THE USE OF VEGETABLE OIL AS A REJUVENATOR FOR ASPHALT MIXTURES

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ABSTRACT

This paper presents a laboratory study to demonstrate the effectiveness of using vegetable oils as rejuvenating agents for aged asphalt mixtures. The work focused on proposing a laboratory aging protocol whereby loose asphalt base course mixtures oven aged at 150°C for a range of durations, namely 2, 4, 6, 8 and 10hrs prior to compaction. Using this technique, 5 mixes were thus produced at various stages/levels of aging. An additional set of 5 batches were produced and aged in the loose state for 10 hours at 150ºC before rejuvenating the mixtures using vegetable oil. Vegetable oil was added at percentages ranging from 4 to 8% by weight of the bitumen. Rejuvenation of the mixture using the vegetable oils was then assessed by the mechanical and rheological performance of the material and binders. Furthermore, the effect of the addition of vegetable oil rejuvenators was also investigated using SARA analysis. Results indicated that the aging and rejuvenating protocol employed in the lab has been effective, with results of the recovered penetration and softening point tests demonstrating that it is possible to rejuvenate an aged mixture thereby improving the recovered penetration by standard penetration grade designation. However further work must be done to improve the correlation between mechanical performance and rheological characterisation particularly if a rejuvenation chart were to be developed.

Keywords: Rejuvenating agents, vegetable oil, aged asphalt, rejuvenation, used vegetable oil

1. BACKGROUND

Bitumens have found uses in many sectors, the most commonly used area is in that of paving. Typically, bitumens are used in road construction as binders for aggregates as they are water resistant, have good adhesive properties and, until recently were cheap and available in vast quantities. Although bitumen can be considered a by-product of the petroleum industry, the availability and price are likely to be market driven. Producers are increasingly likely to look toward 'cracking' the molecules to produce shorter chains of higher value. This, coupled with oil prices reaching an unprecedented highs in recent years, does not fill the industry with much optimism.

On the other hand, atmospheric exposure causes asphalts to gradually age due to weathering and oxidation. The increase in rigidity of the asphalt results in negative consequences with respect to flexural capacity (fatigue cracking) and thermal response (thermal cracking). Therefore, the timely and efficient maintenance of asphalts is crucial. The addition of rejuvenating agents in small quantities can bring asphalt back to life through softening of the binder and restoring flexibility of the mix. It is usually required that the rejuvenating agent will soften the binder in the reclaimed asphalt to the preferred levels for a new mix, and that the rejuvenated binder will have physical properties meeting the local specifications for the new asphalt mixture. The ability of the rejuvenating agent to do this depends on the viscosity and the quantity added to the aged asphalt mixture.

There is a need for a rejuvenating agent that is flexible in its manner of use, which enables the properties of the rejuvenated asphalt to be restored as required across a wide spread of specifications. Ideally, the rejuvenating agent should also have green, environmentally friendly properties, and preferably, the product is made in whole or in part from a recycled or renewable material.

Earlier work carried out by the authors [2] have demonstrated the viability of utilising vegetable oils as extenders and viscosity modifiers for conventional pen grade bitumens. A wide range of non-drying and semi-drying vegetable oils, including numerous edible oils are potentially highly compatible with penetration grade bitumens. As an example, when less than 5% non-drying vegetable oil (by mass of bitumen) is blended into a 40/60 pen, the bitumen is transformed into an 80/100pen binder having indistinguishable rheological properties from a straight run 80/100 pen as measured by DSR temperature and frequency sweeps.

Encouraged by the superb compatibility and fluxing power of vegetable oils, it was decided in this investigation to explore the usefulness of the vegetable oils even further by assessing their capacity to act as rejuvenating agents for aged asphalt mixes.

Initial experiments were carried out using laboratory prepared oven aged asphalt mixtures to eliminate variability obtained when using reclaimed materials. On successful completion of this stage, reclaimed materials were brought from site to the laboratory and mixtures were prepared once again and tested for volumetric and mechanical properties. For final validation of the viability of the proposed process, field trials using a hot mix recycler operated by Bardon Contracting UK were carried out.

2. LABORATORY EVALUATION OF VEGETABLE OIL AS ASPHALT REJUVENATOR

In this experiment, a proprietary basecourse material grading and 40/60 pen binder were employed, the grading of which can be found in table 1. Following mixing with the binder, the loose asphalt mixes were spread in metallic trays and oven aged at 150°C for a range of durations, namely 2, 4, 6, 8 and 10hrs prior to compaction. Using this technique, 5 mixes were thus produced at various stages/levels of aging. The mixes were roller compacted and the slabs were cored producing 24×100mm diameter specimens which were tested for their volumetrics (density and voids) and stiffness (ITSM) properties.

An additional set of 5 batches were next produced and these were all oven aged in the loose state for 10 hours at 150°C. Next, a pre-determined (but different) amount of virgin vegetable oil was added to each of the 10 hour aged asphalt batches and each blend was thoroughly mixed using a heated paddle mixer (2 minutes at mixing temperature). The amounts of oil added to the mixes were 4, 5, 6, 7 and 8% oil by mass of bitumen. The rejuvenated hot mixes were finally roller compacted and the slabs cored producing 20×100mm diameter specimens which were subsequently tested for volumetrics and ITSM.

Table 1: Particle size distribution of asphalt mix adopted in this investigation

Sieve size	Target grading (% passing)
20 mm	100
14 mm	95-100
10 mm	72-88
6.3 mm	53-67
2.0 mm	27-37
1.0 mm	18-28
63 um	6-10

Research findings reported in the literature [1] indicates that the older/aged binder from reclaimed asphalt pavement (RAP) material may not fully blend with new/virgin bitumen which is added to the mix during the process of hot mix recycling. A recycled mix with an inhomogeneous distribution of binder having variable stiffness is likely to result in un-rejuvinated zones or pockets of very stiff/aged binder which in turn act as stress concentrators and sources for fatigue crack initiation. As a result of this work it was considered important to measure performance properties of the mixture such as stiffness and fatigue in order to assess the mechanical behaviour of rejuvenated mixtures rather than purely relying on recovered binder properties.

Average results of the mechanical properties for both the aged and rejuvenated mixtures can be found in table 2.

Table 2: Summary of mechanical properties of aged and rejuvenated mixtures

Mixture Reference	Max. Density (kg/m³)	Procedure C (Sealed)		Procedure A (Un-sealed)		Average Stiffness (MPa)
		Bulk Density (kg/m³)	Air Voids (%)	Bulk Density (kg/m³)	Air Voids (%)	
Control, 0hrs Aging	2494	2310	7.4	2379	4.6	2273
Oven Aged 2hrs	2570	2396	6.8	2452	4.6	3897
Oven Aged 4hrs	2588	2367	8.5	2427	6.2	3754
Oven Aged 6hrs	2584	2390	7.5	2453	5.1	4595
Oven Aged 8hrs	2578	2362	8.4	2430	5.8	4824
Oven Aged 10hrs	2578	2368	8.2	2426	5.9	5288
10hrs Aging, 4% Veg Rejuvenator	2578	2379	7.7	2440	5.4	3096
10hrs Aging, 5% Veg Rejuvenator	2578	2394	7.1	2444	5.2	2676
10hrs Aging, 6% Veg Rejuvenator	2578	2391	7.3	2449	5.0	1809
10hrs Aging, 7% Veg Rejuvenator	2578	2391	7.3	2450	5.0	1833
10hrs Aging, 8% Veg Rejuvenator	2578	2398	7.0	2460	4.6	1630

The increase in stiffness of the compacted cores with increasing loose mix aging time are shown by the red data points in figure 1. The effect of adding oil (rejuvenation) on the 10 hour aged loose mix is also clearly shown in figure 1 (green data points). The results clearly demonstrate the effectiveness of vegetable oil as a rejuvenating agent.

It can be seen from figure 1, that starting from a 10 hour oven aged mix (approx. 5500 MPa), it is possible to rejuvenate that mix back to its original 40/60pen unaged state (approx. 2500MPa) by introducing approximately 5% vegetable oil during the hot mix recycling stage (referred to henceforth as Vegetex 50).

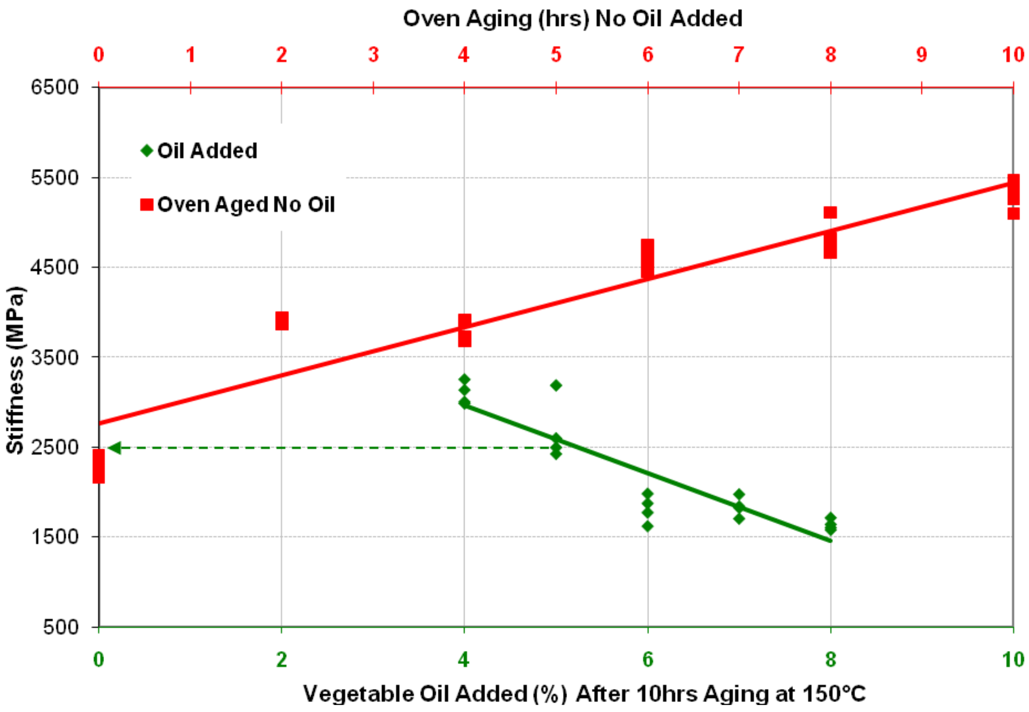

Figure 1: Effect of thermal aging time on stiffness of asphalt mixtures (red line), and the effect of vegetable oil as a rejuvenator on 10hr oven aged asphalt mixtures (green line).

2.1. Indirect Tensile Fatigue Testing (ITFT)

Following initial stiffness testing the specimens were ITFT tested using a Nottingham Asphalt Tester (NAT). Figure 2 shows the 40/60 pen and Vegetex 50 and the 10h aged mix data points all fall roughly into a single fatigue line. This confirms the principle that when asphalt mixture fatigue data are represented on a strain (rather than stress) versus number of cycles to failure plot, for any one mix type (*i.e*. gradation, voids, binder type & binder content) a single fatigue line will represent the performance of that mix. Any changes in stiffness of the binder (*e.g.* due to changes in test temperature or binder ageing) will simply cause the data points to move up and down the same line (which is exactly what has happened to the 10h aged samples).

On the other hand, it is very interesting to note that in figure 2, all of the fatigue results of the oil rejuvenated samples fell above the fatigue line (*i.e.* rejuvenation had unexpectedly caused an improvement in fatigue performance). However, one must not forget that the rejuvenated samples have a slightly higher binder content (due to the addition of vegetable oil) and this extra binder volume is bound to increase fatigue life. Had it been possible to keep the total binder content constant (*i.e.* volume of bitumen + volume of vegetable oil) the results of the rejuvenated samples would have most likely also fallen on the same fatigue line.

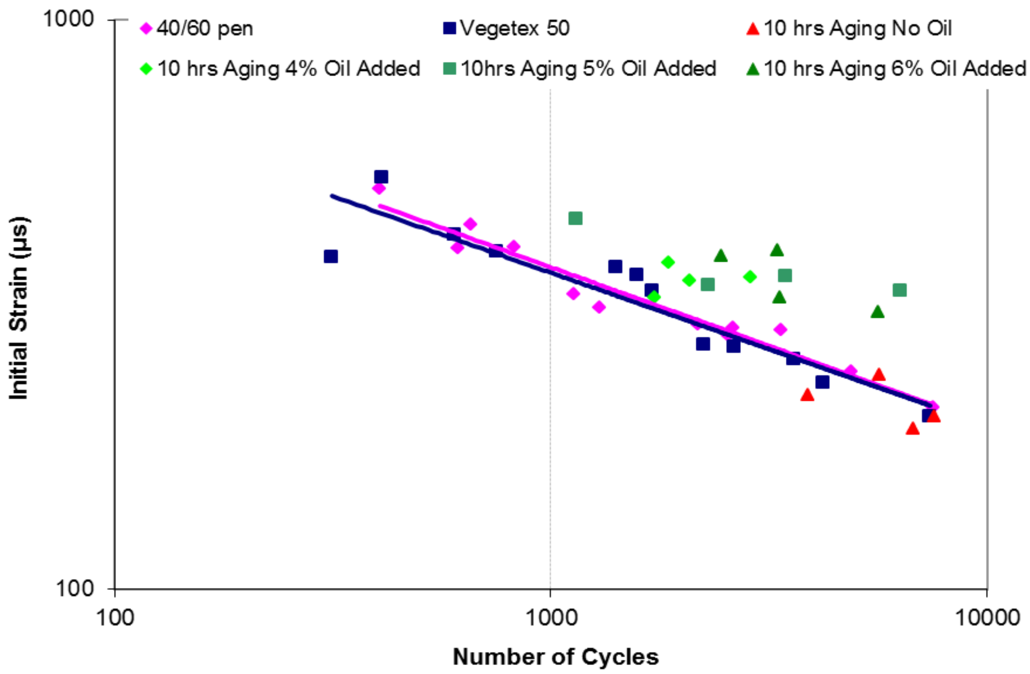

Figure 2: Fatigue performance of the aged and rejuvenated asphalt mixtures.

2.2. Binder Testing

The extent of rejuvenation can be a major uncertainty and cause for concern during hot mix recycling of asphalt. Assumptions are made that all materials are thoroughly mixed and that the blended binder meets requirements such as penetration, softening point or viscosity.

In addition to stiffness and fatigue testing, binder from two sets of mixtures was recovered, allowing penetration and softening point testing to be carried out. The two mixtures chosen for binder testing were as follows; oven aged for 10hrs and oven aged for 10hrs with the addition of 5% vegetable oil rejuvenator. In addition DSR frequency sweeps were also conducted to enable more accurate comparisons to be made between the level of aging and rejuvenation versus conventional penetration grade bitumens. By comparing DSR results of the aged mixtures to standard industry grade bitumens it allows the aging protocol employed in this experiment to be described using industry wide terminology, e.g. aged by one grade.

2.2.1 Recovery of Aged Binder

Binders were recovered from the chosen mixtures in accordance with BS EN 12697-1. A summary of the recovered binder properties in comparison to a virgin sample of 40/60pen recovered from an unaged mixture can be found in table 3. Results of the recovered binder tests demonstrate that it is possible to rejuvenate an aged mixture thereby improving the recovered penetration by a standard penetration grade designation. Results of the recovered penetration suggest that rejuvenation has taken place, however it was difficult to draw accurate conclusions and further testing was conducted using a DSR.

Table 3: Recovered binder results from aged and rejuvenated mixtures.

Mixture Reference	Penetration (dmm)				Softening Point (°C)		
	1	2	3	Avg	1	2	Avg
40/60pen, virgin bitumen	28	22	26	**25**	64.0	64.0	**64.0**
Oven Aged 10hrs	20	19	20	**20**	64.2	64.2	**64.2**
10hrs Aging, 5% Veg Rejuvenator	34	35	35	**35**	61.6	61.6	**61.6**

2.2.2 DSR Frequency Sweeps

In order to validate the findings obtained through penetration and softening point tests performed on the recovered binders, DSR frequency sweeps was also conducted in the region of linear viscoelastic (LVE) response. The DSR tests reported were performed under the following conditions: controlled strain mode of loading, test temperatures ranging from 0 to 80°C in 5°C increments, 0.01 to 10Hz test frequency. The rheological properties of the binders were interpreted in terms of the complex modulus in shear (G*) and phase angle (δ) versus frequency.

Results of frequency sweeps conducted at different test temperatures were shifted along the frequency axis to produce master curves at a reference temperature of 25°C. The 25°C master curves of the two recovered binders compared to a standard 10/20 pen and a 40/60 pen are presented graphically for both the complex modulus and phase angle results in figure 3 and figure 4 respectively.

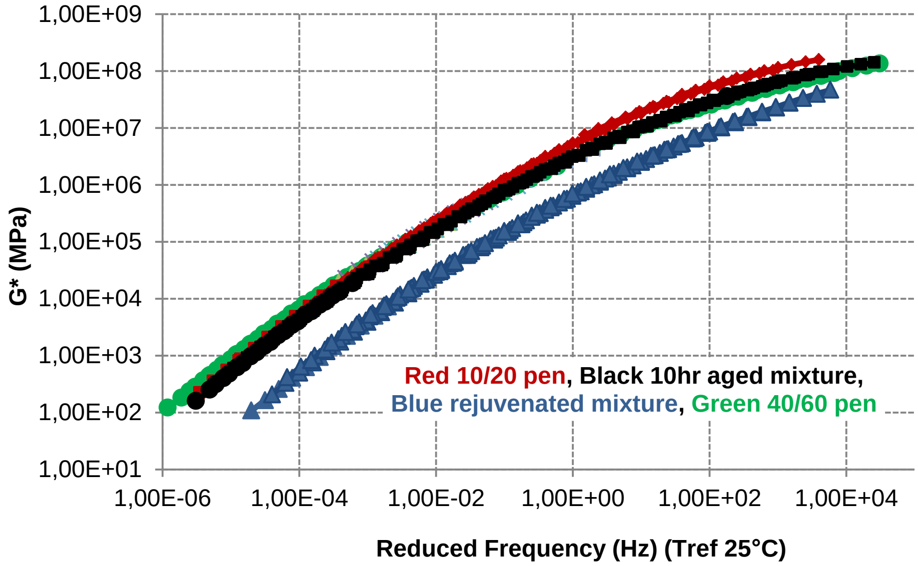

Figure 3: Complex modulus (G*) master curves at 25°C reference temperature for the recovered 10hr aged binder versus the rejuvenated mixture and compared to a standard 10/20pen binder and 40/60pen binder.

In figure 3 there is a reduction in G* for the rejuvenated mixture suggesting that possibly too much rejuvenator has been added. This reduction in stiffness is in line with the increase in recovered penetration results in table 3 and also fits with ITFT results in figure 2 as the rejuvenated mixtures lie above the fatigue line suggesting that the binder used was much softer.

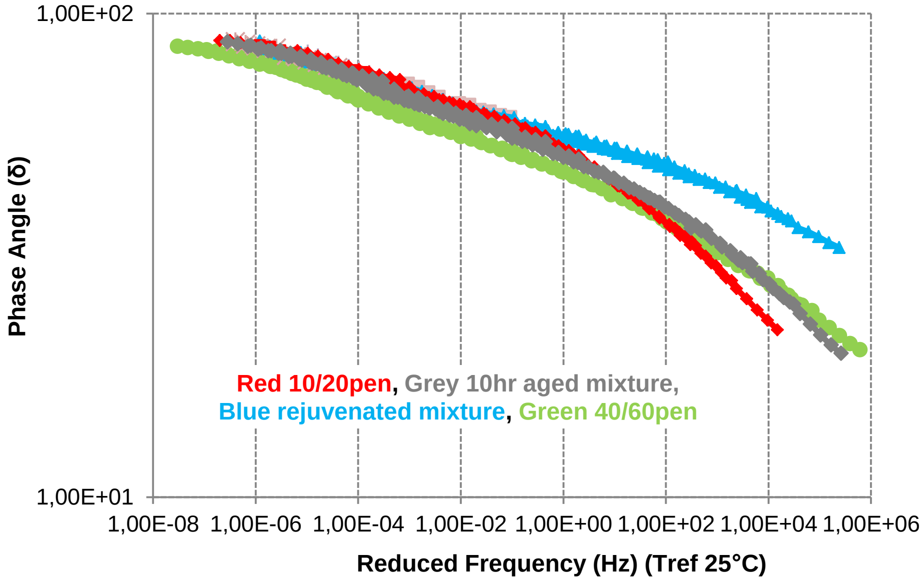

Figure 4: Phase angle δ master curves at 25°C reference temperature for both recovered 10hr aged binder and rejuvenated mixture and also standard 10/20pen binder and 40/60pen binder

2.3 SARA Analysis

In addition to the virgin vegetable oil it was decided that used vegetable oil (UVO) also be utilised and compared against that of the virgin oil. Using the method described earlier by the authors [3], SARA analysis was carried out on binder recovered from three asphalt mixtures, namely;

- Aged asphalt as the control (following 10hr aging protocol)
- Aged asphalt rejuvenated with 5% virgin vegetable oil
- Aged asphalt rejuvenated with 5% UVO

To complete this exercise, additional chromatographic analysis was carried out on the virgin 10/20 pen and 40/60 pen bitumens and also virgin and used vegetable oil are also included in the table of results. Zoorob *et al* [4] reported on a more recent version of the colloidal state of bitumens referred to as the "Colloidal Instability Coefficient or Index" (CI). The Index of Colloidal Instability (CI), which is defined as the ratio of the amount of asphaltenes and saturates to the amount of resins and aromatics, can be used to describe the stability of the colloidal structure in petroleum products and bitumens. The higher CI, the more the bitumen is regarded as Gel-type bitumen. The lower CI, the more stable the colloidal structure. This relationship can be found in equation 1.

CI = [Asphaltenes + Saturates] / [Resins + Aromatics] **Equation 1.**

Results of the SARA analysis and calculated colloidal index (CI) can be found summarised in table 5.

Table 5: SARA analysis, a summary of various binders

Binder	SARA fraction				
	Saturates	**Aromatics**	**Resins**	**Asphaltenes**	**CI**
10/20 pen bitumen	3.65	45.85	31.55	18.96	0.292
40/60 pen bitumen	5.64	37.75	38.17	18.43	0.317
Virgin veg. oil	0.01	0.46	98.74	0.80	0.008
UVO	<0.01	1.18	97.84	0.99	0.010
Aged asphalt (Control)	7.72	8.21	55.37	28.70	0.573
Aged asphalt rejuvenated with virgin veg. oil	11.57	6.35	57.20	24.88	0.574
Aged asphalt rejuvenated with UVO	12.46	5.51	41.25	40.78	1.139

The results in table 5 do not appear to follow any logic, for example the asphaltene content increases from 24.88 to 40.78% when UVO is used instead of virgin vegetable oil to rejuvenate the aged mixture, particularly when the SARA analysis of the UVO on its own is almost identical to the virgin vegetable oil, with both oils lying in the resin fraction. Furthermore, normally as bitumen ages, the CI increases, as shown between the virgin 40/60 pen and aged asphalt control. However, the CI of the 40/60 pen is unexpectedly higher than the 10/20 pen which could be as a result of different crude sources and is considered negligible in the case of these results.
Researchers [5] have documented that the flocculation of colloidal particles is an indication of stability loss within the bitumen; this should therefore provide a good indication of the rejuvenation process. However when rejuvenating the aged asphalt with virgin oil the CI value did not change at all putting a big question on whether the SARA test is suitable for a sample that has a combination of mineral and vegetable oils or indeed the right test for present day bitumen's. Recent work [6] has shown that a more precise description of bitumen as a colloidal dispersion is starting to arise. It shows that a simple solvation parameter allows quantification of the effect of the asphaltenes on the rheological properties of the bitumen. This appears to be a promising approach in order to understand the more complex phenomena such as bitumen aging or the diffusion of rejuvenating oils into the bitumen and as such needs further investigation when evaluating such additives.
A key source of error for this experiment is that only one sample was run per binder type, results therefore serve as an indication only to aid in the explanation of the blending processes. Should additional chemical analysis be carried out 5 test samples should be run per binder type to assess repeatability.

3. DISCUSSION OF LABORATORY RESULTS

The aging and rejuvenating protocol employed in the lab has been effective, with results of the recovered penetration and softening point tests demonstrating that it is possible to rejuvenate an aged mixture thereby improving the recovered penetration by standard penetration grade designation.

The viability of applying the Hirsch model to predict the stiffness of the mix from binder G* values, or alternatively the model can be used to back calculate the binder stiffness at a given frequency has also been demonstrated successfully.
Although initial findings show that mixtures can be rejuvenated through the addition of vegetable oil, laboratory mixtures are of course prepared in a controlled environment therefore it was necessary to verify and validate the initial findings through field investigations. It is therefore recommended that the study be extended to include work on reclaimed asphalt pavement (RAP) mixtures rather than lab simulated aging.

This work needs extending to validate initial findings and examine mixtures fully by evaluating a range of properties.

With more laboratory trials leading to an extended version of figure 1, it would be possible to confidently rejuvenate any old/aged asphalt surfacing back to any target stiffness level.

4.0. MANUFACTURING TRIALS

To verify the results in the field, a hot mix recycler was used to produce bulk samples which were taken back to the laboratory for testing. Trials were also carried out in order to establish how the oil should be added to the mixture. The hot mix recycler utilises 100% RAP. Normal mode of operation sees that 5 tonnes of cold RAP is added to the mixer, heated to approximately 160°C and continuously mixed (using lifters rather than paddles) for 20 minutes. Subject to visual inspection 160/220 pen binder (typically 20kg per 5t batch) maybe added to rejuvenate the mixture, before being released.

Following evaluation of the lab work, a 3t batch with 3% UVO added directly to the plant was produced. Generally the material produced by the hot mix recycler is used in hand lay applications. As a result the batch was evaluated by a contracting crew using conventional hand lay procedures. The crew found the workability to be poor. It was therefore decided to try batches with the maximum oil content used in the laboratory study. The batches were again evaluated by the contracting crew using hand lay procedures. Batches with higher oil contents (9% and 11%) were produced and found to have improved workability. Bulk samples of each were brought back to the laboratory for evaluation.

All mixtures consisted of 100% stockpile RAP, were heated to between 130°C and 160^{0}C, and mixed for a total of 20 minutes. A photograph of the hot mix recycler can be found in figure 5. Figure 6 shows the loading of material into the drum; UVO was added to the material once the drum was full for the first two mixtures trialled. Figure 7 shows the addition of oil for the remaining mixtures trialled. Figures 8 shows the discharge of material from the recycler whilst figures 9 and 10 show laying of material through a mini paver.

Figure 5: Hot mix recycler

Figure 6: Loading material into the recycler

Figure 7: UVO added to the bucket of the loading shovel prior to mixing

Figure 8: Emptying of the material from the hot mix recycler

Figure 9: Material being laid through a minipaver

Figure 10: Material containing 11% UVO, laid through a minipaver

5. DISCUSSION

With a move towards greener and more sustainable material sources once the principle of rejuvenation of rejuvenation had been confirmed in the laboratory using both virgin and used oils, only used vegetable oil should be used going forward. Findings confirm that used vegetable oil can successfully rejuvenate reclaimed asphalt planings. Data from the field trials revealed that the sequence of addition could significantly affect the ability of the vegetable oil to rejuvenate the aged mixture. The rejuvenator has greatest effect when added to heated RAP.

As expected a trend of improved workability was observed with higher oil content, though it was difficult to differentiate between effectiveness of the rejuvenator and increased overall binder content. Furthermore, reclaimed asphalt by its very nature is varied and sampling of such material means that a larger sample size must be assessed in order to fully validate the findings of these trials. Whilst mixtures may perform well initially, if this area of work is to be continued, it would be prudent to evaluate long term aging and durability to examine the possible effect of the localised stiffness. Olard *et al* [7] echoes this recommendation stating that there is little information available about the effect of such high RAP contents on the mechanical properties of the resulting mixtures. Thus, it becomes an important priority to study and determine the effects that various types and percentages of RAP have on combined asphalt binders (RAP binder and virgin binder) and mixture properties [7].

Generally, the development and implementation of asphalt recycling has led to considerable progress with regard to pavement performance as well as environmental concerns, such as workers' health and preservation of natural resources. However, the investigation has also given rise to a number of questions that need to be resolved in order to facilitate further development. The question is; if excessively aged binder components influence the performance of recycled asphalt not only with regard to stiffness, but also with regard to structural stability, which in turn leads to the second question; is there a limit regarding the number of times an asphalt pavement can be recycled in order to avoid the negative influence of aging [8]. Information obtained during experiments suggest that the real question is, what rejuvenating agents can we add and how many time can we rejuvenate without deviating too much from the original chemistry of the bitumen (assuming the original chemistry is also the optimum performing material)? Particularly if each time rejuvenation takes place the rheology changes back into a structure that resembles the original material. It is therefore recommended that in addition to more detail, a mechanical property study to assess the effect of re-recycling should also be addressed.

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REFERENCES:

[1] Analysis of unbalanced binder oxidation level in recycled asphalt mixture using GPC, Doh Y S, Amirkhanian S N, Kim K W, Construction and Building Materials Article In Press, 2007

[2] A Study of the Effect of Bitumen/Vegetable Oil Blends on Asphalt Mixture Performance, Cope M, Allen B and Zoorob S.E, 2007. Fourth International Conference Bituminous Mixtures & Pavements, Thessaloniki, Greece, April 2007.
[3] Vegetex in the laboratory and field, Bailey H. K, and Zoorob S. E, E and E Congress 2012
[4] Review and analysis of the variations in aging rates of equivalent grade road bitumens based on colloidal structure, Part 1: critical review & Part 2 Experimental Investigation, Zoorob S. E and Phillips P, Liverpool John Moores University, February 2006.
[5] Effect of origin and technology on the chemical composition and colloidal stability of bitumens, K. Baginska and I. Gawel, Fuel Processing Technology 85, pp1453-1462, (2004)
[6] The colloidal structure of bitumen: Consequences on the rheology and on the mechanisms of bitumen modification, D. Leueur, Advances in Colloid and Interface Science, 145,pp 42-82, 2009.
[7] Laboratory performance-based assessment of half-warm mix asphalts with high recycling rate by means of the factorial experiment design approach, F. Olard, E. Beduneau, N. Seignez, S. Dupriet and D. Bonneau, Advanced Testing and Characterization of Bituminous Materials, Taylor & Francis Group, London, 2009.
[8] Investigations on bitumen diffusion and structural stability, Karlsson R and Isacsson U, Association of Asphalt Paving Technologists, AAPT, 2003