Internal structure characteristics of crumb rubber modified asphalt binders: An analysis using 3D X-ray Microtomography Imaging

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ABSTRACT. The performance of crumb rubber modified asphalt pavements (CRMAPs) depends on the mix design and aggregate gradation. Even though numerous success stories about the CRMAPs have been reported in the literature, inadequate design can lead to less than desired performance. Understanding the factors contributing to the improved performance CRMAPs over unmodified asphalt pavements is important for developing appropriate designs and also for possibly expanding the limits of aggregate gradation. The objective of this study is to quantify the internal structure of the crumb rubber modified asphalt binders (CRMABs) made with the wet process. 3D geometry of crumb rubber (CR) particles was directly measured and quantified using a high-resolution synchrotron based X-Ray Microtomography (XRM) imaging system. The CRMAB samples were instantly frozen using liquid nitrogen and kept frozen until processing. The 3D internal structure images of these specimens were acquired using the XRM available at Argonne National Laboratory (ANL). These images were analyzed and the change in the size of particles was directly quantified. It was observed that the mixing process causes the crumb rubber particles to partially melt and separate to create small CR ‘chips’. These small CR ‘chips’ homogeneously mix with the asphalt binder creating a polymer-like structure, which improves their engineering (e.g., fatigue) performance.

KEYWORDS: X-ray Microtomography, Wet Process, swelling, image processing
1. Introduction

Crumb rubber has been used as additive in asphalt pavements since 1950s [1, 2, 3]. Benefits of crumb rubber modified asphalt pavements (CRMAPs) have been acknowledged by numerous researchers [4, 5, 6]. Most CRMAPs are made with the following three major methods: wet process, dry process, and terminal blend process. In wet process, crumb rubber (CR) is added to liquid asphalt at temperatures around 325-400 °F (163-205 ºC) and about 15% - 22% by weight of the binder is utilized (1-1.5% by total weight of the mix) [13, 14]. Dry process is the method where the CR particles are added to the mix as a replacement of fine aggregates. The terminally blend process is similar to the wet process, except that less amount of CR is used (~10-12%) and a polymeric additive is used to help suspend the CR particles in the binder.

The characteristics of crumb rubber modified asphalt binders (CRMABs) depend on the rubber, binder type, size of the CR particles, duration and temperature of the reaction and modification method [7,8]. The CR particles mainly react in two different ways during modification process: swelling and/or degradation [9]. Depending on the reaction time and temperature, CR particles swell approximately two to three times of their original size. If the reaction time and temperature are ascended, the particles degrade into the binder. Therefore, the binder properties drastically change. (e.g., viscosity significantly drops with degradation). Although the swelling of particles is proven, the mechanism has not been clearly quantified yet. Xiao et al. (2006) studied the size change of CR particles with respect to the rubber gradation, binder type and reaction time. The variation is evaluated by extracting the binder from CR wet modified binder and by comparing the extracted CR particles with the original ones. However, this method is not adequate enough to understand the swelling behavior since some of the particles might have degraded during extraction [10]. Miknis and Michon (1997) used Nuclear magnetic resonance (NMR) imaging methods to prove the swelling of CR particles in the presence of the hot binder [11]. Airey et al. (2007) studied the binder absorption of CR particles by using Basket Drainage Method. The mass increase of the CR particles was used to evaluate the absorption. This study stated that the absorption rate is mainly controlled by the nature of CR. Besides, it was noted that binder properties such as chemical composition and viscosity directly influence the absorption. [12] However, none of the methods clearly quantifies the swelling and degradation mechanisms.

The objective of this study is to quantify the CR particle sizes change after the wet process modification. X-ray Microtomography (MT), which is a radiographic non-destructive imaging technique, is utilized. [15] Change in the overall volume of the CR particles as well as the size distribution of the particles was computed using 3D image processing methods.
2. Materials

In this study, wet process method was utilized to modify the binders. The base binder was PG 64-22 and CR was blended 15% by weight of binder. The gradation of the CR is shown in Table 1. The base binder was heated up to 190°C and mixed with CR particles with the aid of a digital dual-range mixer at a rate of 2000±100 rpm (rotation per minute) for 60 ± 5 minutes. During the mixing process, the mixing bucket was placed in bucket heater to keep constant mixing temperature. The blade size was selected to be 1/3 of the of bucket size. Thus, CR particles were homogeneously distributed within the binder, with all CR clumps eliminated in the mixture.

<table>
<thead>
<tr>
<th>Sieve No. (mm)</th>
<th>Percent Passing (%)</th>
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<tbody>
<tr>
<td>No. 16 (1.90)</td>
<td>100</td>
</tr>
<tr>
<td>No. 30 (0.590)</td>
<td>100</td>
</tr>
<tr>
<td>No. 40 (0.420)</td>
<td>94</td>
</tr>
<tr>
<td>No. 100 (0.149)</td>
<td>16</td>
</tr>
<tr>
<td>No. 200 (0.074)</td>
<td>2</td>
</tr>
</tbody>
</table>

Table 1. Size distribution of crumb rubber used in this study.

After the binder was modified, it was poured into 7 mm diameter polypropylene micro-centrifuge tubes. After pouring, each specimen was instantly frozen using liquid nitrogen. The purpose was to monitor the change in the microstructure of the CR particle sizes and the samples were kept frozen till it was processed. Kutay and Ozturk (2012) proved that the instant freezing procedure did not damage the binder specimens [16]. This observation was also validated when the 3D XRM images of CR modified samples were analyzed, where there were no visible cracks within the specimens.

3. 3D imaging using X-ray Microtomography

The internal structure of crumb rubber modified binders was obtained by X-ray Microtomography (MT) technique, which is a radiographic non-destructive imaging technique to produce 3D images of the materials [14]. The images of the binder samples were obtained at the 5-BM-C Microtomography beam line at the Advanced Photon Source (APS) facility in Argonne National Laboratory (ANL). The MT system is basically composed of X-ray source (i.e., the X-ray beampipe in Figure 1) and a detector. The specimen is vertically placed between the source and the detector. The detector records the X-ray intensities that is released by the source
and passed through the specimen. The detected intensities are used to calculate the linear attenuation coefficients within the specimen. The resulting 3D image is stored as a series of 2D slices, which are then converted into grayscale images. Each grayscale image represents a slice of the specimen at certain height. The intensity of grayscale image is stored with 8 bits per each pixel, which allows 256 different intensities. When the pixel value is 255, the result is pure white; when it is 0, the result is pure black.

Figure 1. Picture of the Synchrotron-based X-ray Microtomography setup

The detector of MT system composes of cryo-cooled CCD system and optics. The spatial resolution of the images depends on the size of the specimen and varies from 3 microns to about 100 microns. The required energy of the X-ray beamline depends on the sample size and density. For instance, thick and high-absorbing samples should be scanned in higher energy levels. In this study, the energy level was determined to be 20keV, which provided a volume scan of 7 mm diameter, 7 mm tall cylinders. The resulting 3D image of the sample was stored as 1299 2D slices. The size of each slice was 1299 by 1299 pixels, resulting in a 3D image matrix with size 1299 by 1299 by 1299 voxels (3D version of pixel). The image resolution (i.e., size of one side of one voxel) can be calculated as $\Delta x = \Delta y = \Delta z = 7 \text{ mm} / 1299 \text{ pixels} = 0.0054 \text{ mm/voxel} \ (5.4 \text{ micron/voxel})$. As shown in Table 1, ~98% of the crumb rubber particles were larger than 0.074 mm (74 microns). As a result, the minimum size of the CR particles was about 14 times larger than the resolution of the image, allowing a detailed image of CR particles. Also, maximum size of the CR particles (~0.59 mm) was about 1/12 of the size of the 7 mm diameter container.

4. Image Analysis Methods

The analysis of 3D XRM images was challenging since the CR particles and binder were in the same intensity range. Additionally, ring and beam hardening
artifacts seriously degraded the quality of XRM images. In order to get rid of the beam hardening artifact, the images were cropped. The image size was decreased to 650 by 650 by 650 pixels. Then, a Gaussian low pass filter was implemented to the cropped images to remove the ring artifacts and other noise. The cleaned images were then converted to binary (black/white) images using a thresholding algorithm. Then, a connected components algorithm was utilized to label individual CR particles. After each particle was labeled, the volumes and equivalent diameters (i.e., the diameter of a sphere of equivalent volume) of the particle were computed. The volume and size distribution of the particles were computed using algorithms developed in Matlab®. Image processing methods utilized in this study was similar to those detailed in Kutay et al. 2010 [17].

5. Analysis of Results

Figure 2 shows a 2D slice of obtained from the X-ray MT technology. The grayscale intensities in the image correspond to the relative density (i.e., specific gravity) of the materials. Typically the specific gravity of crumb rubber (Gs(CR) = 1.1 to 1.2) is larger than that of the asphalt binder (Gs(asphalt) ~ 1.025). Therefore, the CR particles have brighter color intensity in the X-ray MT image shown in Figure 2. Figure 2 also shows that some of the CR particles have much higher brightness (i.e., density) than others. This is probably because of several types of CR used while preparing the CR. Another observation made while looking at all the X-ray MT slices was that there were very high-intensity (almost pure white) zones in the image. These areas probably correspond to metal chips leftover from the waste tires. Also, some of these bright zones seem to be spread within the asphalt binder. This may be because the metal chips were separated from the CR particles during the 1 hr mixing period at high-temperature (i.e., 190°C). It can also be noticed from Figure 2 that there are very small CR particles (relatively low density – intensity-areas) spread inside the binder, also indicating that high-temperature mixing separates small CR chips from the main CR particles and homogeneously mix into the binder. This phenomenon seems to create a micro-level ‘polymer-like’ structure inside the binder. This structure perhaps helps improve the binder’s resistance to several distresses such as fatigue cracking.
Figure 2. X-ray MT-based 2D slice image of the CR particles in the asphalt binder prepared using the Wet Process.

Figure 3 shows the 3D visualization of the crumb rubber particles in the asphalt binder. As shown, there are many small crumb rubber “chips” in-between the main CR particles, supporting the statement made earlier about the separation of the main CR particles into small pieces during mixing. Such structure is almost like an actual asphalt mixture creating a network of particles where stresses can be transferred. Also, these particles can act as a ‘stopper’ for the micro-cracks developing during fatigue loading.
Figure 3. 3D visualization of the CR particles in the asphalt binder prepared using the Wet Process.

In order to better quantify the properties of the CR shown in Figure 3, image analysis algorithms were utilized to compute the overall volume as well as the size distribution of the CR particles. Table 2 shows the original properties of the CR modified binder where, as mentioned earlier, the CR content (PW) was 15% by weight of the binder. Given that \( PW = \frac{W_{CR}}{W_{binder}} \), where \( W_{CR} \) = weight of CR and \( W_{binder} \) = weight of the asphalt binder, the volumetric percentage of the CR (i.e. PV) can be computed as \( PV = \frac{PW \times G_{s-binder}}{G_{s-crumb rubber}} \), where the \( G_{s-binder} \) is the asphalt binder specific gravity and \( G_{s-crumb rubber} \) is the crumb rubber specific gravity. As shown in Table 2, the original volumetric percentage of the CR was 13.5%. Image-based volumetric percentage of the CR particles was computed by using a basic image-analysis method, where the number of white voxels (which corresponds to CR particles) in the thresholded black/white 3D image are counted and multiplied by the image resolution. As shown in Table 2, the image-based volumetric percentage of the CR particles is 12%, which is less than 13.5%, the original value. The fact that image-based PV is less than the original PV does not necessarily mean that the CR particles did not expand (or shrank) during mixing. Most probably, a portion of the CR have melted and homogeneously distributed into the asphalt binder. This phenomenon is supported by the small “chips” of CR shown in Figures
Based on the original gradation of the CR measured in the lab, there shouldn’t be any CR particles less than 0.075 mm in size. However, as shown in Figure 4, image-based CR gradation indicates that there is ~5% of the particles are less than 0.075 mm in size. This further supports that the CR particles separated into smaller pieces that homogeneously spread into the binder, improving its engineering properties.

<table>
<thead>
<tr>
<th>Original values</th>
<th>Image- based measurement</th>
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<tbody>
<tr>
<td>$G_{s\text{-crumb rubber}}$ (Rubber specific gravity)</td>
<td>1.135</td>
</tr>
<tr>
<td>$G_{s\text{-binder}}$ (Binder specific gravity)</td>
<td>1.025</td>
</tr>
<tr>
<td>PW (%CR by weight)</td>
<td>15.0%</td>
</tr>
<tr>
<td>$PV$ (%CR by volume)</td>
<td>12%</td>
</tr>
</tbody>
</table>

Table 2. Comparison of original and image-based volumetric percentage of crumb rubber in the asphalt binder

It should be noted that Figure 4 also shows that the image-based gradation is slightly coarser than the original gradation. This is the result of inaccuracies related to the automatic particle separation algorithm, where not all the particles were separated successfully in the image. As a result, some of the CR particles that are very close to each other were classified as a single CR as shown by the sub-plot in Figure 4. If the particle separation algorithm were to be 100% successful in separating all the particles in the image, the image-based gradation would be the same or slightly finer than the original gradation.
6. Conclusions

Internal structure of a crumb rubber modified binder prepared using wet process was analyzed using a non-destructive 3D imaging technique called X-ray Microtomography. After mixing, the wet process binder was poured into 7 mm diameter polypropylene micro-centrifuge tube and instantly frozen using liquid nitrogen to preserve the internal structure. Then the sample was scanned using 5-BM-C Microtomography beam line at the Advanced Photon Source (APS) facility located in Argonne National Laboratory (ANL). Once the images were obtained, 3D image analysis methods were utilized to compute the change in the overall volume of the CR particles as well as their size distribution. It was observed that the mixing process causes the crumb rubber particles to partially melt and separate to create small CR ‘chips’. These small CR ‘chips’ homogeneously mix with the asphalt binder creating a polymer-like structure, which improves their engineering (e.g., fatigue) performance. While the original CR gradation had only 2% passing #200 sieve, the image analysis results revealed a gradation that had ~6% passing #200 sieve size.

7. Acknowledgements

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8. Bibliography

Image Analysis of CR Wet Process Binder using X-ray Microtomography


