Sustainability & Environment

**Multi-recycling of asphalt mix with Reclaimed Asphalt and rejuvenator**

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Abstract

Asphalt materials are effectively 100 % reusable. The reuse and recycling of Reclaimed Asphalt (RA) from old pavements has become normal practice. In recent years, the interest of using rejuvenators has brought recycling to the next level for increasing the RA content in new asphalt mix, or with very aged RA binder, or to improve the processing at the mix plant and pavement construction. In Europe, Germany or The Netherlands have been recycling RA for more than 20 years. In Japan, for more than 40 years, RA is reused up to 80 % and the use of a rejuvenator is a common practice. The question of multi-recyclability, therefore, is becoming more of interest. Japan is already at the third if not fourth cycle, Germany already at least second generation of recycling asphalt materials into asphalt materials. So far there are limited studies looking at multi cycle of recycling. Most often they are based on binder evaluation using laboratory binder aging procedures. This paper presents a laboratory investigation of multi-recycling asphalt mix containing RA and a bio-based rejuvenator. For this purpose an asphalt mix was made with 50 % of RA treated with rejuvenator. The mix was aged in lab for short term and long term procedure. After this aging it was reused again after rejuvenation into a new asphalt mix at a 50 % content and re-aged. At each different stages, binder from mix was extracted and recovered for further analysis. Compared to standard binder aging conditioning, the results from mix aging have shown a similar trend in the changes of the properties. The bio-based rejuvenator was able to restore the main properties of the binder even after the second cycle of recycling. It demonstrates the benefit of the rejuvenator to restore and maintain the durability and properties over time.

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# 1 INTRODUCTION

Recycling and the reuse of Reclaimed Asphalt (RA) are becoming a practice of interest for decades for the paving industry, bringing economic and environmental benefits [1]. With technologies and increased need for high performance materials, the use of rejuvenators is foreseen to take recycling to the next level by increasing the RA content in asphalt mix, the use of more adverse RA, containing hard aged binder, or to facilitate the processing at the mix plant [2]. However, this has to be achieved by producing asphalt mix having at-least-equal-to-the-same-level of performances than a standard asphalt mix made with virgin materials only. For this, the rejuvenators have to restore the flexibility at intermediate and low temperature, without compromising the high temperature behaviour against rutting, and ensuring long-term benefits. Furthermore the next question is, will the asphalt mix containing RA and rejuvenator be reusable again at the end of the life?

The performance of materials are already widely investigated and reported [3] [4]. The binder aging and multi-aging is addressed with standard conventional laboratory aging protocol with short term and long term, sometimes even applying double or triple long term aging conditioning [5]. Only a few studies are looking at aging and multi-aging on the asphalt mix itself.

This paper presents and compares results of multirecycling of asphalt mix in laboratory. First, conventional aging conditioning on binder has been conducted to be used as a reference protocol. Secondly, on asphalt mix, made in the lab with 50 % of RA that was subject to aging in oven. Then, this aged asphalt mix was reused again at 50% in a new asphalt mix and then subjected to aging in oven. These results were compared with aging on binder alone. In order to have similar data to compare, only the binder properties, through empirical tests, are discussed either directly from the binders themselves or from binders that were extracted and recovered from the asphalt mixes. They both aim at comparing reference virgin materials with control materials made with RA.

# 2 MATERIALS AND METHODS

## 2.1 Bio-based rejuvenating agent

In the studies, a specific engineered bio-based rejuvenating agent was used [6]. Historically, rejuvenators, such as petroleum flux oils, were used, but worked mostly with high dosage by softening the aged binder through a dilution effect of the asphaltenes. Such an approach did not really help breaking the stronger intermolecular associations of the asphaltenes built through oxidative aging. The specific bio-based rejuvenating agent, is a liquid additive, based on distillate of Tall Oil, that will, with its specific amphipathic chemical structure, disperse the highly polar fractions limiting the agglomeration of asphaltenes. Table 1 presents its main properties.

Table 1. Properties of the rejuvenating agent

| Flash point | Viscosity at 60 °C | Density | Cloud point |
|---|---|---|---|
| > 280 °C | 22 cSt | 0.93 | < -25 °C |

This rejuvenating agent has been already evaluated in research projects [8] on binder [6] and asphalt mixes [7] and as well with field applications [9]. A dosage of 5 % is able to restore by two grades, the flexibility of an aged binder [10].

## 2.2 Bitumen evaluation

In a preliminary evaluation, aging on binder alone was considered as a reference normalised protocol. A virgin bitumen 35/50 was used as a reference binder and compared with an aged binder with and without the rejuvenator. The aged binder was also evaluated as a control binder. The blend of rejuvenator and aged binder was made at 135 °C on a heating plate. The rejuvenator was added at a dosage of 5 % per weight of aged binder. The blend was stirred by hand with a glass rod for 30s and then kept in oven for 15min with stirring every 5 min for 30s, no maturation time was needed.

The three different binders, virgin binder, aged binder and aged binder + rejuvenator were subjected to binder aging. The Rolling Thin Film Oven Test (RTFOT), according to EN 12607, and Pressure Aging Vessel (PAV), according to EN 14769 protocols were used. They correspond respectively to short-term aging, occurring during manufacture, transport and laying, and long-term pavement aging under road conditions [11]. While it is not always recognised as exactly mimicking the real aging conditions, it provides indications on aging behaviour.

All the binders, along the different aging conditioning were characterised with penetration value at 25 °C and softening point temperature as part of the empirical testing defined for EN specifications.

## 2.3 Asphalt mix evaluation

The asphalt mixes, used for lab aging, were coarse mixes made with granite aggregates. The binder contents and gradation curves were maintained equal for all evaluated mixes. The binder content was 5.17 %, equal to the one from the RA source used. Aging in oven was performed on the mix. The binder was later extracted from the mixes according to EN 12697-3. No additional testing on the mix was performed at this point of the study. Four different asphalt mixes were used, Mix I, a control mix with no RA, Mix II, a reference mix with 50 % RA and no additive but with soft bitumen, Mix III, a 50 % RA mix with the rejuvenating agent and Mix IV, a mix made with 50 % of aged RA mix with rejuvenator, which mimic a second cycle of recycling as illustrated in Figure 1.

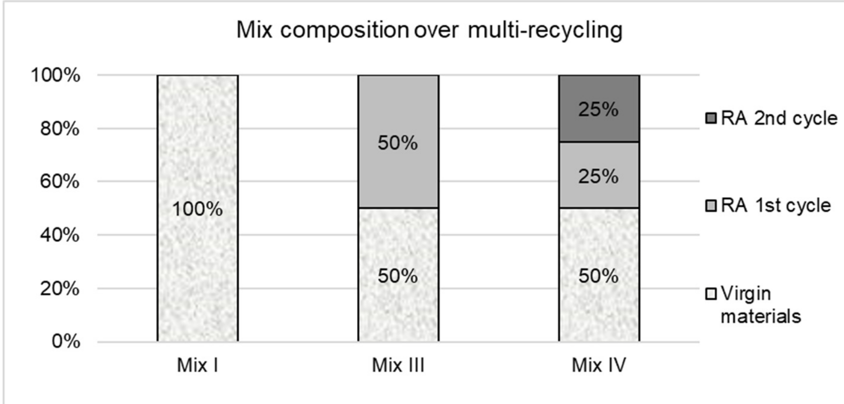



Figure 1. RA Mix composition after multirecycling

Table 2 shows the main properties of the mixes. Each mix was then compacted using the gyratory compactor in the same way, in order to obtain the same air void content for the three different mixes. This void content was initially defined for Mix I after 60 gyrations. The job mix formula used and the granite aggregates lead to a high void content of above 10 %. The other mixes needed fewer gyrations to achieve the same void content. While this high level of voids may not be suitable for standard asphalt mix, for the purpose of the study, it allowed having higher degree of aging through the mixes. Afterwards the binder was extracted and recovered from the mix to be further characterised.

Table 2. Composition of the different asphalt mixes

| | Mix I | Mix II | Mix III | Mix IV |
|---|---|---|---|---|
| | Control mix | Reference 50% RA mix | Mix 50% RA and RP1000 | |
| Virgin aggregates | 94.83 % | 47.42 % | | 47.42 % |
| RA content | 0 % | 50 % | | 50 % of aged mix III |
| Virgin binder* | 5.17 %<br>35/50 binder | 2.59 %<br>70/100 binder | 2.46 %<br>35/50 binder | |
| Rejuvenating agent* | 0 % | | 0.13 % | 0.13 % |
| Gyration and void content | 12.9 % after 60 gyrations | 12.9 % after 16 gyrations | 12.9 % after 15 gyrations | 12.9 % after 47 gyrations |

* per total weight of asphalt mix

For asphalt mix aging in laboratory, different methods can be used [12] [13] either on loose mixes or on compacted mixes. For the purpose of the study, the following protocol based on the Rilem protocol [14], was used, on compacted sample with high void content:

- Original conditioning as before mixing process and aging conditioning
- A first “short-term” aging where the loose mixes were left in an oven at 135 °C for 4 hours
- A “long-term” aging where compacted samples at high void content were left in oven at 85 °C for 9 days
- An “extra long-term” aging with extra 9 days in oven at 85 °C after the long-term conditioning.

All the binders, including the ones from mix extraction, were characterised with penetration value at 25 °C and softening point temperature as part of the empirical testing defined for EN specifications.

## 3 BINDER AGING EVALUATION

For the binder evaluation, the dosage of the rejuvenator was determined to restore the penetration value of the aged binder (AB) similarly to the virgin bitumen and still meeting the minimal softening point temperature specification. While the virgin bitumen was supposed to be graded as a 35/50, it displayed slightly harder value, but still was used for comparison. The aged binder was equivalent to a 10/20 pen grade bitumen, thus a 5 % dosage was used to restore similarly to the virgin reference bitumen. The blend, aged binder + rejuvenator, was able to restore the penetration value between 35 and 50 0.1mm and still meeting the minimal softening point temperature.

The three binders were subject to short-term and long term aging. Table 3 displays the main results. The penetration index (PI) is included as per determined according to EN 12591 Annex A. it provides a useful indication of the temperature susceptibility. The higher the PI value, the lower the variation of properties along with temperature the material is. This is important properties especially for the resilience of pavement towards global warming climate change. As it can be seen for the 35/50, with aging the PI increased. The RA binder PI displayed a value already close to 0, which is a positive effect of aging, and with the additive this PI value is not adversely impacted keeping a good value. It is worth to noticed that after aging the aged and treated aged binders PI changed less than the virgin bitumen 35/50.

Table 3. Binder Aging of binders

| | Unit | Specifications | 35/50 PEN graded | Aged Binder | AB + 5% additive |
|---|---|---|---|---|---|
| | | | Original (not age conditioned) | | |
| PEN | 0.1mm | 35-50 | 29 | 15 | 30 |
| SP | °C | 50-58 | 54 | 67 | 58 |
| PI | | | -1.44 | -0.16 | -0.53 |
| | | | After RTFOT | | |
| PEN | 0.1mm | | 23 | 14 | 27 |
| SP | °C | | 59 | 70 | 61 |
| PI | | | -0.80 | 0.19 | -0.22 |
| Mass change | % | ≤ 0.5 | 0.10 | 0.07 | -0.06 |
| Retained PEN | % | ≥ 50 % | 79 % | 93 % | 90 % |
| Delta SP | °C | ≤ 8 | 5 | 3 | 3 |
| | | | After RTFOT + PAV | | |
| PEN | 0.1mm | | 15 | 11 | 19 |
| SP | °C | | 65 | 76 | 66 |
| PI | | | -0.44 | 0.58 | 0.06 |
| Mass change | % | | 0.51 | 0.42 | 0.46 |
| Retained PEN | % | | 52 % | 73 % | 63 % |
| Delta SP | °C | | 12 | 9 | 8 |

Figure 2 displays the evolution of penetration value at 25 °C on the y-axis vs. the softening point temperature on the x-axis. For each lines, the point on the left end is for original, the middle point after RTFOT and the right end point further with PAV. Results show that the 5 % dosage on the aged binder can restore the penetration value similarly to the virgin binder and still maintaining a high softening point temperature. Over the aging period the aged binder became even harder with a residual penetration value close to 10 0.1mm. A contrary, the aged binder + rejuvenator followed the same trend than the virgin bitumen but with less changes in properties. Would have the effect of the rejuvenator disappeared over the aging, it would have been back to the similar values as the aged binder.

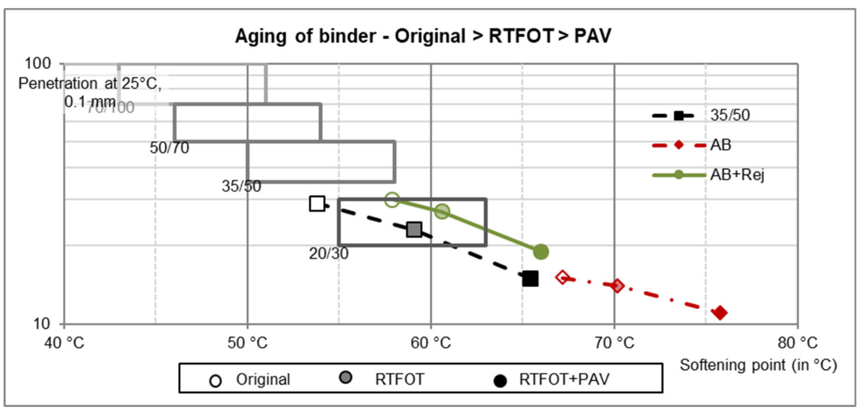
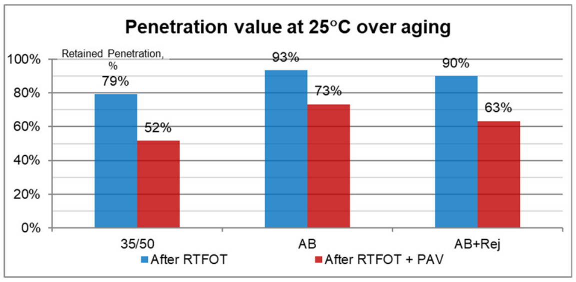



Figure 2. Change in binder properties over aging

Figure 3 shows the relative change, on the penetration values on the left, and softening point temperature on the right. The retained penetration is determined as the decrease of the penetration value in percentage compared to the original value. The retained penetration was greater for the treated aged binder compared to the virgin bitumen, meaning there was less change in property after RTFOT, but also after PAV. The delta of softening point temperature is the difference of aged compared to the original binder. As expected, over aging the softening point temperature increased. Similarly to the penetration value, the change is less for treated aged binder as compared to virgin bitumen.



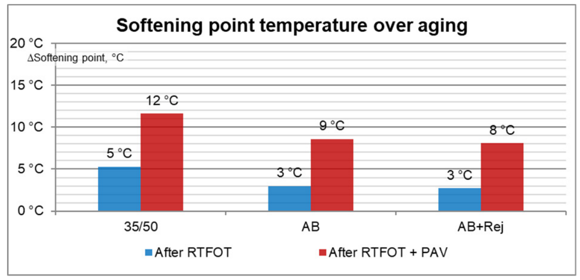




Figure 3. Change in penetration value (left) and softening point temperature (right) over aging

With this first study, the aged binder treated with the rejuvenator had fewer changes in properties compared to the virgin bitumen, meaning they are less prone to age over time.

# 4 ASPHALT MIX AGING EVALUATION

## 4.1 Asphalt mix aging on first recycling cycle

For the mix evaluation, the RA came from a field stockpile and was characterised prior to be used. No tar was detected. The properties of the recovered binder were close to the aged binder used in the binder aging evaluation, with penetration value at 25 °C of 17 0.1mm and softening point temperature of 68.0 °C, in the same range than a 10/20 pen graded binder. The dosage of the rejuvenator was set at 5 % per weight of the RA binder. The RA mixes were manufactured in lab with 50 % RA per weight of final mix. Three mixes were considered at first attempt. Mix I, a virgin asphalt mix with 0 % RA, Mix II, a mix with 50 % RA but no rejuvenator and Mix III, a mix with 50 % RA treated with 5 % of the rejuvenator. The three different mixes were subjected to lab mix aging with short-term aging on loose materials and long-term, and additional extra long-term, aging on compacted samples with high void content. At each stage of the conditioning aging, materials were collected and binder was extracted and recovered from the mixes for further characterisation with penetration value at 25 °C and softening point temperature, amongst others. Table 4 displays the results.

Table 4. Asphalt mix aging with RA and rejuvenating agent

| | Unit | RA | Mix I | Mix II | Mix III |
|---|---|---|---|---|---|
| | | Fresh | | | |
| PEN | 0.1mm | 17 | 40 | 36 | 43 |
| SP | °C | 68.0 | 55.2 | 62.8 | 54.4 |
| PI | | 0.22 | -0.49 | -0.84 | -0.50 |
| | | After short-term aging 4h at 135 °C | | | |
| PEN | 0.1mm | na | 24 | 22 | 24 |
| SP | °C | na | 63.8 | 62.8 | 64.6 |
| PI | | | 0.13 | -0.21 | 0.26 |
| Retained PEN | % | na | 60 % | 61 % | 56 % |
| Delta SP | °C | na | 8.6 | 8.2 | 10.2 |
| | | After long-term aging 9 days at 85 °C | | | |
| PEN | 0.1mm | na | 19 | 20 | 20 |
| SP | °C | na | 69.6 | 63.6 | 66.4 |
| PI | | | 0.63 | -0.24 | 0.22 |
| Retained PEN | % | na | 48 % | 56 % | 47 % |
| Delta SP | °C | na | 14.4 | 9 | 12 |
| | | After extra long-term aging 2x9 days at 85 °C | | | |
| PEN | 0.1mm | na | 17 | 14 | 16 |
| SP | °C | na | 72.4 | 68.8 | 70.8 |
| PI | | | 0.85 | -0.02 | 0.51 |
| Retained PEN | % | na | 43 % | 39 % | 37 % |
| Delta SP | °C | na | 17.2 | 14.2 | 16.4 |

As a first comparison with binder aging, it was observed after short-term aging a significant difference between binder and mix aging protocols. On the virgin binder without any RA, the retained penetration after RTFOT was only 76 %, while, for mix aging, it was 60 %. The same trend is recordable afterwards for the blend and mix containing RA. This suggests that, lab aging on asphalt mix, as set, is more severe than lab binder aging, both for short and long-term aging. This was already noticed in other researches [15].

Figure 4 displays the evolution, on the left, of the penetration value at 25 °C and on the right of the softening point temperature, at the different steps of aging, for the three mixes and with dotted line for the RA and virgin binder.

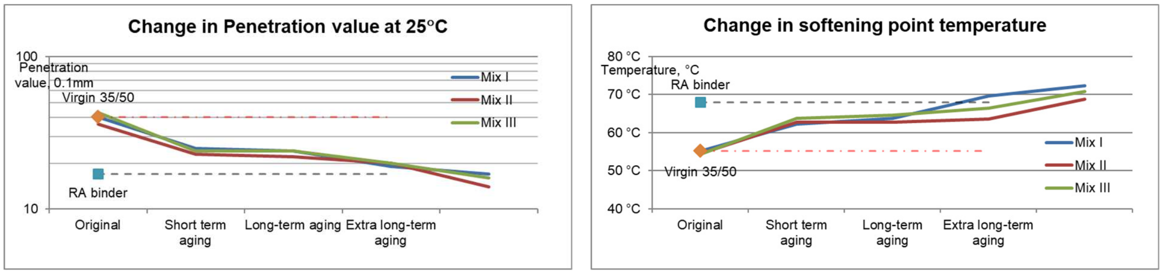



Figure 4. Asphalt mix aging, change in penetration value (left) and softening point temperature (right)

The three curves are following the same trend with hardening over each aging steps, almost overlapping for the penetration value and slight difference for the softening point temperature. Considering the repeatability of the test, for binder recovered from the asphalt mixes, between the control mix with no RA and the mixes with RA there is no significant differences for penetration value and for the softening point temperature there is a slightly more change for the mix with no RA. The effect of the rejuvenator remained over the aging steps even after extra long-term aging and at least did not age more than the virgin mix. It is also worth highlighting that while for the control mix I, with no RA, there was a constant increase in softening point temperature, the mixes with RA displayed limited change between the short and long-term aging. With regards to penetration index, the rejuvenated mix, Mix III, had similar value than the mix with no RA, Mix I, while for mix II it was negatively affected.

### 4.2 Second recycling cycle mix aging evaluation

After this first step of lab asphalt mix aging, the Mix III, after 9 days long-term aging, was reused again at a rate of 50 % in a new asphalt mix, Mix IV. As the residual penetration value at 25 °C was close to the initial RA, the same dosage was applied, 5 % per weight of this new RA binder. **Error! Reference source not found.**~~Figure 1~~ shows the RA composition for the different recycling cycles considered. The second cycle contained 25 % of the initial RA and 25 % of "new" RA as per first recycling cycle. The same could be applied for the rejuvenator considering that the mix III contained already 50 % of 5 % dosage, equal to 2.5 % and was finally treated with additional 5 % dosage.
Then, this mix IV was again subjected to lab mix aging, similarly to the first recycling cycle with short term aging in oven at 135 °C for 4 h and long term aging at 85 °C for 9 days on compacted sample with high void content. At each steps, samples were collected for binder extraction and recovery for all characterisation of the binder. Table 5 compares the results for the multi-recycled Mix IV, virgin Mix I and first recycled Mix III.

Table 5. Asphalt mix aging for multi-recycled asphalt mix

| | Unit | Mix I | Mix III | Mix IV |
|---|---|---|---|---|
| | | | Fresh | |
| PEN | 0.1mm | 40 | 43 | 35 |
| SP | °C | 55.2 | 54.4 | 59.6 |
| PI | | -0.49 | -0.50 | 0.12 |
| | | After short-term aging 4h at 135 °C | | |
| PEN | 0.1mm | 24 | 24 | 23 |
| SP | °C | 63.8 | 64.6 | 69.8 |
| PI | | 0.13 | 0.26 | 1.03 |
| Retained PEN | % | 60 % | 56 % | 66 % |
| Delta SP | °C | 8.6 | 10.2 | 10.2 |
| | | After long-term aging 9 days at 85 °C | | |
| PEN | 0.1mm | 19 | 20 | 21 |
| SP | °C | 69.6 | 66.4 | 70.8 |
| PI | | 0.63 | 0.22 | 1.01 |
| Retained PEN | % | 48 % | 47 % | 60 % |
| Delta SP | °C | 14.4 | 12.0 | 11.2 |

Figure 5 shows the change of the recovered binder after first and second cycle of recycling with rejuvenator. Again the main change in properties occurred during the short term aging as compared to long-term aging, most likely an artefact of mix short term aging being more aggressive than RTFOT or what is really happening at mix plant. Nevertheless, the comparative analysis showed that the mix III, 1st recycling cycle aged similarly or even less that the virgin mix with no

RA and only virgin bitumen. Then, while the second recycling cycle just restored enough the penetration value to meet a 35/50 pen grade, after the full aging short and long term aging the final properties were not worse than the mix I with only virgin bitumen.

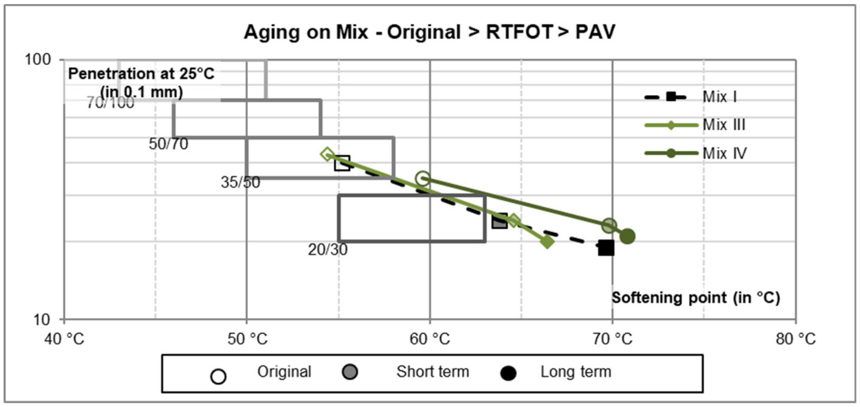



Figure 5. Multirecycling mix aging

## 5 CONCLUSION

While recycling is getting more and more popular due to economic and environmental benefits, the use of rejuvenator is foreseen as it is bringing more value when increasing RA content in the mix, or with very hard RA, or facilitating the processing during asphalt mix production. However, the use of current rejuvenators available today on the market should not harm the quality of materials over time and furthermore the question should be raised, will asphalt mix containing already RA treated with rejuvenator be recyclable at the end of its life?
For this purpose, a specific study on multi-recyclability of asphalt mix made with 50 % RA and specifically designed bio-based rejuvenator has been conducted. It encompassed, a standardised bitumen aging evaluation through binder laboratory short-term and long-term aging. Then aging was duplicated on 50 % RA asphalt mix with specifically lab mix aging protocol. Aged asphalt mix was then recycled again at 50 % RA and compared with a virgin asphalt mix with 0 % RA.
Binder aging was assessed with conventional means for aging, through RTFOT and PAV tests. The comparison with a neat 35/50 bituminous binder, an aged binder and a treated aged binder with the bio-based rejuvenator has shown that restoring the initial properties of aged binder, at least equal to virgin binder, is possible and that this rejuvenating effect remained over aging with fewer changes in properties as compared to the virgin binder.
Asphalt mix aging was conducted on loose materials for short-term aging in oven at 135 °C for 4 hours and compacted samples with high void content for long-term and extra-long term aging in oven at 85 °C for 9 days and also two 9 day cycles respectively. As a first comparison between binder and asphalt mix aging, the mix short term aging was more severe than the binder RTFOT aging. Then, the comparison with an asphalt mix made with only virgin materials and a mix made with 50 % of RA has shown a similar trend. It is possible to restore, at least to the same level of the virgin mix, the binder as extracted from the mixes and there were fewer changes in properties for mixes with RA even with extra long-term aging.
Finally a second 50 % recycling cycle with the same bio-based rejuvenator did show even less change of binder properties as compared to virgin mix. It demonstrated that the use of the bio-base rejuvenator did not impact the future recyclability of asphalt mix containing high RA content.

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