 15 the opposite was noticed: the higher the filler content, the higher the transition temperature.



Figure 34 – Temperature (°C) vs. Loss modulus (MPa).

The complex modulus can be defined as the measure of the overall resistance to deformation of a material. As shown in section 3.4.3, it can be split into a storage modulus (related to elastic behaviour) and a loss modulus (related to viscous behaviour). Figure 34 shows that loss modulus of the mastics starts to increase at lower temperature than the pure binder. When the loss modulus starts to increase, that is the point when molecules are more free to move along the displacement imposed to the specimen enabling a so called rearrangement of the molecules and giving some ductility to the material; this was discussed in the section 3.4.4. This would give the mastics better relaxation properties at lower temperature compared to the pure binder. For the 6% modified binder also a displacement to the left of the point in temperature where the loss modulus starts to increase is visible which could result in an earlier (in temperature) viscoelastic behaviour when compared to the pure binder.



Figure 35 - Temperature (°C) vs. Storage Modulus (MPa).

In the storage modulus graph (Figure 35) the transition temperature could be determined. It is defined as the point where there is a big drop in the storage modulus, together with an increase in the loss modulus. This is the definition for the Glass Transition Temperature according to the Standard ASTM E1640 [13]. Although clearly specified, some difficulties were encountered to precisely define this point because of the atypical shape of the storage modulus graphs such as an increase of it before the expected big drop. For this reason the temperature found in this research is being named "transition temperature", as a temperature where the binder is expected to have a more viscous behaviour than solid or brittle.

#### 4.3.3.1 - Test results analysis

The storage modulus of the modified binder has a drop point at a lower temperature than for the pure binder. This could give an improvement on its low temperature behaviour.

The differences in the filler types used are also visible. Although Duras 15 showed a lower performance for every filler/binder ratio, it is still close to the Wigro 60K results. The use of a filler in fact lowers the transition temperature for the modified binder which could indicate a better fatigue behaviour of the mastic.

### 4.4 – Direct Tensile Tests (DTT)

DTT tests were performed according to the ASTM D6723-02 [15] aiming to determine the stress/strain curve and the stress/strain at failure. This test was done for the pure binder, 6% modified binder and the same mastics as used for the DMA tests. In addition, for comparison, tests with mastic using filler Duras 15 (f/b=1,0) and no polymer modification were performed.

Specimens were prepared using a silicon mould. After heating the mixed samples, they were poured in the silicon mould and covered with another silicone plate allowing the surplus of mastic/binder to overflow. After cooling down to room temperatures, the specimens were trimmed and measured. Not having enough end supports for the specimens, it was not possible to prepare specimens for the three different temperatures (-10°C, 0°C and -10 °C) tested at the same time. For each sample tested, the specimens were prepared twice or three times to be tested in three different temperatures. This could give some statistical deviation of the results.

According to the ASTM D6723-02 [15] at least six specimens should be tested and the best four results taken to calculate the average stress and strain at failure. Due to the Altraflex-2006 dispersion instability in C-Fix and C-Fix brittleness properties, a much higher number of tests were needed to have reasonable results. The mixture of Altraflex-2006 and C-Fix can not be in the oven for a period longer than 10 minutes. This would cause phase separation of the two components. So, when melting the Altraflex-2006 modified C-Fix to pour in the silicon moulds, extra care had to be taken in order to have a good mixed sample of the modified binder in the specimens. Also, the brittleness of the C-Fix material makes more difficult to deal with small and delicate specimens. The cleaning and measurements of the specimens demand extreme care, otherwise micro cracks could be present, distorting the end results. When tests needed to be redone, the whole batch for that sample and temperature was repeated. The complete test data set of the valid results is presented in the Appendix 1.

Those reasons above mentioned explain the high number of tests done for some samples. Finally, at least six specimens were tested for each one of the binders and mastics. The average of the failure stress and strain of the four best results measured for each sample are presented in Table 7.

		DTT					
	Temperatur	$\sigma_f$	$\sigma_f$ Std.	٤ <sub>f</sub>	ε <sub>f</sub>	Toughnes	#
	е	mean	Dev.	mean	Std.Dev.	S	samples
	[ºC]	[Mpa]		[%]		[GJ/m <sup>3</sup> ]	tested
Pure C-Fix		1,060	0,13	0,560	0,12	4,318	11
6% Altraflex-2006 modif. C-Fix		1,082	0,16	0,798	0,10	5,535	14
Mastic Wigro 60K - f/b=1,0		2,475	0,43	0,656	0,21	15,548	9
Mastic Wigro 60K - f/b=0,5	-10ºC	2,208	0,24	0,775	0,07	9,960	14
Mastic Duras 15 - f/b=1,0		2,467	0,30	0,826	0,16	13,820	6
Mastic Duras 15 - f/b=0,5		1,659	0,11	0,829	0,05	7,700	10
Mastic Duras 15 - f/b=1,0 - NO POLYMER		2,551	0,23	0,883	0,06	13,157	5
Pure C-Fix		1,112	0,15	1,167	0,13	8,440	6
6% Altraflex-2006 modif. C-Fix		1,250	0,07	1,807	0,02	11,557	8
Mastic Wigro 60K - f/b=1,0	0°C	2,782	0,78	1,048	0,20	23,986	9
Mastic Wigro 60K - f/b=0,5		2,076	0,31	1,295	0,20	18,000	11
Mastic Duras 15 - f/b=1,0		2,415	0,60	1,187	0,37	26,160	5
Mastic Duras 15 - f/b=0,5		1,651	0,09	1,377	0,17	14,330	6
Mastic Duras 15 - f/b=1,0 - NO POLYMER		2,709	0,53	1,361	0,17	24,025	6
Pure C-Fix		1,001	0,01	2,916	0,10	15,300	6
6% Altraflex-2006 modif. C-Fix		0,846	0,06	3,362	0,56	21,736	10
Mastic Wigro 60K - f/b=1,0		2,998	0,32	1,578	0,10	26,964	10
Mastic Wigro 60K - f/b=0,5	+10°C	1,717	0,00	2,065	0,00	31,050	14
Mastic Duras 15 - f/b=1,0		2,465	0,05	2,598	0,15	37,240	6
Mastic Duras 15 - f/b=0,5		1,478	0,24	3,689	0,44	37,310	8
Mastic Duras 15 - f/b=1,0 - NO POLYMER		2,480	0,21	2,804	0,45	47,540	8

Table 7 - Direct Tensile Test results.

For clearer visualization, graphics (Figure 36 to Figure 38) are plotted for each different temperature. Comparison of the pure and 6% modified binder shows that more ductility is present in the modified version for each temperature tested. It is reachable higher level of strain comparing at the same temperature. In fact, not much higher stresses are found in the modified binder and even lower for the  $+10^{\circ}$ C temperature.



Figure 36 - DTT test results for -10°C.

Adding filler to the modified binder mostly increased the stress at failure. When comparing the two different filler types used, almost similar results were obtained for both of them. The absence of calcium hydroxide does not influence too much the test results. For the low temperature and f/b=1,0 basically same results in strength were measured.



Figure 37 - DTT test results for 0°C.

At 10°C a slightly higher strength of Wigro 60K filler is noticed when compared to the 0°C and -10°C results, but much higher ductility of the Duras 15 filler. This could improve the permanent deformation resistance for Duras 15 mastic at high serviceability temperatures.



Figure 38 - DTT test results for 10°C.

Using the same DTT results, an approximated calculation of the toughness of the binder and mastic was possible. It is an approximation because although most of the stress/strain curves were linear, a slight non linear behaviour was observed at higher temperatures. Examples of stress/strain curves are presented in Figure 39 to clarify this.











Figure 39 - Stress/strain curve for each binder and mastic assessed in the DTT test set-up for -10°C, 0°C and +10°C.

This slightly non linear behaviour allowed for a direct approximation considering all the stress/strain curves to be linear in the calculation of the toughness. The toughness is calculated based on the average of the best four test results. Toughness is defined as the energy the material can absorb prior to rupture. It is calculated by determining the area under the stress/strain curve as indicated in the Figure 22. The results are presented in Figure 40.



Figure 40 - Calculated toughness using DTT test results.

Adding Altraflex-2006 to C-Fix has increased its toughness for each temperature investigated. The toughness of the mastics is higher than that of the pure and modified binder. Wigro 60K has given higher toughness than Duras 15 for -10°C. Instead, for +10°C Wigro 60K has a lower toughness than Duras 15, probably because calcium hydroxide would be more active at this temperature. This would give higher stiffness to the mastic, but lower ductility decreasing the strain level at breaking point.

### 4.4.1 - Test results analysis

Adding filler to the modified binder, increased the stress and strain at failure. The filler is expected to fill the voids completing the stone gradation in the smallest size, and form a mastic seen as a bonding agent increasing its strength, improving the viscous behaviour of this mastic, which increases stability of the asphalt concrete mixture [10]. Calcium hydroxide in the filler is expected to prevent stripping of the asphalt mixture and make the filler less sensitive to the influence of moisture. Greater water sensitivity of the filler would lead to damage of the road

surface. That is the reason why in the Standard RAW it is prescribed the use of a medium filler with calcium hydroxide for porous asphalt concrete.

At low temperatures, the modified binder presented a slight higher toughness than the pure C-Fix. For each filler type, a higher filler content resulted in a higher toughness at low temperatures (-10°C and 0°C).

From the results obtained for Wigro 60K, at -10°C and 0°C it was observed that the higher the filler content the higher the toughness. For +10°C the opposite is noticed. This is possibly due to the high calcium hydroxide content being more active at higher temperatures. Compared with Duras 15, the toughness of mastics with Wigro 60K was higher at the lower temperature tested (-10°C). At higher temperatures the toughness of Wigro 60K mastics was lower than Duras 15 mastics.

From the results obtained for Duras 15, at -10°C and 0°C it is also observed that a higher filler content resulted in a higher toughness. For +10°C basically the same toughness level was measured. Tests were also done with a mastic sample without Altraflex-2006, with Duras 15 (f/b=1,0). For this mastic, at low temperature (-10°C and 0°C) a slightly lower toughness was obtained probably due to the absence of Altraflex-2006. A much higher toughness level was obtained at the higher temperature (+10°C).

The addition of filler has increased the toughness of the binder. Not much difference was noticed between the Wigro 60K and Duras 15. For C-Fix binder, the calcium hydroxide does not seem to make a substantial difference. For this reason, the Duras 15 mastic (f/b=1,0) was applied in the relaxation tests. The same filler binder ratio prescribed in the Dutch standard is kept, but no calcium hydroxide is added.

### 4.5 – Relaxation test

Relaxation tests give information on the material stress dissipation properties. Relaxation behaviour is important because stress can be dissipated due to thermal and/or traffic loading, decreasing the residual stress present in the mastic or binder. This would prevent or minimize the cracking formation. It may give also information on the healing properties of the binder and mastic.

Based on the results of the DMA and DTT tests, the pure and 6% Altraflex-2006 modified C-Fix was used for the relaxation tests. Also the Duras 15 mastic (f/b=1,0) with 6% Altraflex-2006 modified C-Fix was tested.

Strain controlled tests were performed in the following way. A small displacement (from 15 until 24% of the breaking strain) is applied and kept constant. Due to this displacement, a tensile stress is developed and this stress is then monitored for a certain time (in this case 10 minutes). This test was performed in the Direct Tensile Test set-up. As the relaxation shape curve depends on the speed of the applied deformation, three different strain speed application levels were used:

- low 1 mm/min;
- medium 10 mm/min;
- high 300 mm/min.

The relaxation is plotted as a percentage of the maximum measured stress during the test for each binder and mastic. This makes comparison easier and clearer. The lower the temperature, the lower the strain range that can be applied. Due to the possible molecules rearrangement in the low speed strain application, in general a higher strain levels were feasible. For higher speeds, normally lower strain levels were reachable and higher stresses developed. Table 8 shows the results for the samples tested.

	Relaxation	n after 10 i	minutes			
	Temperature	Speed	Displacement	Area	Max.Sress	Relaxation
	[ºC]	[mm/min]	[mm]	[mm²]	[MPa]	[%]
		1	0,05	36,3	0,580	68
Pure C-Fix		10	0,04	36,3	0,342	75
		300	0,04	35,8	0,829	63
		1	0,07	36,3	0,672	73
6% Altraflex-2006 modif. C-Fix	-10°C	10	0,07	36,6	0,751	78
		300	0,07	36,3	0,807	75
		1	0,07	38,3	1,381	49
Mastic Duras 15 - f/b=1,0		10	0,07	38,3	1,079	44
		300	0,07	37,6	1,621	41
		1	0,08	35,9	0,590	81
Pure C-Fix		10	0,07	35,9	0,572	88
		300	0,08	37,4	0,764	86
		1	0,11	36,0	0,388	72
6% Altraflex-2006 modif. C-Fix	0°C	10	0,11	36,0	0,447	74
		300	0,12	36,7	1,139	85
		1	0,09	38,1	0,989	68
Mastic Duras 15 - f/b=1,0		10	0,09	38,1	1,484	71
		300	0,09	38,1	1,813	70
		1	0,18	37,3	0,481	85
Pure C-Fix		10	0,17	37,3	0,743	94
		300	0,17	37,3	0,767	90
		1	0,21	35,4	0,472	80
6% Altraflex-2006 modif. C-Fix	+10°C	10	0,21	35,4	0,743	85
		300	0,21	35,4	0,878	87
		1	0,17	38,7	1,329	85
Mastic Duras 15 - f/b=1,0		10	0,17	38,7	1,817	82
		300	0,17	38,7	1,817	85

Table 8 - Relaxation test results.

Adding Altraflex-2006 to the C-Fix binder has increased relaxation ranges at low temperature (-10°C) and decreased at higher temperatures (0°C and 10°C). This is in fact what it should be expected when modifying binders with polymer. The addition of Duras Filler 15 to the mastic has decreased the relaxation at low temperature, but has kept the same relaxation ranges for higher temperatures. The displacement rate did not have much influence in the end relaxation ranges results. Basically the end relaxation was in the same range for the same temperature and material, but different displacement speed applied. Figure 41 to Figure 43 show the relaxation plots for low strain speed application at different temperatures. Test results obtained at other displacement rates are presented in the Appendix 2.



Figure 41 - Relaxation test at -10°C - low loading speed (1 mm/min).

Surprisingly, the relaxation plots revealed some unexpected results mostly at the low temperature tested. As seen in the Figure 41 and Figure 42, there is as increase in the stress level during the relaxation test. This phenomenon happened mostly at -10°C, although some similar behaviour happened also at 0°C.



Figure 42 - Relaxation test at 0°C - low loading speed (1 mm/min).

Due to different displacement rate, a different response of the material is obtained.



Figure 43 - Relaxation test at 10°C - low loading speed (1 mm/min).

When compared with a normal grade bitumen (70/100) the relaxation level of the C-Fix binder is relatively low. Even at -10°C (loading speed of 3 mm/min) for the previous cited bitumen, the relaxation reaches 100% after 5 minutes [3]. For C-Fix binder, this amounts 68 to 75%. Modifying the C-Fix with Altraflex-2006 has increased the relaxation ranges for this low temperature to amounts like 73 to 78% (see Table 8 for details). This fact could be important in the cracking sensitivity of the C-Fix binder.

A comparison could be made between the relaxation results obtained in these sets of experiments (for pure and 6% modified C-Fix) with relaxation tests done on blended C-Fix in other researches. In the research by Khedoe [3], the temperature used was -10°C and the speed 0,3 mm/min for the binder C-9. In this blended C-Fix binder, 20% of its content is composed out of bitumen 70/100. This blend produces a softer C-Fix, with higher penetration values and lower softening point. For C-9 binder, the relaxation level obtained was around 56% after 10 minutes. For pure C-Fix (or C-5) at the same temperature and displacement rate around 3 times bigger than the one used by Khedoe (i.e. 1 mm/min), the relaxation was 68% for pure C-Fix (C-5) and 73% for 6% Altraflex-2006 modified binder after the same 10 minutes. Considering the fact that the blended C-Fix is softer than pure C-Fix, lower relaxation should be expected for the results in C-5 tests.

The mastic used in Khedoe's research was mixed using Wigro 60K (with C-9) instead of Duras 15 (with C-5) as used here for relaxation tests. In this sense, for the same temperature and a lower displacement rate used for C-9, much lower relaxation was measured for Wigro 60K mastic (with C-9) than the one for Duras 15 mastic: 25% against 49% in this research. This could be due to the more active filler used, which makes the mastic much stiffer than the weak filler Duras 15, even if the calcium hydroxide is supposed to be less active at lower temperature ranges.

### 4.5.1 - Test results analysis

According to the Table 8, addition of Altraflex-2006 has increased somewhat the end relaxation at -10°C, although unexpected patterns were monitored in the relaxation tests at low temperature. When compared with normal grade bitumen such as 70/100, even the polymer modified C-Fix has much less relaxation properties. The bitumen relaxes completely after 5 minutes, while the 6%

Altraflex-2006 modified C-Fix reaches after 10 minutes 78% of relaxation at the same test temperature.

The extra ability of relaxing the thermal and traffic loads could possibly give the modified C-Fix binder less residual stresses resulting in possible less cracking formation. The same is valid for the mastic prepared with Duras filler 15.

### 4.6 – Adhesion zone fatigue test (Stone columns test)

This test is meant to model the adhesion zone between the mastic and the aggregates, giving information on the fatigue behaviour of the binder tested. The samples used were the Pure and 6% Altraflex-2006 modified C-Fix. Greywacke was the stone used in the test, after being prepared as described in section 3.7. Tests were performed using a 40 Hz full sine load at 10°C, with different torque levels aiming to have three test durations: short, medium and long [21]. The results of the tests are presented in the Table 9.

Material	Test duration	Torque	Time to failure	Upper/lower stone diameter	Initial raw phase	Final raw phase	Initial displ.	Final displ.
		[µ.N.m]	[Hours]	[mm]	[°]	[°]	[rad]	[rad]
	T	100	10,96	-	3,798	8,598	1,38E-03	2,38E-03
	Long duration	100	12,73	6,76/6,84	3,784	8,653	1,41E-03	2,05E-03
		100	2,03	6,86/6,90	4,521	8,982	1,44E-03	2,08E-03
		130	2,23	-	4,681	9,331	1,79E-03	2,58E-03
		130	0,88	-	3,680	7,398	1,75E-03	2,80E-03
		130	1,43	-	3,929	7,046	1,71E-03	2,42E-03
Duro C-Fiv	Medium	130	6,76	6,90/0,00	4,195	7,542	1,81E-03	2,35E-03
	duration	130	1,65	6,86/6,88	4,134	7,871	1,76E-03	2,58E-03
		130	14,89	6,80/6,74	4,250	8,637	2,08E-03	2,63E-03
		130	0,67	6,88/6,86	3,890	7,662	1,65E-03	2,55E-03
		130	0,45	6,86/6,88	4,566	7,445	1,84E-03	2,40E-03
		170	0,11	-	4,866	5,710	2,55E-03	2,70E-03
	Short duration	170	0,03	6,78/0,00	4,409	4,362	2,78E-03	2,85E-03
		170	0,50	6,86/6,90	4,479	5,766	2,32E-03	2,53E-03
L	Long duration	100	4,23	6,86/6,82	5,143	19,830	1,53E-03	2,31E-03
		100	9,89	6,88/6,88	4,896	21,410	1,56E-03	2,47E-03
		100	8,98	6,82/6,84	5,170	21,490	1,50E-03	2,52E-03
		130	0,46	6,76/6,74	6,294	14,980	2,06E-03	3,16E-03
		130	0,09	6,88/6,90	5,783	10,800	2,00E-03	2,80E-03
6%	Modium	130	0,37	6,84/6,82	5,558	11,730	1,97E-03	2,69E-03
Altrafiex-	Mealum	130	0,75	6,78/6,78	5,167	12,770	2,08E-03	3,14E-03
modified	duration	130	0,10	6,88/6,90	6,270	12,000	2,02E-03	2,96E-03
C-Fix		130	1,63	6,88/6,80	5,141	18,050	1,87E-03	3,02E-03
		130	0,25	6,94/6,88	5,788	13,280	1,99E-03	3,17E-03
		170	0,01	6,78/6,78	7,335	8,437	3,34E-03	3,54E-03
	Short duration	170	0,03	6,88/6,88	7,004	10,040	2,54E-03	3,01E-03
		170	0,04	6,88/6,88	6,090	9,669	2,58E-03	3,02E-03
		170	0,05	6,88/6,88	6,573	10,460	2,65E-03	3,22E-03

Table 9 - Stone columns fatigue test results.

As it can be noticed, the duration of the test until failure obtained did not show a regular pattern. This is the case for the pure C-Fix as well as for the 6% Altraflex-2006 modified C-Fix. There was a big scatter in failure time for the same torque level, possibly due to test uncontrollable variables such as the stone column diameter, the parallelism of the surfaces in contact with the binder film, the perfect alignment of the stone cores when preparing the specimens as well as when performing the test, etc. Those variables are still being studied regarding the repeatability of the stone columns test.

If a plot is made of the data based on the failure time, the 6% modified C-Fix presents on average a lower failure time than the pure C-Fix binder. Figure 44 shows the results. Modifying C-Fix with a thermoplastic elastomer could bring expectations on higher failure time than the pure C-Fix, due to the elastic behaviour of the polymer. This would possibly decrease the crack formation, or delay it, giving the material better fatigue resistance. This behaviour was not measured during the test.



Figure 44 - Stone column test results: Failure time [h] vs. torque applied [µ.N.m].

Figure 45 shows how the raw phase angle varies during the adhesion zone fatigue test performed on pure and 6% Altraflex-2006 modified C-Fix. The raw phase angle is the phase angle directly measured by the DSR equipment on which no correction factor is used (for instance to deduct the equipment inertia).

Figure 45 shows that the phase angle remains rather constant for the fatigue tests performed on the stone columns/pure C-Fix samples while for the stone columns/modified C-Fix samples mostly a gradual increase of the phase angle with time is observed. Clearly on the long duration test.



Figure 45 - Failure time vs. Raw phase angle.

The facts discussed in Figure 45 are presented in a normalized graph in the Figure 46. It is the same data, only the horizontal axis is made dimensionless. For medium and short duration 6% Altraflex-2006 modified C-Fix test, the increase in the raw phase angle along the test is much less pronounced as mentioned previously. The fact that the phase angle gradually increases during the fatigue test on the modified C-Fix indicates that damage is developing early during the test and gradually increases. The pure C-Fix binder constant phase angles measurements with a sudden and fast failure at the end could indicate the brittle type of failure of the adhesion zone. If both binders really behave differently under repeated load in the adhesion zone is difficult to confirm based on these tests results.



Figure 46 - Normalized number of cycles vs. Raw phase angle.

Another approach to assess the test output data can be the calculation of the dissipated energy. To determine the dissipated energy, thee components must be known: stress, strain and phase angle between stress and strain. The shear stress can be calculated for stone column DSR testing:

$$\tau = \frac{2.T}{\pi . r^3}$$

Where:

 $\tau$  = Maximum shear stress [MPa];

T = Applied torque [N.mm];

r = specimen radius [mm].

The strain can be calculated by:

$$\gamma = \frac{r.\varphi}{h}$$

Where:

 $\gamma$  = Maximum shear strain [-];

 $\varphi$  = Rotational angle [rad];

h = bitumen film thickness between two stone columns (here assumed 15 µm).

The dissipated energy per cycle thus can be determined using following equation:

 $w_i = \pi . \tau_i . \gamma_i . \sin \delta_i$ 

Where:

 $w_i$  = Dissipated energy during ith cycle [MPa];

 $\tau_i$  = Maximum shear stress or stress amplitude during ith cycle [MPa];

 $\gamma_i$  = Maximum shear strain or strain amplitude during ith cycle [MPa];

 $\delta_i$  = Phase angle between strain and stress amplitude during ith cycle [rad].

It is commonly believed that dissipated energy is for the larger part translated into heat. A smaller part of it is associated with material damage. The increase in dissipated energy is often used by researchers to explain fatigue damage development [23]

The rotational deformation and the measured phase angle tend to slowly increase during the fatigue tests. Limited changes are visible at the beginning of the test, but a gradual increase of the dissipated energy per cycle  $w_i$  develops with increasing the number of load repetitions. At the

end of the test  $w_i$  increases exponentially [23].

Figure 47 shows the results of the calculated total dissipated energy for all the tests performed with pure and 6% Altraflex-2006 modified C-Fix. The graph clearly shows that more energy is dissipated in the fatigue tests on the 6% modified C-fix.



Figure 47 - Total dissipated energy per specimen tested vs. Number of cycles for pure and 6% Altraflex-2006 modified C-Fix.

When the dissipated energy per cycle is plotted for both binders, again the 6% Altraflex-2006 modified C-Fix shows a gradual increase of the dissipated energy. Plus, it indicates the more brittle behaviour of pure C-Fix. See Figure 48.



Figure 48 - Increase in the dissipated energy per cycle along the test duration for pure and 6% Altraflex-2006 modified C-Fix.

### 4.6.1 - Test results analysis

In this test, the expected improvement in the fatigue behaviour of the 6% modified C-Fix was not measured. In contrary, the modified binder presented slightly worse results regarding the fatigue life than the pure binder, with earlier viscous behaviour leading to shorter failure time in general. The calculated dissipated energy corroborated the same results. An early and higher increase rate in the dissipated energy is present for the 6% Altraflex-2006 modified binder.

As mentioned before, this tests might have still uncontrolled variables which compromises its repeatability. Further tests could give better results and better understanding in the fatigue behaviour of the C-Fix binder with the Altraflex-2006 modification, but time constrictions limited

the test period. As C-Fix has a high transition temperature and higher stiffness when compared with bitumen, it has also a more brittle behaviour, which was translated in a more difficult pattern development for the fatigue test. This could be due to high crack sensitivity at higher temperatures than when compared with bitumen, which could demand special treatment when dealing with fatigue test.

# 4.7 – Ageing tests

Hardening of the binder is most of the time referred to as ageing. The hardening of the binder results in a decrease in penetration, an increase in softening point and an increase in penetration index (PI). Binders are affected by the presence of oxygen, ultra-violet radiation and by changes in temperature.

Four main mechanisms of bitumen hardening have been identified being:

- oxidation;
- loss of volatiles;
- physical hardening;
- exudative hardening.

The ageing procedure adopted, as explained in chapter three, made use of the weatherometer manufactured by ATLAS. This equipment reproduces outside temperatures, moisture and sunlight providing a very useful tool for speeding up log-term ageing of binder and polymers.

The two samples aged in this research were:

- 6% Altraflex-2006 modified binder;
- 6% Altraflex-2006 modified binder + Wigro 60K (f/b=1,0).

The samples were aged for 1.000 hours , subjected to UV ( $60 \text{ W/m}^2$  in the range of 300-400 nm), 70% relative humidity, a room temperature of 40°C resulting in a surface temperature of 70°C. These parameters were decided based on the Dutch weather conditions, and comparison of the artificial laboratory ageing results with real in-situ aged materials has proven them to be related to 3 to 4 years ageing [27]. These details are given in the section 3.8.3.

The procedure used at LOT project [21] at TU Delft was chosen for the samples preparation. A 2 mm thin layer of the binder or mastic should be prepared and laid on a silicone paper tray. The tray dimensions are dependent on the amount of material to be aged and the space availability in the laboratory weatherometer. According to the Suntest XXL dimensions, it was possible to make two trays of 30x30 cm. After mixing, the samples were poured in the pre-weighed paper tray on top of a scale. The exact amount in order to have those 2 mm thickness is placed based on the controlled weight. Figure 49 shows an example of the sample preparation.



Figure 49 - Sample preparation for ageing procedure [21].

The results are presented in the Table 10, Figure 50 and Figure 51. There was a considerable decrease in the penetration values for every temperature, and a remarkable increase in the softening point.

	Bitumen 70/100 [3]	C-Fix - C5	C-Fix + 6% Altraflex- 2006	C-Fix +12% Altraflex- 2006	1000 hours aged C-Fix + 6% Altraflex- 2006
Penetration 25°C [0,1mm]	64	5	10	10	5
Penetration 40°C [0,1mm]	301	15	20	22	10
Penetration 60°C [0,1mm]	-	38	47	49	29
Softening point [°C]	45,6	84	97	106	111
Penetration Index	-1,8	0,5	2,9	3,8	3,1
Density [Ton/m <sup>3</sup> ]	1030	1090	-	-	-

 Table 10 - Comparison with unaged conventional binder tests results and 1000 hours aged 6% Altraflex-2006 modified C-FIx.

The rheological changes for the 6% Altraflex-2006 modified C-Fix (see Table 10 and Figure 50) and the mastic with Wigro 60K (see Figure 51) correspond to typical oxidation hardening and volatile loss following an ageing procedure: increase of complex shear modulus and reduction of phase angle.



Figure 50 - Complex shear modulus and phase angle master curves for unaged and 1000 hours aged 6% modified C-Fix at 20°C.

For the mastic, although the typical hardening has also occurred, the filler might have influenced in the less pronounced phase angle decrease and complex shear modulus increase.



Figure 51 - Complex shear modlus and phase angle master curves for unaged and 1000 hours aged 6% Altraflex-2006 modified C-Fix + Wigro 60K at 20°C.

For comparison purposes, the data from Figure 50 (master curves for aged and unaged 6% Altraflex-2006 modified C-Fix) are shown in the Figure 52 together with the phase angle and complex shear modulus master curves for pure C-Fix in the. The aged 6% modified C-Fix complex shear modulus master curve has moved in the direction of the pure C-Fix binder master curve. At very low frequency ranges only, it overtakes the values for pure C-Fix binder.



Figure 52 - Complex shear modulus and phase angle master curves at 20°C -Comparison between pure C-fix and 6% Altraflex-2006 modified C-Fix (aged and unaged).

# 4.7.1 - Test results analysis

C-Fix, when compared with bitumen 70/100, has shown to be more resistant to long term ageing than the bitumen [26]. In the Shell investigation, the rotating flask test (RFT) has been used to simulate the short term ageing and long term ageing. For the long term ageing, an adaptation of the RFT called Long term ageing in the RFT (LTRFT) was performed.

Here, the 6% Altraflex-2006 modified C-Fix binder and mastic have been submitted to a long term ageing in the weatherometer for 1000 hours. Comparison with related bitumen is not possible, but modified C-Fix has showed considerable ageing resulting in a higher softening point and a lower penetration values. The mastic has shown, proportionally to the binder, lower ageing when comparing the complex shear modulus and phase angle master curves.

The 6% Altraflex-2006 modified aged C-Fix binder has shown to age in the direction of the pure binder when the master curves are compared. Complex shear modulus increased and reached those values of pure C-Fix, which could give an advantage on modifying C-Fix with Altraflex-2006: extra years on the life span of the pavement before ravelling due to oxidation of the binder (ageing) would start to occur.

# 5. Conclusions and recommendations

The microscopy tests done in the initial phase of the research, gave a good insight of the dispersion of the Altraflex-2006 in the binder. For 6% modified C-Fix, the distribution of the polymer can be classified as a continuous binder phase. The phase is homogeneous and only small polymer particles with a roundish shape can be visualized. At 12% Altraflex-2006 however medium size polymer particles can be recognized. The experience in mixing Altraflex-2006 and C-Fix also helped in defining the optimal polymer content, which appeared to be 6%.

The penetration increased with addition of Altraflex-2006 to C-Fix. The Softening Point also increased, which did not bring the expected lower mixing temperature. The increase in the calculated Penetration Index indicates that adding the modifier could improve the already good permanent deformation resistance of the binder.

The DSR master curves showed little change in the properties of the modified binder, although in general a decrease of the complex shear modulus and the phase angle was observed for the modified binder. Especially the lower stiffness at low temperatures is an important result because it indicates less brittle behaviour.

DMA tests have shown a decrease in the transition temperature when modifying the binder with Altraflex-2006. The addition of filler has decreased the transition temperature even more. The mastic with the Wigro 60K filler (filler/binder ratio 1) showed lowest transition temperature.

Direct Tensile Tests have shown higher toughness for the 6% modified binder at all temperatures investigated compared to the pure C-Fix. Duras Filler 15 has higher toughness at high temperature (10°C) for both filler/binder ratios tested. Wigro 60K mastics have a higher toughness at low temperature (-10°C) also for both filler/binder ratio studied. Wigro 60K could make the mastic stiffer, but could decrease its ductility and consequently its toughness at higher temperatures when calcium hydroxide is more active. Duras filler 15 seems to give more ductility to the mastic compared to Wigro 60K. For this reason Duras 15 was the filler chosen for the relaxation tests.

Addition of Altraflex-2006 has increased the end relaxation at low temperatures when comparing the modified and the pure binder, although when compared with B70/100, C-Fix presents still considerably lower relaxation ranges. The filler Duras 15 seems to give more relaxation at low temperature (-10°C) when compared with Wigro 60K. The extra ability of relaxing traffic and thermal loads could possibly give the modified C-Fix and the mastic (produced with Duras 15) less residual stresses resulting in possible less cracking formation.

Despite better results in the relaxation test for the 6% Altraflex-2006 modified C-Fix, the same was not present in the stone columns assessment. In this test, earlier higher phase angle and dissipated energy was measured and calculated, giving the 6% modified C-Fix shorter failure time.

In the ageing protocol used, the comparison with aged 6% Altraflex-2006 modified C-Fix with the unaged pure C-Fix could possibly give an advantage on using this modifier. The 6% Altraflex-2006 modified aged C-Fix binder has shown to have properties close to those of the pure unaged binder.

### 5.1 – General conclusions

The results of the tests done in the modified binder showed some improvements in the penetration values, extra ductility (higher tensile strain at break), higher toughness for all the temperatures tested, lower complex shear modulus and extra relaxation. In general, the results could point to an improvement in the C-Fix binder properties regarding brittleness.

Less cracking sensitivity could possibly be expected due to the addition of Altraflex-2006. Altraflex-2006 modified C-Fix would possibly be more suitable for PAC mixture than the pure C-Fix, although still high transition temperature is present, possibly leading to an earlier brittle behaviour when compared with bitumen.

When the mastic is concerned, the Duras filler 15 seems to give an average better result in relaxation, although higher transition temperature was found when compared with the mastic produced with Wigro 60K. The toughness performance of the Duras 15 mastic was relatively comparable with Wigro 60K, with even better results at higher temperatures.

Regarding the ageing, even if C-Fix has better ageing properties than B70/100, a considerable ageing was measured. Comparison of the aged 6% Altraflex-2006 modified C-Fix binder is made with the pure unaged C-Fix master curves could indicate some advantage on using Altraflex-2006 modified C-Fix.

All in all the results are promising but not enough yet to support the indication of C-Fix as a binder for PAC mixture. The transition temperature is still reasonably high. The reduction of the phase angle in the 6 and 12% modified binder does not reach higher frequencies in the DSR master curves that could possibly mean a less viscous behaviour at lower temperatures. Due to the low relaxation, also a residual stress is expected to develop in the mastic, even if this point has improved with the use of Duras 15 compared with Wigro 60K. The modifications made in this research are not yet decisive in indicating C-Fix for porous asphalt.

### 5.2 - Recommendations

If the Altraflex-2006 grains could be processed in a different way (smaller size, or different shape in which there would be more specific surface available for faster heating and consequently melting), higher amounts than 6% could be investigated. In this research, 12% of Altraflex-2006 was also used in the first phase of the research, but some difficulties were encountered in the laboratory to mix this amount. This was, among other issues, the reason it was decided to use a lower amount (6%) of Altraflex-2006 in the second phase of the research. The long time needed to melt the polymer particles (possibly due to large polymer grains), the high melting temperature of the polymer itself, the high temperature sensitivity of C-Fix binder (the smallest drop in temperature results in a big drop in the fluidity of the binder) hindering the mixing process, those are some of the biggest issues faced in the laboratory during the experiments phase in this research.

Also, after mixing the C-Fix binder with Altraflex-2006, one should be aware that this solution is not stable. This means that after storage at room temperature or lower, heating will be necessary to melt samples once again to prepare specimens for laboratory tests. This melting process should be extremely short (from 5 to 10 minutes only), with a good mixing before samples would be applied in the specimens' preparations, specially when dealing with Altraflex-2006 modified C-Fix only (without filler). Problems were quite recurrent in finding inconsistent results of some tests due to polymer/binder segregation caused by long oven heating period of the samples (around 1 hour). As mentioned before in this research, this is not a real problem in the asphalt plant as Altraflex-2006 is meant to be added in the pug-mill, but extra care should be taken in dealing with these materials in the laboratory to prevent inconsistent results.

If in fact, as suggested in the previous paragraphs, the Altraflex-2006 grains could be processed in a different way, different percentages could be tried such as 6, 9 and 12% by weight of the C-Fix binder in the tests performed in the second phase of this research. As 6% seemed to be the higher amount generally in use (in the literature research) in polymer bitumen modification, it would be not wise to go much beyond 12% of Altraflex-2006 (as it is made of around 40% SBS polymer). Also because of the high asphaltenes content of C-Fix, higher amounts of polymer could be more difficult or even unfeasible to process.

Tests comparing the results of Altraflex-2006 with other types of polymers shall be useful. Examples of such polymers are natural rubber or crumb tyre rubber which are expected also to improve fatigue cracking formation (it could improve the C-Fix binder brittle behaviour). Also comparison of Altraflex-2006 with pure SBS polymer could give the real contribution of Altraflex-2006 in the modification. Again, one should be aware of the high asphaltenes content of C-Fix, which could result in low solvency to polymers.

Tests with the modified C-Fix binder in the PAC aggregate mixture would also be extremely useful assessing the possible improvements achieved. Tests such Monotonic Indirect Tensile Test (MITT) could be performed to give an insight of the tensile strength of the mixture, or Cyclic Indirect Tensile Test (CITT) for an insight on its resilient modulus. Also fatigue tests performed on mixtures prepared with the modified C-Fix (four point bending beam test, for instance) could give a good information on the fatigue behaviour (main concern in using C-Fix due to its brittleness) of the C-Fix asphalt concrete. In this research, for time constrictions, comparison with a normal pen grade bitumen (B70-100) was not possible. These extra experiments with bitumen may increase the time needed for the research, but would also make it more complete, allowing comparison between them (C-Fix and bitumen). Comparisons with the results obtained by Khedoe [3] and Woldekidan [24] in some of these tests results are possible.

These tests with the PAC mixture made with Altraflex-2006 modified C-Fix could give a more complete picture, with possible comparison of the results already available of the pure and modified C-Fix binder only. For instance, the fatigue behaviour of the modified C-Fix binder showed to be worse than the pure C-Fix binder in the stone columns adhesion zone fatigue test. With the four points bending beam tests, this characteristic could be further investigated to check whether these results are valid or not. Same approach is valid for Relaxation Tests, or Direct Tensile Tests, for instance.

The ageing of C-Fix in-situ should be determined, which would give a great contribution to all the laboratory results that have been done in C-Fix ageing tests. Real pavement sections actually made on C-Fix should be catalogued in order to be checked later in time. These kinds of data would support the investigations made in the laboratory, as it is available with bitumen asphalt concrete.

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