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The Influence of Mineral Aggregates on Bitumen Ageing

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Abstract: A typical basic aggregate (limestone) and a typical acidic aggregate (granite) were mixed with a 100/150 penetration grade bitumen and its 'short-term' aged equivalent to produce four different 28mm dense bitumen macadam (DBM) asphalt mixtures. Subsets of the four asphalt mixtures were also subjected to the SHRP short- and long-term ageing protocols to further age the different mixtures. The ageing properties of these asphalt mixtures, together with their recovered binders, were tested at the different ageing stages using stiffness modulus (ITSM) and complex modulus (DSR) tests respectively. The results from the ITSM and DSR tests were used to indirectly and directly assess the influence of mineral aggregates on bitumen ageing (short-term initial and long-term extended). The testing results show that both the binder and aggregate types can significantly affect the ageing properties of bituminous materials. In addition, it is found that the mineral aggregate can also influence the proportion of ageing amount at different ageing stages.

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***Key words**: Ageing; Bitumen; DSR; ITSM; Mineral aggregate; Oxidation; Stiffness.*

Introduction

Age hardening of bitumen has been widely recognized as one of the main factors that can significantly deteriorate the service quality of bituminous pavements. Findings from previous studies have shown that the ageing properties of bituminous (asphalt) mixtures are influenced by both the bitumen and the mineral aggregate [1-4]. In terms of the influence of aggregate, the findings also indicated that, besides absorbing oily components from bitumen, the petrographic nature of different aggregates may have added effects on the age hardening of bitumen [5-7].

In general (excluding oily component absorption), mineral aggregates can affect the ageing of bituminous materials in the following ways: (1) mineral components on the surface of aggregates may catalyze bitumen oxidation [8]; (2) the mineral aggregate surface may adsorb some of the polar functional groups within the binder and as these adsorbed components contribute to the formation of viscosity build-up, this will delay the increase of binder viscosity upon ageing [9]; and (3) the adsorption of binder fractions by the aggregate surface will disrupt the fractional proportional balance within the bitumen which may lead to a less well dispersed binder and thereby promote the rate of bitumen ageing [10]. The first and third effects increase oxidative age hardening while the second effect reduces ageing.

In order to investigate the influence of aggregate type on bitumen ageing, a typical basic aggregate (limestone) and a typical acidic aggregate (granite) were used to produce different 28 mm Dense Bitumen Macadam (DBM), continuously graded asphalt mixture specimens. The specimens were artificially aged using the SHRP short-term ageing (STA) and long-term ageing (LTA) protocols [11]. Ageing properties of both the asphalt mixture specimens and the recovered binders from these mixtures were tested at different ageing stages (short- and long-term) using the Indirect Tensile Stiffness Modulus (ITSM) and Dynamic Shear Rheometer (DSR) respectively. In addition to the two different aggregate types, two different binders consisting of a 100/150 penetration grade bitumen (V) and its 'short-term' aged equivalent (A) were used in the asphalt mixtures so that the effect of binder type on ageing could be investigated. A total of eight different asphalt mixtures comprising different aggregates, different binder grades and aged conditions were evaluated. Evidence is provided to show that both the mineral aggregate and binder type can affect the ageing properties of bituminous materials.

Experimental Design

Materials

In this study, four different 28 mm DBM continuously graded asphalt mixtures were fabricated with the following material combinations:

- Virgin binder (100/150 pen) + limestone aggregate (LV),
- Virgin binder (100/150 pen) + granite aggregate (GV),
- Aged binder ('short-term' aged 100/150 pen) + limestone aggregate (LA) and,
- Aged binder ('short-term' aged 100/150 pen) + granite aggregate (GA).

The virgin (100/150 pen) binder had a penetration of 122 dmm and a Ring & Ball softening point of 44.2°C. The 100/150 pen binder was also 'short-term' aged using a high shear mixer (with a rotate speed of 3500 rpm) at 165°C for 2.5 days to produce a binder with a penetration and softening point of 69 dmm and 50.2°C respectively. The high shear technique used in this study was found to be equivalent to the 'short-term' aging achieved by the standard rolling thin film oven test (RTFOT) at 163°C and 75 minutes (the penetration and softening point of the same binder after RTFOT aging were 69 dmm and 49.6°C respectively) [12]. The use of the high shear ageing approach was chosen in order to provide a

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Table 1. Batching Details for the Limestone.

Material	Percentage	Sieve Size (mm)								
		37.5	28	20	14	10	6.3	3.35	0.3	0.075
28 mm	20	100	89	14	6.0	5.0	4.2	2.5	0	0
20 mm	10	100	100	87	4.6	2.0	2.0	2.0	1.2	1.0
14 mm	10	100	100	100	87.2	15.2	3.0	2.5	1.9	1.4
10 mm	10	100	100	100	100	81.1	7.0	2.2	1.3	1.2
6 mm	8	100	100	100	100	100	93	18	8.0	8.0
Dust	42	100	100	100	100	100	100	82	25	13.5
Total	100									

Table 2. Batching Details for the Granite.

Material	Percentage	Sieve Size (mm)								
		37.5	28	20	14	10	6.3	3.35	0.3	0.075
28 mm	20	100	85	21	2.4	1.0	0.8	0.7	0.6	0.4
20 mm	10	100	100	88	9.0	1.4	1.1	0.9	0.6	0.3
14 mm	10	100	100	100	74	12	1.0	0.8	0.6	0.3
10 mm	10	100	100	100	100	95	22	6.0	2.7	2.1
6 mm	8	100	100	100	100	100	83	18	4.7	2.9
Dust	42	100	100	100	100	100	100	98	25	11.5
Total	100									

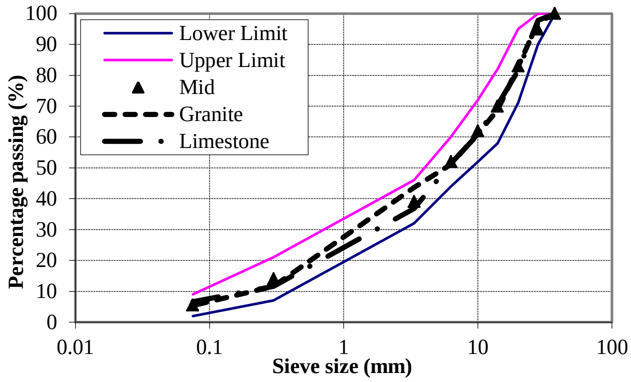

Fig.1. Gradations for the Aggregates in this Study.

sufficient quantity of 'short-term' aged 100/150 pen binder for subsequent asphalt mixture specimen manufacture.

In terms of aggregates, a typical 'basic' aggregate (limestone) and a typical 'acidic' aggregate (granite) were used in this study so that the effect of a typical carboniferous versus siliceous aggregate on bitumen ageing could be shown. The limestone is from Dene Quarry of the UK, and the granite is from Mount Sorrel. Tables 1 and 2 present the batching details of these two aggregates to produce the 28 mm DBM asphalt mixture and Fig. 1 graphically illustrates their gradations.

Previous studies [13-15] have indicated that binder content (and related binder film thickness) can significantly affect the ageing of asphalt mixtures. A high binder content (thick binder film) can effectively limit the effects of age hardening of the asphalt mixture. Therefore, to aid the investigation of the influence of aggregate type on ageing, a relatively low binder content (thin binder film) of 4% by mass was used as the target binder content in this study. In addition, as the ageing of bitumen requires the presence of sufficient oxygen, a relatively high target air void content of 8% was designed for the asphalt mixture specimens.

Although both the target binder content and the percentage of each fraction size are exactly the same for the limestone and granite asphalt mixture specimens, the differences in the detailed gradation of the different fraction sizes associated with the two aggregates and the differences in their absorption values (0.7% for limestone and 0.3% for granite) will lead to differing binder film thicknesses. It was therefore necessary to check the binder film thickness for the two asphalt mixtures. In this study, a newly developed method [16], which takes into account the different sources of aggregates, the percentage of aggregates maintained on each sieve size and the aggregate shapes, was adopted to calculate the binder film thickness and the results are given as follows:

- Binder film thickness in the asphalt mixture with limestone: 6.9 μm;
- Binder film thickness in the asphalt mixture with granite: 8.4 μm.

It was recognized that a difference of 1.5 μm in the binder film thickness between the two asphalt mixtures in this study could not be neglected and special attention was paid to this factor during the data analysis.

The Roller Compactor was used in this study to compact the four asphalt mixture combinations into slabs (with size of 305×305×100 mm) so that sufficient specimens can be produced for following tests. Two slabs were fabricated for each material combination and five specimens were cored from each slab and trimmed into cylinders with a diameter of 100 mm and a height of 60 mm. The layout of specimens from each slab is shown in Fig. 2.

Testing Programme

As stated above, there were four material combinations in this study: LV, GV, LA and GA. These four material combinations were tested in two modes, as shown in Fig. 3. The first mode involved the compaction of asphalt mixture specimens directly after mixing, while

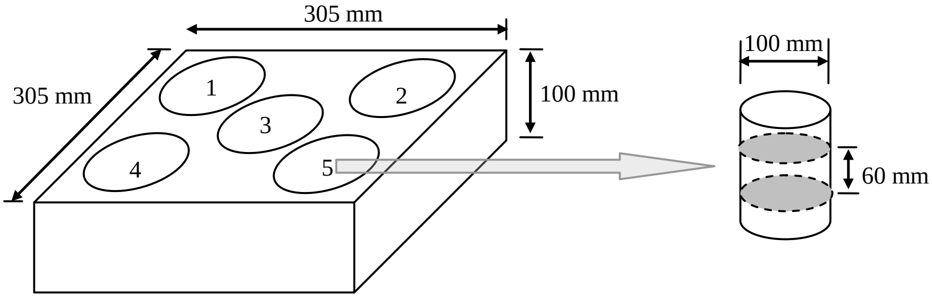

Fig. 2. Layout of Cores from Fabricated Slabs.

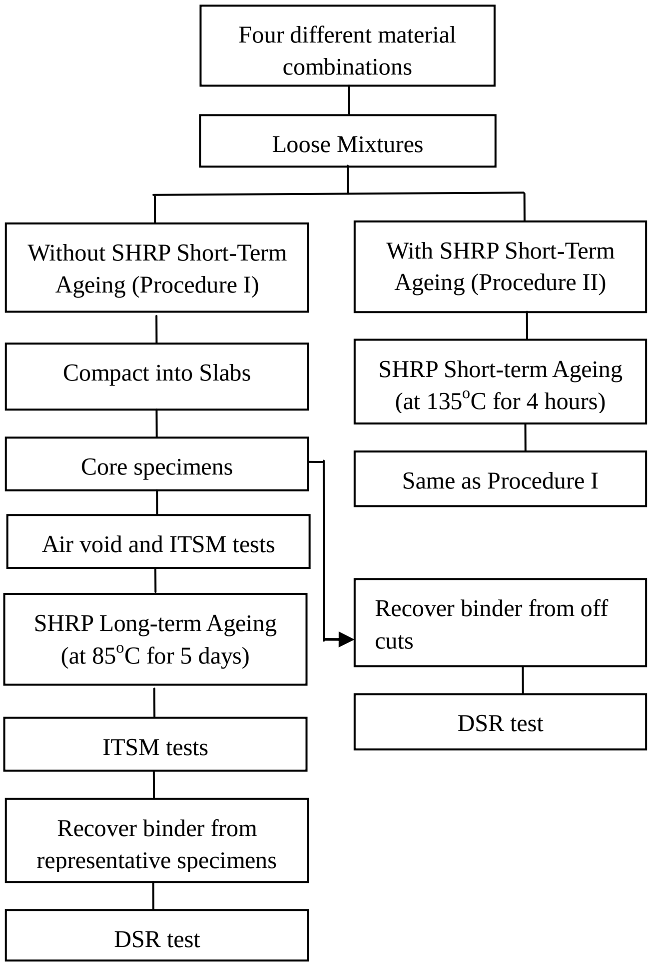

Fig. 3. The Testing Sequence.

the second mode included the short-term ageing of the loose material after mixing using the SHRP short-term oven ageing protocol (4 hours at 135°C) before compaction.

The stiffness of the cored specimens for the different asphalt mixture types was measured using the ITSM test at 20°C with a pulsed load rise time of 124 milliseconds. The specimens were then subjected to the SHRP long-term oven ageing protocol (at 85°C for 5 days), followed by a further ITSM test. Finally, two representative specimens for each material combination were selected and used for the binder recovery and rheological testing.

In addition to the binder recovered from the cores (equivalent to long-term ageing), binder was also recovered from short-term aged asphalt material. This was achieved by using the compacted slabs that had been cored and recovering the binder from the remaining parts of the slabs. The binder recovery was done according to the standard BS EN 12697-4:2005 [17]. A sample of the asphalt was soaked in dichloromethane (methylene chloride) to remove the bitumen from the aggregate into solution. In order to remove the aggregate, the bitumen solution was firstly decanted through a 63μm sieve to remove the larger sized aggregate. The other insoluble matter was then removed from the bitumen solution by centrifuging at an acceleration of at least 15000 m/s^2 for 25 minutes in a sample tube centrifuge. After removal of the aggregate, the bitumen solution was concentrated by atmospheric distillation in a fractionating column. The last traces of solvent were removed from the concentrate by distillation at a temperature of 100℃ above the expected softening point or 175℃, whichever is the higher, with the pressure reduced from atmospheric pressure 100 kPa to 20 kPa with the aid of a stream of carbon dioxide gas. The recovered binder (short- and long-term aged) was then subjected to DSR frequency sweep tests using the Bohlin Gemini 200 model DSR with the following testing conditions:

- Mode of loading: Controlled-strain,
- Temperatures: 0 to 80°C (5°C intervals),
- Frequencies: 0.1 to 10 Hz,
- Plate geometries: 8 mm diameter with a 2 mm gap (0 to 35°C) and 25 mm diameter with a 1 mm gap (25 to 80°C).

Based on the above testing programme and material description, all the specimens in this study can be divided into 8 groups according to the materials they contain and the ageing procedures they were subjected to prior to testing. Table 3 gives a general description for all the asphalt mixture specimen groups that have been used in this study.

The influence of aggregate type on bitumen ageing was assessed by comparing the stiffness modulus of asphalt mixtures produced with the different aggregates. The stiffness results were obtained after short-term ageing (ITSM testing of specimens immediately after compaction) and long-term ageing (ITSM testing of specimens after SHRP long-term age conditioning). The original stiffness modulus results immediately after compaction were taken to represent short-term (initial) bitumen ageing, while the stiffness modulus results after age conditioning were used to represent long-term (extended) bitumen ageing. In addition to the ITSM testing of asphalt mixture specimens, the aged bitumen from the various asphalt mixtures was recovered and subjected to DSR testing to investigate once again initial and extended ageing of bitumen as influenced by aggregate-bitumen interaction.

Table 3. Specimen Description.

Asphalt Mixture ID	Aggregate Type	Binder Condition	Pre-Compaction Conditioning	No. of Specimens
LV_U	Limestone	Virgin	None	10
GV_U	Granite	Virgin	None	10
LA_U	Limestone	High Shear Aged	None	10
GA_U	Granite	High Shear Aged	None	10
LV_STA	Limestone	Virgin	Short-term Aged	10
GV_STA	Granite	Virgin	Short-term Aged	10
LA_STA	Limestone	High Shear Aged	Short-term Aged	10
GA_STA	Granite	High Shear Aged	Short-term Aged	10

Note: L: Limestone G: Granite V: Virgin bitumen
A: Aged bitumen U: Unaged mixture STA: Short term aged mixture

Table 4. Maximum Density Results for Different Asphalt Mixtures.

Asphalt Mixture Type	Maximum Density (kg/m3)
Limestone Aggregate & Virgin Binder	2508
Limestone Aggregate & High Shear Aged Binder	2502
Granite Aggregate & Virgin Binder	2500
Granite Aggregate & High Shear Aged Binder	2494

Experimental Results

Mixture Volumetrics

The air void content for each asphalt mixture specimen was determined using the sealed specimen method according to the standard BS EN 12697-6:2003 [18]. The maximum densities for the four asphalt mixture types, required for the air void calculation, were measured according to the standard BS EN 12697-5:2009 [19] and presented in Table 4.

It should be noted that, with the same aggregate, the maximum densities of asphalt mixtures with aged binder are slightly lower than those with virgin (unaged) binder. As the batching details for the mixtures with virgin and aged bitumen are identical, their masses (aggregate and bitumen) will be the same with the only plausible reason for changes in density being possible changes in mixture volume. Part of the ageing process of the virgin 100/150 pen bitumen will include the loss of lighter, volatile, oily components of the bitumen [10, 20, 21]. The aged binder will therefore contain a smaller amount of lighter, oily fractions compared to the virgin bitumen. When these two binders are mixed separately with the aggregates, the larger amount of oily components in the virgin binder means that potentially more bitumen is absorbed into the aggregate, leaving less free bitumen within the mixture and therefore resulting in a smaller mixture volume and a larger maximum density.

Using the maximum densities shown in Table 4, the air voids for all the specimens were calculated with the average, standard deviation and coefficient of variation of each group summarised in Table 5. From the relative high coefficient of variation for each group, it can be seen that the air voids at different positions of the slab are not the same. It was observed that most of the specimens from the centre of slabs had a void content smaller than the average void content of the specimens from the same slab. This indicates that mixtures in the centre of each slab are subjected to better compaction and are normally denser than the mixtures from the side of slab. In addition, Table 5 also shows that, with the same material combinations, the mixtures that were short-term aged (STA), as described in Section 3, have a larger void content than the ones without short-term ageing (U). This could be the result of the increased difficulty of compacting the mixtures which now contain a slightly harder (aged) binder. The absorption and/or adsorption of binder fractions by the aggregate surface might also be the cause of this phenomenon.

ITSM Testing of Asphalt Mixtures

Table 5. Air Void Content and ITSM Results.

Asphalt Mixture ID	Air Void Content			ITSM Stiffness Before Ageing			ITSM Stiffness After Ageing		
	Average (%)	Std Dev (%)	CV (%)	Average (MPa)	Std Dev (MPa)	CV (%)	Average (MPa)	Std Dev (MPa)	CV (%)
LV_U	8.6	0.8	9	1747	196	11	3500	414	12
GV_U	10.8	0.8	7	1097	179	16	1455	174	12
LA_U	9.2	0.9	10	2373	243	10	4096	392	10
GA_U	10.9	0.9	8	1356	149	11	2194	179	8
LV_STA	9.8	1.0	10	1874	210	11	3235	522	16
GV_STA	11.3	0.6	5	1411	131	9	1770	147	8
LA_STA	9.4	1.0	11	2446	260	11	3837	486	13
GA_STA	10.8	0.7	6	1679	182	11	2014	106	5

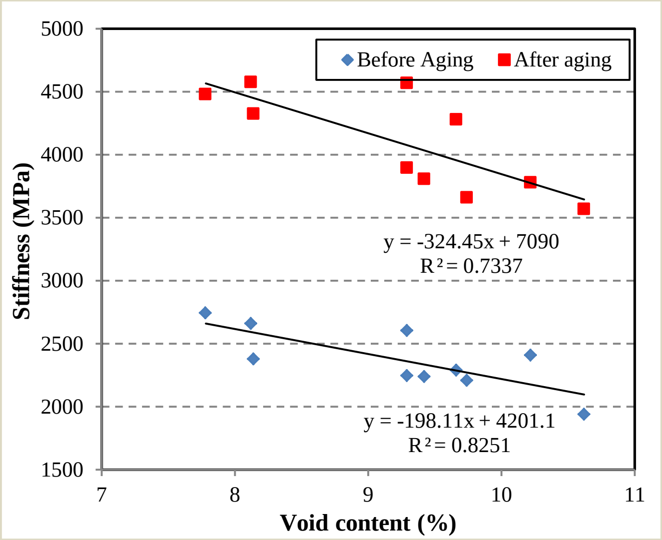

Fig. 4. ITSM Results for LA_U Group.

The stiffness of each specimen was measured both before and after they were aged (SHRP long-term ageing), using the ITSM test at 20^{o}C. Table 5 includes the average, standard deviation and coefficient of variation of the ITSM results for each of the eight asphalt mixture groups.

From the void content results in Table 5, considerable differences can be observed among different groups, particularly between the limestone and granite asphalt mixtures (approximately 9% air voids versus 11%). Accordingly, the average stiffness of each group may not reliably reflect the aggregate influence as they are not at the same void content. In order to eliminate the influence of void content on the study, plots of stiffness versus void content were produced for each group. A linear regression line representing the relationship between the stiffness and void content was then added to each group (eight asphalt mixture types) both before ageing (BA) and after ageing (AA). Based on the stiffness versus air void content relationships, a calculated stiffness for each group was predicted at an air void content of 8%. As an example, Fig. 4 shows the mixture stiffness versus air void content for the limestone aggregate with aged 100/150 pen bitumen without short-term ageing of the loose material before compaction (LA_U).

The results show that the asphalt mixtures with limestone aggregates have higher ITSM values than the granite mixtures that contain the same type of binder. The higher stiffness values for the limestone aggregate mixtures could be attributed to a number of factors. Firstly, it can be seen from Fig. 1 that the gradation of granite aggregate in this study was not as continuous as that of limestone aggregate. This could lead to poorer aggregate-aggregate contact and a larger air void content (as seen in Table 5) for the granite asphalt mixtures and therefore a lower ITSM stiffness. Secondly, slight differences in the surface texture of these two aggregates can lead to different friction and interlock capacities, which may result in different stiffness of the mixtures. Thirdly, as stated previously, the ability of the limestone aggregates (with a water absorption value of 0.7%) to absorb the oily components of the bitumen are higher than that of the granite aggregates (with a water absorption value of 0.3%). The binders coating the limestone aggregates would therefore potentially be harder than those coating the granite aggregates, resulting again in higher ITSM values for limestone aggregate mixtures. Finally, the calculated binder film thicknesses showed that the binder films in granite mixtures were thicker than those in limestone mixtures. According to the findings from previous studies [13-15], the thicker binder films in granite mixtures could also lead to lower ITSM values.

Both the average value and the theoretically predicted stiffness (at 8% air void content) of each group have been used to study the influence of aggregates on bitumen ageing through the calculation of ageing indices (expressed as the ratio of ITSM value after long-term ageing to initial ITSM value) as seen in Table 6. The ageing indices determined in terms of the stiffness at 8% air void content of asphalt specimens are shown in Fig. 5. The ageing indices in Table 6 and Fig. 5 show that, during the SHRP long-term ageing, there is more ageing in the limestone aggregate mixtures compared to the granite aggregate mixtures.

DSR Testing of Recovered Binders

In order to quantitatively illustrate the effects of different ageing conditions and the influence of aggregates on the age hardening of bitumen within an asphalt mixture, the complex modulus (G^*) values at 25^{o}C and 0.4 Hz from the DSR tests on recovered binder for each of the mixture groups were presented in Table 7. In addition to the recovered binder results (after asphalt mixture compaction and after long-term ageing), the G^* values for the unaged bitumen were also included and ageing indices were calculated relative to the unaged binder (*i.e.* ratio of G^* value of recovered binder after ageing to G^* value of original, unaged bitumen). The summarised results of the ageing indices for the

Table 6. Average and Calculated ITSM Stiffness and Ageing Indices.

Asphalt Mixture ID	Average			Calculated (8%)		
	Stiffness BA (MPa)	Stiffness AA (MPa)	Ageing Indices	Stiffness BA (MPa)	Stiffness AA (MPa)	Ageing Indices
LV_U	1747	3500	2.00	1770	3648	2.06
GV_U	1097	1455	1.33	1490	1878	1.26
LA_U	2373	4096	1.73	2616	4494	1.72
GA_U	1356	2194	1.62	1537	2346	1.53
LV_STA	1874	3235	1.73	2157	3684	1.71
GV_STA	1411	1770	1.25	1849	1960	1.06
LA_STA	2446	3837	1.57	2676	4395	1.64
GA_STA	1679	2014	1.20	2199	2363	1.07

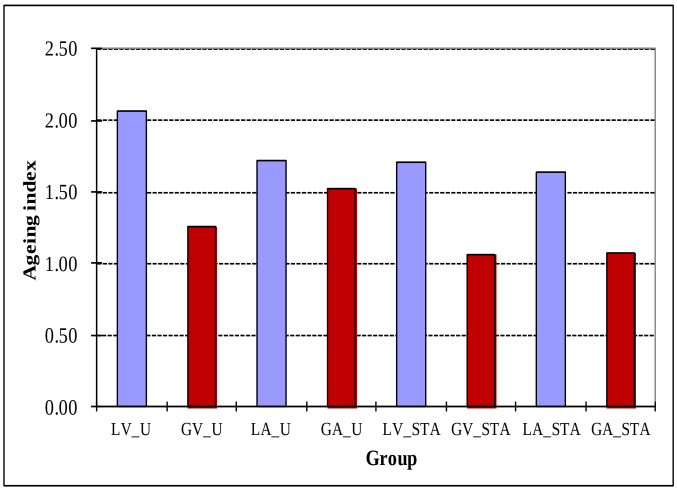

Fig. 5. Ageing Indices in Terms of Calculated Asphalt Stiffness at 8% Air Void Content.

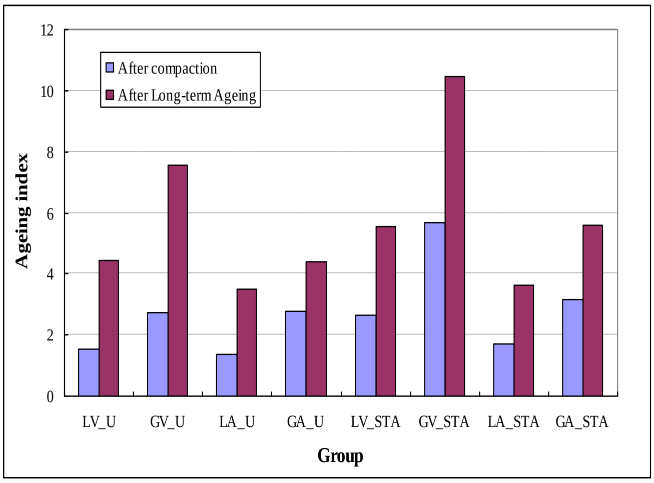

Fig. 6. Ageing Indices in Terms of Complex Moduli of Binders Recovered from Mixtures.

Table 7. Complex Moduli at 25oC and 0.4Hz from the DSR Tests.

Asphalt Mixture ID	*G** Before Mixing (kPa)	After Compaction		After LTOA	
		*G** (kPa)	Ageing Indices	*G** (kPa)	Ageing Indices
LV_U	84	129	1.54	371	4.43
GV_U	84	229	2.73	635	7.57
LA_U	225	308	1.37	790	3.51
GA_U	225	628	2.79	987	4.38
LV_STA	84	221	2.64	466	5.56
GV_STA	84	475	5.67	878	10.47
LA_STA	225	383	1.70	819	3.64
GA_STA	225	706	3.14	1257	5.58

different asphalt mixture groups based on DSR testing of the recovered binders are shown in Fig. 6.

The ageing indices in Table 7 and Fig. 6 show that for a particular binder type, the ageing indices for the binders mixed with granite are much higher than those for the binders mixed with limestone, indicating increased age hardening of the binders in the granite aggregate mixtures. However, it can be seen from Fig. 5 that the ageing indices of the limestone mixtures in terms of ITSM stiffness are higher than those of the granite mixtures, which implies that, during the long-term ageing simulation, stronger age hardening has happened to the limestone mixtures. In order to analyse this phenomenon more clearly, the binder stiffness ratios (BSRs) in terms of bitumen complex moduli (at 25^{o}C and 0.4 Hz) at the same stages of the ITSM tests are calculated using the following equation:

$$BSR = \frac{Complex\ \ Modulus\ \ after\ \ \ LTA}{Complex\ \ Modulus\ \ after\ \ Compaction} \tag{1}$$

The results of the calculation using the above equation are summarised in Fig. 7. It can be seen that the BSRs for the binders recovered from limestone mixtures are also higher than those for the binders recovered from granite mixtures. This result is coincident with the results of the ITSM stiffness of mixtures.

Discussion

The Effects of Binder Type on Ageing

As aforementioned, two different binders were used in this study: the virgin 100/150 pen binder (V) and the 100/150 pen binder that was ‘short-term’ aged at 165^{o}C for 2.5 days using a high shear mixer (A). Because binder A is much harder than binder V, it is expected that, with the same aggregate type and ageing time, asphalt mixtures containing binder A will have a higher stiffness and their recovered binders will be harder than those recovered from asphalt mixtures containing binder V (as seen in Tables 6 and 7).

From Fig. 6, it can be seen that the ageing properties of bitumen can be significantly affected by binder type. When mixed with a particular aggregate (either limestone or granite in this study) and undergoing the same ageing procedure, the ageing indices for V binders are generally higher than those for the A binders. It is a reasonable result as binder A has already been aged at 165^{o}C for 2.5 days and, therefore, the amount of oxidizable molecules (potential for ageing) it contains should be smaller than that in binder V, resulting in a smaller amount of oxidative age hardening in the A binders during the testing programme.

In terms of the ageing properties of the mixtures, it can be seen from Fig. 5 that, for the limestone group, the ageing indices for asphalt mixtures containing binder V are higher than those for asphalt mixtures containing binder A, which correlates well with the DSR results and the observations made in Section 3.3. However, a completely opposite result is observed for the granite group. In this group, the ageing indices for asphalt mixtures containing binder V are lower than those for the binder A mixtures, which seems to imply that during the SHRP long-term ageing, stronger age hardening has happened to the mixtures containing binder A. However, it should be noted that, the ageing index expressed by mixture stiffness is calculated by dividing the mixture stiffness after long-term ageing by the mixture stiffness before long-term ageing; and in fact, the binders inside the mixtures before long-term ageing has already undergone age hardening during mixture mixing, compaction and short-term ageing. Therefore, a more reasonable conclusion that can be drawn from this phenomenon is that, compared with binder A, a bigger proportion of binder V ageing happens during the period before the SHRP long-term ageing.

The Effects of Air Void Content and Binder Film Thickness

Before the effect of aggregate type on ageing is discussed, it should be firstly noted that, for a particular bitumen, the ageing properties of asphalt mixtures and their recovered binders are affected not only by the type of aggregate they contained, but also by the varying air void content and binder film thickness due to the differences in the total gradation and shape factors between the various aggregates. Therefore, special attention should also be paid to these two factors, *i.e.* the air void content and binder film thickness, when the effect of aggregate type on ageing is analysed.

It has been found from the results shown in Table 5 that, at the same testing stage, the granite mixtures had a higher void content than the limestone mixtures with the same binder. This higher void content might also lead to higher ageing indices for the binders recovered from the granite mixtures due to the extra oxygen available to the bitumen. However, as it can be seen from Fig. 6, the ageing indices for the binders recovered from granite mixtures after slab compaction are already much higher than those for the binders recovered from limestone mixtures. During the time before slab compaction, there is no difference in 'air void content' among the loose mixtures with different aggregates. Therefore, the difference in air void content is not considered to be a major factor leading to this result.

With regard to the binder film thickness, the calculations carried out in Section 2.1 have shown that the binder film thickness of the granite mixtures (8.4 μm) is thicker than that of the limestone mixtures (6.9 μm). According to the findings from previous studies, the binder film thickness can significantly affect the ageing of binders in asphalt mixtures. In general, when mixing with a specific type of aggregate and undergoing the same ageing procedure, the binder in thinner film thickness will become harder than the thicker ones. However, Table 7 shows that, in this study, it is the binder from granite mixtures (with thicker binder film thickness) that has higher ageing indices. This indicates that, compared with the effects of differences in binder film thicknesses, the influences of aggregate types are significantly more dominant in this study.

The Effects of Aggregate Type on Ageing

According to the above analysis, it is therefore believed that the differences in ageing indices and/or BSRs are mainly caused by the varying aggregate types. In terms of aggregate type effects, references have stated that besides absorbing oily components from bitumen, mineral aggregates have some added effects on the age hardening of bitumen [4-8]. Firstly, the charged and polarised aggregate surface can adsorb polar groups (either naturally occurring ones or the oxidation products) within bitumen, which may decelerate the bitumen hardening process. In addition, some mineral components on the surface of aggregates can catalyze bitumen oxidation. However, the catalytic activity of the aggregate surface can be limited by the adsorption of polar components from bitumen to aggregates.

Anderson *et al*. [22] stated that aggregates with the least adsorption of highly polar fractions (e.g. quartzite and granite) exhibit the greatest catalytic effect in bitumen oxidation, whereas those showing the largest adsorption (e.g. limestone) exhibited the smallest catalytic effect. This statement is proved to be correct by the data shown in Fig. 6. Firstly, the stronger catalytic ability of the granite led to more oxidative ageing of the binders in the granite mixtures resulting in an increased ageing index. Secondly, the limestone adsorbed a greater amount of polar components from the bitumen which, as stated by Petersen *et al*. [5], could protect the adsorbed components from oxidation. This would result in a decrease in ageing indices for the binders recovered from limestone mixtures. Thirdly, Petersen *et al*. [5] declared that some of the polar component adsorption might be irreversible and most of these are not recoverable through normal bitumen recovery techniques. Therefore, it is possible that some polar components were still kept on the limestone surface after bitumen recovery, which can also lead to lower ageing indices for the binders recovered from limestone mixtures due to the lower polar components concentrate. Finally, the different abilities of these two aggregates in absorbing oily fractions might also be one of the main causes for this phenomenon. The absorbed binder was inside the aggregate, it was protected from oxidation and/or evaporation; therefore, when the absorbed components were recovered back to the bitumen, it led to a softer recovered binder for the limestone mixture.

An interesting phenomenon can be observed by comparing Fig. 5-7. The ageing indices in terms of bitumen complex moduli (shown in Fig. 6) illustrate that, during either the period before LTA or the whole ageing programme, stronger age hardening has happened to the granite mixtures; whereas the ageing indices in terms of asphalt stiffness (shown in Fig. 5) and the BSRs in terms of bitumen complex moduli (shown in Fig. 7) show that, during the long-term ageing simulation, stronger age hardening has happened to the limestone mixtures. It is therefore quite evident that, for a particular bitumen, the aggregate type can not only significantly affect the ageing of bituminous materials, but also can it influence the proportion of total age hardening in different ageing periods.

With regard to the effect of aggregate type on the proportion of ageing that happened in different ageing periods, it is believed to be caused by the degradation of adsorbing and catalyzing abilities of aggregates. As stated in previous studies [23-25], only the charged sites on the aggregate surface are able to interact with (adsorb) the polar components of bitumen. For the limestone (basic) mixtures where adsorption dominates, as the ageing proceeds, the constant adsorption would lead to reduced charged active sites on the aggregate surface and, therefore, its ability of decreasing the amount of ageing will be weakened. In addition, Petersen *et al*. [5] stated that the interactions of bitumen components with the aggregate surface (adsorption) reduce the surface catalytic activity of bitumen oxidation. For the granite (acidic) mixtures, although the catalytic effect is dominant in the bitumen ageing, adsorption of polar components from bitumen to aggregate surface still happens during ageing. This will deactivate the granite surface in accelerating the bitumen ageing, which can result in smaller proportion of age hardening in later stage of granite mixture ageing.

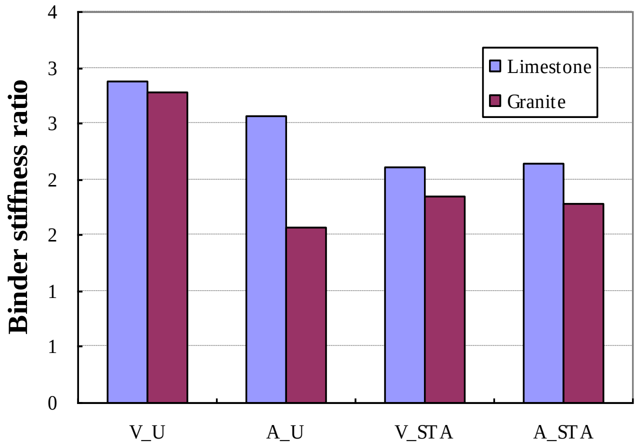

Fig. 7. Effect of Aggregate Types on the Binder Stiffness Ratio in Terms of Complex Modulus (LTA/comp.)

Conclusions

In this study, an ageing test programme involving two different bitumen types and two aggregate types was conducted by testing the stiffness of asphalt mixture specimens and the complex modulus of recovered binders at different stages of oxidative ageing. Based on this study, several conclusions can be drawn as follows:

- The testing results showed that both the bitumen and aggregate types can significantly affect the ageing properties of the bituminous materials;
- It was observed that, with the same material combinations, the short-term aged (STA) mixtures have a larger void content than the ones without short-term ageing (U), which indicates that ageing procedure can considerably increase the difficulty of compacting the mixtures;
- The aggregate type can not only significantly affect the ageing of bituminous materials, but also can it influence influence whether the majority of the total age hardening occurs in the initial ageing period or in the subsequent period;
- It is deduced from the testing results that part of the adsorbed polar components and most of the absorbed oily components were recovered from the aggregate surface back to the bitumen during binder recovery. Because these adsorbed and absorbed fractions do not contribute to the stiffness-built of the mixture, special attentions should be paid to the use of rheological properties of recovered binder on bitumen ageing studies in the future.

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