Characterisation of oxidative ageing in asphalt concrete using a non-collinear ultrasonic wave mixing approach

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A study to assess oxidative ageing of asphalt concrete using non-collinear wave mixing of two dilatational waves is presented. Criteria were used to assure that the detected scattered wave originated via wave interaction in the asphalt concrete and not from non-linearities in the testing equipment. These criteria included the frequency and propagating direction of the resultant scattered wave and the time-of-flight separation between the two primary waves and the resulting scattered wave. It was observed that asphalt concrete exhibits non-linear behaviour. It was also observed that the non-linear response decreases with increased ageing until approximately 24 h of ageing, after which the non-linear response exponentially increases. This observation is consistent with previous studies, including the acoustic emission response to thermal loading, and with changes in the dynamic modulus and fracture energy with increasing ageing.

Introduction

Although possessing remarkable toughness in its original state, asphalt concrete (AC) becomes brittle and prone to damage with time, in the form of costly pavement cracking. The time required to reach an unacceptable level of embrittlement depends upon a number of factors and varies widely from pavement to pavement, even within a given region and mixture type. Given the annual costs associated with repair of pavement damage caused by mechanical and thermal loads, considerable interest exists in testing methods to estimate damage in AC mixtures. Current methods to evaluate the existing conditions of AC pavement surfaces for sustainability-based pavement asset management are based upon the binder’s rheological properties and are time consuming, costly and, by themselves, may cause additional damage.

Oxidative ageing is a key contributor to the deterioration of asphalt concrete pavements[4-7]. Exposure to environmental conditions causes gradual oxidative ageing of the asphalt concrete, where the highest aged material is located at the surface, while the material at the bottom of the pavement is significantly less aged. Over time, increasing ageing at the surface leads to a pavement with graded material properties through its thickness, where the material near the surface has warmer embrittlement temperatures and higher stiffness compared to the bottom of the pavement. Increasing ageing also results in significant loss of adhesion between the binder, aggregates and fines, which contributes to an increase in the micro-flaw population in the mastic and at the interfaces between the mastic and the aggregates[8]. As the pavement is subjected to thermal loads (for example cold climates and temperature cycling) and mechanical loads (for example traffic loads), microcracks develop and coalesce to form larger cracks. Repeated loading and exposure to environmental conditions eventually leads to significant deterioration of asphalt concrete pavements. Although surveys are conducted to monitor the condition of pavements and to determine when preventative or corrective maintenance is necessary, accurate assessment of the amount of pavement deterioration has remained a challenge. Clearly, there is a need for a rapid in-situ non-destructive evaluation technique capable of assessing the level of oxidative ageing in the top layers of asphalt concrete pavements.

Conventional acoustic and ultrasonic structural health monitoring methods, including traditional guided wave studies, are based upon principles that are valid under the assumption of linear elasticity. These include the effects of reflection, scattering, transmission and loss of energy by material absorption and by leakage to adjacent systems, as in the case of guided wave systems. Under these assumptions, the presence of defects leads to phase or amplitude variation of the ultrasonic wave (signal), keeping the frequency of the receiving wave the same as the frequency of the wave emitted by the sending transducer. In non-linear ultrasonics, the frequency of the received wave differs from the frequency of the emitted wave (ie from the frequency of the wave generated by the sending transducer), implying that a non-linear transformation of ultrasonic wave energy by the media has occurred, as in harmonic generation[9-7].

In linear elastic wave propagation, superposition holds: two or more waves can cross paths and their resultant (in the region where they cross paths) is the addition (ie superposition) of those waves. However, superposition does not hold when the media is non-linear. Because of the presence of higher-order terms in the non-linear equations of motion, when two waves cross paths, ie intersect, a third wave, ie a scattered wave, may arise. However, in order for a strong scattered wave to occur, resonance and polarization conditions must be met[10-20].

Using equilibrium conditions and virtual work, the third-order non-linear equations of motion were developed by Murnaghan[11] in 1951. He proposed a set of third-order elastic constants (TOECs), which are commonly referred to as $l$, $m$ and $n$. In 1954, Landau and Lifshitz[12] suggested using the relationship between the stress tensor and the elastic energy function as the basis for deriving the non-linear equations of motion and proposed a different set of TOECs, referred to as $A$, $B$ and $C$; the derivation was carried out by Goldberg[13] in 1961. The two sets of TOECs are linear combinations of each other. In 1963, Jones and Kobett[14] solved the third-order non-linear elastic equations of motion and developed criteria for the occurrence of a scattered wave. Experimentally, elastic non-linear behaviour has been observed for a variety of materials using the non-collinear wave mixing approach for a broad range of ultrasonic
Non-collinear wave mixing has proved useful in a variety of practical applications, which include the determination of higher-order elastic constants\textsuperscript{11-14}, and in the inspection and detection of material degradation, such as plastic deformation and fatigue damage accumulation in metals\textsuperscript{19} or ageing in polymers\textsuperscript{17}. Johnson and Shankland\textsuperscript{12,13} also reported scattered waves as a result of the non-linear interaction of two intersecting waves of different frequencies in crystalline rock and developed criteria to assure that the scattered wave is due to non-linearities in the material response and not caused by non-linearities inherent in the instrumentation used.

Distributed damage, such as damage induced by fatigue, typically consists of phenomena initially affecting the material at many locations in the form of local variations in the material microstructure. In metals such as aluminium, these local variations in microstructure consist of fatigue-induced entanglement of dislocation substructures, such as the sidebands. These sidebands, which accumulate at the grain boundaries, produce strain localisation, which eventually leads to microcrack initiation with an increased number of fatigue cycles. While these local changes in microstructure do not change macroscopic elastic moduli or produce variations in linear acoustic parameters, such as acoustic velocity and attenuation, the stress field in the dislocation(s) creates a local non-linear stress-strain response in the localised volume of the dislocation(s). These local variations in the microstructure on the scale of single grains develop before the initiation of a visible crack. This degradation of the material significantly reduces the material's resistance to crack growth when microcracking coalescence occurs at a later time. The ultimate strength of most structural materials is limited by the presence of these microscopic microstructural variations, ie defects, which can be produced by mechanical or thermal loading and subsequently serve as nuclei of the fracture process. Because these nuclei (local variations) are significantly shorter than the acoustic wavelength at the frequencies normally used in ultrasonic non-destructive evaluation, linear acoustic parameters (ie attenuation, velocities, etc) are usually not sensitive enough to detect this kind of microscopic degradation of the material structural integrity.

In addition to the non-linear behaviour of the individual constituents of asphalt concrete, ie binder, crushed stone and fines, the presence of micro-flaws in the composite structure causes a non-linear distortion in the propagating wave. As the asphalt concrete is subjected to oxidative ageing, two counter-acting effects take place within the mastic (binder + fine material). The stiffness of the mastic increases with ageing, while the adhesive properties of the mastic decrease. Until a critical amount of ageing, the positive effects of the increased stiffness overcome the negative effects of the reduction in adhesion. Beyond this critical point, the adhesion loss becomes the dominating effect. The loss in adhesion leads to weaker bonds between the mastic and the aggregates and between the fines in the binder, which leads to a weaker internal structure of the composite. This degradation of the material significantly reduces the material's resistance to crack growth later when microcracking coalescence occurs. The ultimate strength is limited by the presence of these micro-flaw populations, which will then serve as nuclei of the fracture process.

As in metals, the presence of these micro-flaw populations causes a non-linear distortion in the propagating wave. Therefore, as the oxidative ageing of the asphalt increases, the material displays increasingly non-linear behaviour. Air voids, which are also present in the material based on standard design mixture guidelines, may also have a similar effect on the propagating wave.

Using linear ultrasonics, McGovern et al\textsuperscript{23-25} studied the effects of oxidative ageing of asphalt concrete mixtures upon the acoustic emission response during thermal cooling and upon both ultrasonic longitudinal and shear velocities and corresponding attenuations. In this study, the non-linear behaviour of asphalt concrete as a function of laboratory-induced oxidative ageing is investigated by a non-collinear wave mixing approach, using two dilatational waves similar to the methodology used by Johnson and Shankland\textsuperscript{12,13}. For the purpose of comparison, the asphalt concrete mixtures and the different levels of oxidative ageing are the same in both studies\textsuperscript{24,25}.

### Asphalt concrete and its non-linear mesoscopic nature

Asphalt concrete belongs to a class of materials that have non-linear mesoscopic elasticity (NME). For example, rocks, soils and powdered aluminium have non-linear mesoscopic elasticity (NME)\textsuperscript{24,25}. This class of materials typically has a bricks-and-mortar character. In these materials, the bricks (quartz grains in the case of rocks and crystallites, such as quartz, feldspar and clay particles, in the case of soils) interface with one another across a distinctive, elastic phase, which behaves as the mortar (a system of asperities in the case of rocks and a system of fluid layers and fillets in the case of wet soil). For these materials, the typical elastic modulus is an order of magnitude smaller than the corresponding modulus of the bricks. Having ten times as much displacement and about 10% of the volume means that the mortar, while only a minor phase in terms of volume concentration, is responsible for much of the compliance in the composite and may carry strains as high as two orders of magnitude greater than the strains in the bricks or rigid particle phase.

This behaviour has also been observed in asphalt concrete mixtures, where the bricks are the aggregates (ie crushed stone) and the mortar is the asphalt binder, which is approximately 5% by volume. Asphalt binders exhibit both linear and non-linear viscoelastic behaviour and the level of non-linearity increases with stress or strain levels\textsuperscript{24,25}. Masad et al\textsuperscript{30} found the average binder strain range to be 9 to 12 times the mixture strain, and that the binder maximum strain can be as high as 90 times that of the mixture strain; this induces non-linear behaviour in the binder and consequently in the mixture as well. Masad et al\textsuperscript{35} also studied the viscoelastic response of un-aged and aged binders. The non-linear response of binders and mixtures was also studied by Delgadillo\textsuperscript{35}, and a comprehensive investigation on low-temperature cracking in asphalt pavements was carried out by Marasteanu et al\textsuperscript{36}.

Clearly, in addition to thermal cracking induced by the thermal stresses during cooling, the high level of stresses and strains in the binder will increase the non-linear response of the asphalt concrete mixture.

### Specimen preparation and linear characterisation

Six gyratory compacted asphalt concrete specimens were prepared using PG 64-22 binder with a target asphalt content of 5.9% by weight of the total mixture. The aggregate structure had a nominal maximum aggregate size (NMAS) of 9.5 mm and it consisted of aggregates from four different stockpiles: 65% of coarse aggregate (CM16), 23% of manufactured sand (FM20), 10.5% of manufactured sand (FM02) and 1.5% of mineral filler (MF). This mixture design followed Superpave guidelines.

The asphalt concrete mixtures were mixed using a standard bucket mixing procedure at a temperature of 155°C. The mixtures were oxidatively aged by placing the uncompacted mixtures in an oven at 135°C. Each of the six specimens was subjected to a different amount of ageing, namely 0, 12, 24, 28, 32 and 36 h. Ageing beyond 36 h was not carried out because the rough, cratered surfaces of specimens aged past 36 h caused coupling difficulties with the sensors, preventing ultrasonic testing. To ensure uniformity of the ageing process (ie uniform exposure to oxygen), the mixtures were hand-stirred every 12 h.
Cylindrical specimens (150 mm in height and with 150 mm diameters) were created by compacting the aged mixtures with a servo-controlled gyratory compactor (IPC Servopac) at a temperature of 135°C. Each of the cylinders was cut to obtain a 5 cm-thick cylinder, see Figure 1. Appropriate angles were then cut into the cylinders so that the transducers could be mounted directly onto the flat faces of the specimen. Figure 1 shows the geometry and dimensions of the extracted test samples from each of the cylindrical compacted gyratory specimens.

Figure 1. Geometry of test specimens: (a) gyratory compacted specimen; and (b) extracted test specimens with different levels of oven ageing at 135°C. The angles at which these specimens were cut were such that the sending and receiving transducers were positioned at the appropriate angles to satisfy resonance and polarisation conditions, see Figure 3.

In a previous study\cite{24,25}, ultrasonic phase velocity and attenuation measurements for dilatational and shear waves were taken on six specimens constructed with the same mixture design and oxidatively aged using the same process as described above (see Figure 2). These ultrasonic results are presented as a function of frequency and ageing amount. It can be seen that the velocity (over all frequencies) increases with increased ageing until 24 h of ageing; after 24 h, the velocity decreases with increased ageing. Similarly, the attenuation (over all frequencies) decreases with increased ageing until 24 h of ageing, after which it increases with increased ageing. Please refer to McGovern et al\cite{24,25} for a more detailed discussion on these results.

Using a non-collinear wave mixing approach

Jones and Kobett\cite{11} considered the scattered wave that may result from the interaction of two primary monochromatic waves. The two waves travelling in directions $k_1$ and $k_2$, with frequencies $f_1$ and $f_2$, respectively, may be dilatational and/or shear (polarised either in or out of the $k_1$-$k_2$ plane) and may interact to produce a scattered dilatational or shear wave with a sum ($f_1 + f_2$) or difference ($f_1 - f_2$) frequency, which propagates in the $k_0$ direction.

The polarisation of the scattered shear wave depends on the type and polarisation of the two primary waves. In addition to resonance conditions, polarisation conditions must also be met in order for interaction to be possible for a given set of primary waves. As a result, there are only a finite set of possible interaction cases; out of fifty-four potential non-linear scattered waves (resulting from nine possible interaction cases of different bulk primary waves), only nine potential non-linear scattered waves satisfy both the resonance and polarisation conditions. In this study, only the non-collinear interaction of two dilatational waves is considered, where two dilatational waves ($k_1, f_1$ and $k_2, f_2$) are forced to interact to produce a scattered shear wave ($k_0 = k_1 - k_2, f_0 = f_1 - f_2$). For resonance and polarisation conditions to be met, the following equations need to be satisfied:

$$\cos(\phi) = \left( \frac{c_1}{c_0} \right)^2 \left[ 1 - \frac{f_1}{f_2} \left( \frac{c_1}{c_2} - 1 \right) \left( \frac{f_2}{f_1} + 1 \right) \right] \quad (1)$$

$$\tan(\gamma) = -\frac{f_2 \sin(\phi)}{f_1 - f_2 \cos(\phi)} \quad (2)$$

In Equations (1) and (2), $f_1$ and $f_2$ denote the frequencies of the two primary dilatational waves travelling in directions $k_1$ and $k_2$, respectively, $c_1$ and $c_2$ denote the dilatational and shear velocities, respectively, $\phi$ is the angle between $k_1$ and $k_2$, and $\gamma$ is the angle between $k_0$ and $k_1$. For the case described by the above two equations, the non-linear scattered wave is polarised in the $k_1$-$k_2$ plane. To assure that the scattered wave originates from the non-linear interaction in the material and not from non-linearities inherent to the testing instrumentation, Johnson and Shankland\cite{10,12} proposed three criteria: (1) frequency – the frequency of the observed scattered wave must match the frequency predicted by theory; (2) amplitude – the amplitude of the scattered wave must be proportional to the product of the amplitudes of the primary waves; and (3) directionality – the propagating direction of the scattered wave must match the direction predicted by theory (Equations (1) and (2)). In this study, the experimentally-observed time-of-arrival of the scattered wave is also considered, which should closely match the time-of-arrival predicted using ray-path analysis.

Experimental set-up

Prior to cutting the asphalt concrete specimens, the appropriate angles must be chosen such that wave interaction occurs and the resulting scattered wave is received. The reader is referred to Equations (1) and (2). The frequency ratio $f_2/f_1$, interaction angle $\phi$ and scattered wave angle $\gamma$ are all interdependent quantities. In other words, once one parameter is chosen (for example $\phi$), the other two (for example $\gamma$ and $f_2/f_1$) are set. Typically, this task is relatively simple: for a known set of ultrasonic material properties ($c$ dilatational and shear velocities/attenuations), one parameter can be chosen, in consequence setting the other two. Then, the specimen dimensions can be selected and appropriate angles cut into the specimen.

In this study, the goal is to use the non-collinear wave mixing approach to estimate the ageing level of asphalt concrete. It follows that the testing set-up should be the same for all specimens. The ultrasonic velocities vary with the level of ageing, which complicates the matter of choosing the appropriate testing.
set-up (ie specimen dimensions, interaction/scattered angles and frequencies) that will work across all aged specimens. To assess the specimen ageing, a testing set-up will be chosen based on the virgin specimen parameters. Determining a final testing set-up is an iterative process, where the goal is to find one testing set-up that meets the conditions, which will now be outlined.

**Interaction angle, \( \phi \)**

The interaction angle will be chosen such that all six specimens (0 to 36 h of ageing) will interact at the same angle, resulting in different \( f_1 / f_2 \) and \( \gamma \) for specimens having different ageing levels. The interaction angle will be chosen based solely on its effects on the scattered wave angle and frequency ratio.

**Primary and scattered wave frequencies**

The primary wave frequencies should be chosen such that the waves propagate with minimal distortion (due to scattering and attenuation) and generate a non-linear scattered wave with the same characteristics. To achieve this, the wavelength of the propagating wave should be larger than the aggregate size. The nominal maximum aggregate size for this mix is 9.5 mm. The upper frequency bound for shear waves is 110 kHz, which corresponds to a shear wave wavelength (= 9.5 mm) through the 36 h-aged specimen. The dilatational waves have wavelengths greater than 9.5 mm for all ages for the measured frequency range (≤350 kHz). Furthermore, the two primary wave frequencies should be chosen such that their resulting velocities are similar, to be consistent with the theory presented in Equations (1) and (2).

Above 100 kHz, the dilatational wave velocities all tend to plateau (see Figure 2). Thus, the frequencies of the primary waves should be chosen to be greater than 100 kHz, because Equations (1) and (2) were derived with the assumption that \( k_1(f_1) \) and \( k_2(f_2) \) have the same velocity. Above 200 kHz, the attenuation increases dramatically. In fact, for specimens aged 36 h, a signal could not be detected through the thickness of 30 mm above 250/200 kHz for dilatational/shear waves. Therefore, the primary wave frequency should be restricted to 200 kHz or below. It is difficult to detect shear waves below 50 kHz with the transducers employed in the set-up; as a result, the scattered shear wave should have a frequency above 50 kHz.

Therefore, the primary waves should be chosen within a frequency range of 100 to 200 kHz, and the resulting scattered shear wave should be within a frequency range of 50 to 110 kHz. Also, the scattered wave frequency \( f_s \) should have a sufficient separation from the primary wave frequencies \( f_1 \) and \( f_2 \), so that they can be easily separated in the frequency domain. Of course, the choice of frequencies should also consider the effects it has on the interaction and scattered wave angles.

**Scattered wave angle, \( \gamma \)**

All specimens will be cut according to the conditions specified using the virgin parameters. The amount of deviation between the angle for which the specimens are cut and the actual scattered angles will affect how well the scattered wave is received. For this reason, a case should be found that minimises the difference in scattered wave angles between the virgin specimen and the other aged specimens, so that even if the receiving transducer is not oriented in the ideal location it can still receive the non-linear scattered wave.

**Specimen dimensions**

Asphalt concrete is highly attenuative (see Figure 2), which will greatly diminish the amplitudes of the primary and scattered waves as they propagate through the specimen. This attenuation loss should be minimised by minimising the distance through which the wave propagates to ensure that the scattered non-linear wave can be detected by the receiver. The propagation distances should be at least one wavelength long, such that the waves are stabilised by the time they interact. For simplicity, \( k_1 \) and \( k_2 \) can be chosen to have the same propagation distances.

**The non-linear wave generation parameter, \( \beta \)**

To characterise the non-linearities in the asphalt concrete with respect to ageing, a non-linear wave generation parameter is introduced. The amplitude of the scattered wave is proportional to the product of the primary wave amplitudes at the time of interaction \([14,15]\). The primary waves suffer attenuation as they propagate through the specimen before they interact, and the scattered wave is further attenuated as it travels to the receiver. Accounting for the attenuation, the received amplitude of the scattered wave can be described by the following expression (assuming perfect coupling conditions):

\[
A_{w,ie}^{(k)} = \beta_{ie} A_{w,ie}^{(1)} A_{w,ie}^{(2)} \exp \left[ -\left( \alpha^{(1)} + \alpha^{(2)} \right) D_{we} \right] \exp \left[ -\alpha^{(1)} D_{we} \right]
\]

where:

- \( \beta = \) conversion efficiency
- \( A_{w,ie}^{(k)} = \) Transmitted amplitude of \( k \) (volts)
- \( \alpha^{(k)} = \) Attenuation coefficient of \( k \) \((Np/m)\)
- \( D_{we} = \) Propagation distance of \( k \) (m)

The conversion efficiency parameter \( \beta \) is a dimensionless parameter, which accounts for the fraction of the interacting waves that is converted to the scattered wave. The propagation distances are approximated by the plane-wave assumption. The attenuation coefficients were measured empirically.

Normalising the amplitude by the attenuation, and denoting it by:

\[
A_{w,ie}^{(k)} = \frac{A_{w,ie}^{(k)}}{\exp \left[ -\left( \alpha^{(1)} + \alpha^{(2)} \right) D_{we} \right] \exp \left[ -\alpha^{(1)} D_{we} \right]}
\]

then:

\[
\beta_{ie} = \frac{A_{w,ie}^{(k)}}{A_{w,ie}^{(k)}}
\]

Assuming that variations in the coupling conditions between testing set-ups can be neglected, the transmitted amplitudes of the primary waves \( A_{w,ie}^{(1)} \) and \( A_{w,ie}^{(2)} \) will be the same for all tests performed. As a result, the conversion efficiency parameter for the aged mixture, \( \beta_{ie} \), can be normalised by the conversion efficiency parameter for the virgin, ie unaged, mixture, \( \beta_0 \), to characterise increasing levels of ageing:

\[
\beta_{ie} = \frac{\beta_{ie}}{\beta_0}
\]

By way of the above formulation, the non-linear wave generation parameter is independent of the attenuation, so that it represents the conversion efficiency of the energy transferred from the primary waves interacting to produce the scattered non-linear wave. Thus, this parameter is a good indicator of the material’s inherent non-linear behaviour.

**Experimental description**

Using the considerations outlined in the previous section, the specimens were cut so that two longitudinal transducers could be mounted on the faces of the specimens and generate two longitudinal primary waves \( k_1 \) and \( k_2 \) that interact at 31° to produce a non-linear scattered shear wave, which is received at 42° with respect to \( k_2 \). These angles were chosen based on virgin specimen properties and used for all six specimens. Table 1 contains the theoretical values
Table 1. Average dilatational and shear velocities (between 140-200 kHz), corresponding frequency ratio $f_3/f_1$, and scattered wave angle $\gamma$ for an interaction angle $\phi$ of 31°. For time-of-flight calculations, the shear velocity at the scattered wave frequency $f_3$ is also presented.

<table>
<thead>
<tr>
<th>Amount aged (h)</th>
<th>Dilatational velocity mean (140-200 kHz) $c_i$ (m/s)</th>
<th>Shear velocity mean (140-200 kHz) $c_s$ (m/s)</th>
<th>Velocity ratio $c_i/c_s$</th>
<th>Interaction angle $\phi$ (°)</th>
<th>Frequency ratio $f_3/f_1$</th>
<th>Angle of scattered wave $\gamma$ (°)</th>
<th>Scattered wave frequency $f_3$ (kHz)</th>
<th>Shear velocity at $f_3$ $c_s$ (m/s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>3559</td>
<td>1992</td>
<td>0.560</td>
<td>0.700</td>
<td>–42°</td>
<td>60</td>
<td>1384</td>
<td>31°</td>
</tr>
<tr>
<td>12</td>
<td>3785</td>
<td>2066</td>
<td>0.546</td>
<td>0.710</td>
<td>–43</td>
<td>58</td>
<td>1343</td>
<td></td>
</tr>
<tr>
<td>24</td>
<td>4005</td>
<td>2088</td>
<td>0.521</td>
<td>0.724</td>
<td>–44</td>
<td>55.2</td>
<td>1343</td>
<td></td>
</tr>
<tr>
<td>28</td>
<td>3285</td>
<td>1629</td>
<td>0.496</td>
<td>0.738</td>
<td>–46</td>
<td>52.4</td>
<td>1013</td>
<td></td>
</tr>
<tr>
<td>32</td>
<td>2780</td>
<td>1313</td>
<td>0.472</td>
<td>0.751</td>
<td>–47</td>
<td>49.8</td>
<td>904</td>
<td></td>
</tr>
<tr>
<td>36</td>
<td>2868</td>
<td>1273</td>
<td>0.444</td>
<td>0.764</td>
<td>–49</td>
<td>47.2</td>
<td>648</td>
<td></td>
</tr>
</tbody>
</table>

*Note: The specimens (for all ages) were all cut to the angles determined for the virgin specimen. Care was taken to find a case where the difference in the scattered wave angle was minimal (ie $\gamma_{\text{vir}} - \gamma_{\text{aged}} \approx 0$°) so that the receiving transducer could detect the scattered wave for all specimens. Keeping the angles constant causes the frequency ratio at which the non-linear wave interaction occurs to shift.

for all aged specimens. The velocities used to calculate the velocity ratio were computed using the mean velocities across 140-200 kHz (see Figures 1 and 3). This set-up avoids complications that arise with the implementation of wedges (for example refracted angle, generation of multiple refracted modes, etc). The specimen dimensions were cut such that $k_1$ and $k_2$ will propagate a distance of 5 cm and $k_3$ will propagate a distance of 4 cm. A plastic template was created using a 3D printer to ensure reproducibility of transducer placement between tests. The plastic template supported the asphalt concrete specimens using three screws to minimise the contact area between the test specimens and the supporting structure surface.

A pulser-receiver (Ritec RPR 4000) was used to generate and amplify a 15-cycle sinusoidal signal at $f_1 = 200$ kHz. This signal ($k_1$) was sent to a dilatational wave transducer (Panametrics V413, centre frequency 500 kHz). A function generator (Krohn-Hite Model 5920) was used to generate an 8-cycle sinusoidal wave, which swept from $f_3 = 110$ kHz to 180 kHz in 1 kHz increments. This signal ($k_3$) was amplified using a gated amplifier (Ritec GA-2500A) and sent to another dilatational wave transducer (Panametrics V413, centre frequency 500 kHz). The number of cycles in the tonebursts was chosen to ensure the intersection of the primary dilatational waves; (2) the inherent dispersion in the asphalt concrete (especially at high frequencies); and (3) the presence of the dominating, ie large amplitude, primary waves in the imperfect subtraction (a portion of the signal energy is lost in the conversion to the non-linear scattered wave; this leads to the signal resulting from the addition of the signals obtained when the dilatational transducers are operated individually being larger than the signal obtained when the two longitudinal transducers are operated simultaneously). To circumvent the low amplitude and maximise the ability to detect the scattered wave, the following steps were taken: (1) a high sample rate (50 MHz) was used in order to avoid trigger jitter, as suggested by Johnson and Shankland[12,13]; and (2) to mitigate the scattering effects, an average of 500 waveforms were collected. For each ageing level, 10 independent measurements were taken, which required removal of the three sensors, removal of the used couplant and subsequent application of new couplant and repositioning of the three sensors.

Figure 3. Schematic diagram of the ultrasonic data collection system illustrating the angle of interaction of the two longitudinal waves and the location of the shear transducer to receive the generated scattered shear wave. The blue and red regions denote the areas of signals $k_1$ and $k_3$, respectively, caused by the beam spread. The overlap region is the volume of interaction. Note that the beam spread from $k_3$ is slightly higher than from $k_1$ due to the difference in frequencies.

Experimental results

To ensure that the received wave is a scattered wave resulting from non-linear interaction between the primary waves within the specimen, the following selection criteria (proposed by Johnson and Shankland[12,13]) were used, ie amplitude, directionality, frequency, and time-of-arrival criteria.

Amplitude criterion

The amplitude of the non-linear signal must be proportional to the product of the amplitude of the primary dilatational waves. An experiment was conducted where it was observed that, as the voltage of the primary waves was increased, the amplitude of the non-linear signal also increased in a manner proportional to the amplitudes of the primary waves.

Directionality criterion

The propagating direction of the scattered wave must match the propagating direction predicted by theory. The geometry of the samples was selected using the virgin parameters shown in Table 1, which were calculated using Equations (1) and (2). As a result,
since the scattered non-linear wave was received, the directionality criterion was satisfied. Care was taken to ensure that even though the scattered wave angle $\gamma$ for the aged specimens deviated from the virgin scattered wave angle, it still hit the transducer face.

**Frequency criterion**

The frequency of the non-linear scattered wave must closely match the frequency predicted by theory ($ie f_2 = f_1 - f_3$ at the appropriate value of $f_2/f_1$). The experimental set-up is based on virgin specimen properties; therefore, for each aged specimen, the frequency ratio at which interaction takes place will vary. To verify that the non-linear interaction took place at the predicted frequency ratio, the amplitude of the non-linear scattered wave was monitored as $f_2$ was swept and $f_1$ was held constant. The maximum amplitude of the non-linear scattered wave should occur when $f_2$ reaches the frequency where $f_2/f_1$ matches the ratio predicted by the theory (Figure 4).

The amplitude of the scattered wave may be measured by taking the fast Fourier transform (FFT) of the difference signal and recording the amplitude at the appropriate frequency ($f_2$) as $f_2$ was swept. Because the frequency of the non-linear scattered wave, $ie f_2$, changes with the amount of ageing of the specimen, instead of monitoring the $f_2$ amplitude via the FFT, a 4th-order Butterworth bandpass filter (30-90 kHz) was used. These filter limits ensured that the primary waves were filtered out as well as any other very low frequencies. Recording the difference wave amplitude over a finite band of frequencies (instead of a discrete point as it is done with the FFT method) ensured that even when $f_2$ changed (with ageing), the $k_2$ amplitude could still be monitored. It was verified that filtering with such a broadband filter and taking the FFT yielded nearly the same results for the amplitudes. For the different specimens, the non-linear frequencies were theoretically predicted to be: $(f_2/f_1)_0 = 0.68$; $(f_2/f_1)_1 = 0.55$ to 0.90; $(f_2/f_1)_2 = 0.70$; $(f_2/f_1)_3 = 0.764$.$f_3$ and $(f_2/f_1)_4 = 0.71$ and $(f_2/f_1)_5 = 0.68$.

Data collected from the specimen aged 12 h is shown in Figure 4 as a representative example of the measured amplitude of the filtered difference signal ($ie$ the non-linear scattered wave amplitude). As $f_2$ is swept, the non-linear wave amplitude reaches a maximum at $f_2/f_1 = 0.68$ ($f_2 = 136$ kHz). This is close ($\pm 4\%$ difference) to the predicted value of $f_2/f_1 = 0.71$ ($f_2 = 142$ kHz). Theoretically, the amplitude of the received shear wave should have vanished when $f_2$ deviates from 136 kHz. The width of the amplitude curve shown in Figure 4 is probably due to wave scattering induced by the presence of the stochastic nature of the aggregate structure, which leads to different propagation paths of the wave energy and to the large interaction volume. The relatively large interaction volume and the stochastic nature of the asphalt concrete structure, $ie$ aggregate-binder matrix, may have the effect of smearing the theoretical frequency ratio over a range of values centred about the theoretically-predicted value.$^{[21,22]}$. The theory assumes the test sample material to be isotropic and homogeneous and the two interacting longitudinal waves to be monochromatic.

The frequency ratios at which the maximum non-linear wave amplitudes occurred were recorded from all the measured data and plotted as a function of specimen ageing, as shown in Figure 5. The theoretical prediction was calculated using the mean of the measured velocities across 140 to 200 kHz and is denoted by the dashed line. The measured amplitudes of the scattered waves, corresponding to specimens with different ageing levels, reach a maximum close to the theoretical predictions (Table 2). The largest deviation from the theoretical prediction is for the case with 24 h of ageing (8.65% difference). Even though the 36 h had a low percent difference (1.18%), it had the highest standard deviation amongst the measurements. Although there is a statistically significant observable trend for the experimentally-observed frequency ratio as a function of ageing, it is not very sensitive, as the errors are high relative to the change with ageing.

**Table 2. Experimentally-observed and theoretically-predicted frequency ratios corresponding to the maximum amplitude of the scattered wave**

![Figure 4. Experimentally-obtained amplitude of scattered shear wave, $ie$ difference signal ($f_2 = f_1 - f_3$) as $f_2$ is swept from 110 kHz to 180 kHz ($f_2/f_1 = 0.55$ to 0.90), while $f_1$ is held constant at 200 kHz. This analysis was performed for all specimens to obtain the data shown in Figure 5. The plot shown above is from the specimen oven-aged for 12 h and shown as a representative case. The dashed blue line represents the experimentally-obtained maximum and the dashed red line represents the theoretically predicted maximum, which was obtained using the experimentally-determined velocity data.](image)

![Figure 5. Experimentally-observed frequency ratio at which the maximum non-linear scattered wave amplitude occurs. The theoretical amplitude was predicted (see Equations (1) and (2)) using the experimentally-obtained dilatational and shear velocities.](image)
Time-of-arrival

Finally, the results were also viewed in the time domain to see whether a time separation existed. If the non-linear signal originated from the equipment, for example the function generator, amplifiers, transducers, etc, a separation in time between the primary waves and the difference signal should not exist. In other words, the non-linear signal inherent to instrumentation should arrive at the same time as the primary waves. However, when the difference signal is due to non-linear wave mixing within the asphalt concrete, a time separation should exist. The theoretical time of arrival of the difference signal, ie scattered wave, can be calculated assuming mean velocities, see Table 1, and a straight ray-path analysis.

As a representative example, Figure 6 shows a set of time domain records obtained from the asphalt concrete specimen aged for 36 h: (a) when both sending transducers are operating simultaneously; (b) when the sending transducers are operated individually, one at a time; and (c) the resulting scattered wave, ie the difference signal. The theoretical arrival time for the difference signal is found to be 81.5 μs, which matches closely (~8.8% difference) with the observed arrival time of 74.9 μs. The stochastic nature of the asphalt concrete also causes disparities in the independent measurements, as even a slight variation in the placement of the transducers can alter the paths of the travelling waves.

Figure 6. Time domain records required to obtain the non-linear scattered shear wave: (a) time record obtained when both sending transducers were operated simultaneously; (b) time record obtained when sending transducers were operated one at a time and the received waveforms added; and (c) non-linear scattered wave, ie difference signal, obtained from subtracting the signals obtained from operating the sending transducers individually from the signal obtained when operating the two sending transducers simultaneously. The theoretically-predicted time of arrival for the difference signal matches closely (~8.8% difference) with the experimentally-observed time of arrival. The records are all normalised by the maximum amplitude of the record in (b). The difference signal was scaled up 25 times

The non-linear wave generation parameter, β

The non-linear scattered wave generation parameter, β, is an indicator of the conversion efficiency of the energy transferred from the primary waves, due to the non-collinear interaction, to produce the scattered non-linear wave. Figure 7 shows β normalised with β₀ (corresponding to the virgin mixture). The normalised parameter decreases with an increased amount of ageing until 24 h, when it begins to exponentially increase with increasing ageing. Similar trends have been observed in other experiments, such as fracture energy obtained via DC(T) tests, acoustic emission responses and dynamic modulus estimates via ultrasonic tests, in which the behaviour of asphalt concrete as a function of ageing has been studied.

Figure 7. Normalised non-linear parameter, β, versus different levels of oven-ageing. The parameter β is normalised with the parameter, β₀, which corresponds to the virgin, ie unaged, mixture. For each ageing level, each of the ten independent measurements required removal of the three sensors, removal of the used couplant, and subsequent application of new couplant, and reposition of the three sensors

Due to the nature of the experimental set-up (parameters were based upon the material properties of the virgin specimen), the scattered wave will not always hit the received transducer in the best possible manner (ie normal incidence at the transducer centre), and the experimentally-measured beta parameter will be affected by this deviation. However, since the scattered wave hits the receiving transducer increasingly off-centre and at an angle as ageing increases, the measured values of β may increasingly be an underestimate of its true value as ageing increases. Figure 2 shows that the attenuation increases drastically with ageing. Should attenuation not be taken into account, the observable trend of the non-linear wave amplitude with respect to ageing shown in Figure 7 will be less pronounced. However, accounting for attenuation reveals that the asphalt concrete exhibits increasingly strong non-linear behaviour with ageing.

Conclusions

Asphalt concrete test samples were manufactured with various amounts of laboratory-induced oven ageing and assessed using a non-linear ultrasonic approach involving the mixing of two non-collinear dilatational waves. A non-linear scattered wave was observed as a result of the interaction of two intersecting waves in the asphalt concrete specimens, even though the material is highly attenuative and dispersive. Criteria were used to confirm that the detected scattered wave originated as a result of wave interaction in the asphalt concrete and not from non-linearities in the testing instrumentation. A non-linear wave generation parameter was defined to describe the conversion efficiency of energy from the primary waves into the non-linear scattered wave. By monitoring the non-linear wave generation parameter, it was observed that the non-linearities exhibited by the asphalt concrete decrease until about 24 h of ageing, after which the non-linear response increases exponentially with increasing ageing. The observed non-linear response has the potential of being used for the assessment of oxidative ageing of asphalt concrete, including durability and damage accumulation due to thermal and mechanical loading and other environmental conditions.

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