

First Observation of the Blending Zone Morphology at the Interface of Reclaimed Asphalt Binder and Virgin Bitumen

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ABSTRACT

One of the challenges in designing recycled asphalt mixtures with a high amount of RAP is to estimate the blending degree between RAP binder and the added virgin bitumen. The extent of blending is crucial in this case as asphalt concrete response is influenced by the final binder properties. This paper focuses on the evaluation of interaction and extent of blending between RAP-binder and virgin bitumen by studying the microstructures of the 'blending zone' with atomic force microscopy (AFM). AFM is used to probe the change of microstructural properties from a RAP-binder and virgin bitumen to the blending zone of these two. Averaged microstructural properties have been observed in thin film blends of RAP-binder and pure bitumen. The morphology of the blending zone (spatial extent of about 50 μm) exhibits domains of a wide range of microstructure sizes from 160 nm to 2.07 μm and can be considered to be a completely blended 'new material' which has been observed directly for the first time. The fully blended binder properties are found to be in between those of the two individual binders, as could be inferred from the averaged microstructural properties as derived from AFM-images of the blending zone. This is also consistent with the results of mechanical tests by dynamic shear rheometer on the same materials. Finally a design formula is proposed that relates the spatial dimensions of the blending zone to temperature and mixing time. This relation will eventually allow for translating the results of this study from small length scales up to the engineering level.

INTRODUCTION

The use of reclaimed asphalt pavement (RAP) as a recycling component in new hot mix asphalts (HMA) is common practice nowadays. From environmental and economic perspective the use of RAP as an ingredient in new asphalt mixes is a very attractive development. It reduces the problem of waste disposal of road construction materials, and also reduces the use of scarce raw materials. For example, in the Netherlands approximately 4 million tons of reclaimed asphalt became available in 2010 of which approximately 75% was recycled into new hot mix asphalt (1). Today, the asphalt industry is facing the challenge to increase the RAP content to 70% without compromising the mechanical and other desired properties of the asphalt structure (currently in the Netherlands a maximum RAP content of 50% is allowed, and only 20% in the surfacing of porous asphalt,). Meanwhile there exists a growing need to increase the amount of RAP in porous asphalt concrete mixtures, as in the near future major maintenance works on highway pavements will be carried out, which leads to an increased availability of porous asphalt concrete derived RAP. Similar developments are seen all over Europe, in the USA (2), and Asia.

Crucial for the use of high quantities of RAP within asphalt mixtures for new pavement structures is the requirement that the overall performance of the system should be at least at the level of the material without RAP added. This is a severe constraint, especially because the bituminous binder in the RAP has aged, thus has chemically changed, a process that is not anticipated to reverse by only mixing it with a virgin – i.e. non-recycled - material. Therefore significant research efforts have been directed towards the performance of asphalt mixtures as a function of RAP content (for a literature review see (3)). The main performance characteristics that are considered in these studies are fatigue, rutting, low temperature cracking and raveling.

It has been claimed that addition of up to 50% RAP leads to an increase of elastic modulus, tensile strength and improved rutting resistance of the mixtures (4). Hence asphalt concrete mixtures containing RAP perform equally or better than mixtures with only virgin aggregates (4,5). Other authors found that up to addition of 15% RAP no effect on low temperature (cracking) behavior can be observed (6). Field studies show similar trends: application of up to 30% RAP in mixtures showed equal or better performance than virgin mixes, though it was noted that the material performed worse with respect to fatigue (7). In another field study (7,8) it was found that warm-mix asphalt (WMA) containing RAP is softer (lower dynamic modulus) than a HMA containing the same amount of RAP. The authors attributed this to incomplete blending of the RAP and virgin binders.

Thus two issues come up in relation to the recycling of asphalt that needs further investigation: the fatigue behavior of RAP-mixes, the degree of blending of RAP and virgin binders, and how this would affect the performance of the mixtures. To avoid premature failure of HMA due to fatigue, recycling requires to soften the aged and stiff RAP binder by adding softer virgin bitumen or a rejuvenator. In this way the fatigue characteristics of RAP-mixtures may relate directly to the degree of blending. However, there is a lack of knowledge on the degree of blending of the RAP-binder with virgin bitumen. To determine the blending degree of virgin bitumen in recycled asphalt mixtures, blending charts and equations such as the log-pen rule are used (2,9,10). In this design method it is assumed that RAP binder is homogeneously and completely blended with virgin bitumen during the mixing process (5): the so-called linear blending scheme. Other ways of estimating the degree of blending of RAP and virgin binder are based on the mechanical response of the RAP-

mixture (stiffness as a function of percentage RAP, viscosity, BBR, cracking and rutting parameters), which is then interpreted with an appropriate model (11).

Thus, the assumption of a homogeneously blended mix is seldom valid as concluded by the laboratory and field studies mentioned before. On the contrary, some designers consider RAP as a black rock and assume no blending at all. These two extreme assumptions can eventually lead to the wrong estimation of design parameters (12,13,14). Moreover it is still unresolved whether a homogeneous or a partial blending of the binders are required for an improved fatigue life. Thus it is necessary to quantify the degree of blending between RAP binder and virgin bitumen to setup a correct mix-design approach.

To study the details of the blending process and the blending result of mixing a neat and RAP derived binder, new nanotechnology based methodologies will be explored. Direct observation of RAP binder and virgin bitumen is one of the several techniques which has been used in studying the blending degree. Among those methods, using nano-indentation was yet not successful in measuring and characterizing the blending zone (15). In this contribution atomic force microscopy (AFM) is being explored to gain insight in the blending of RAP and virgin bitumen.

It has been known for a long time that bitumen exhibit unique properties at the micron and nanometer length scales. AFM-images of bitumen show that a rich variety of microstructure can develop on its surface (16,17,18,19). The observed microstructure can develop to different degrees and in different microstructural details depending on the crude origin of the bitumen, the thermal history of the sample, etc. The microstructure has earlier been proven to be a unique and reproducible property of a bitumen (19,20), therefore it is a tool worth exploring in RAP research.

The specific details of the microstructure are believed to originate from bitumen chemistry: a variety of intermolecular associations driven by molecular polarity, size or shape could explain the observed structuring at the micro meter scale. Changes in the molecular composition or ordering, or both, will result in significant changes in the microstructural properties. Furthermore, also chemo-mechanical and structural properties of the material will relate to details of the microstructure (21,22), which makes AFM a versatile research tool for a material as complex as bitumen. Within the research that is reported here, the AFM-derived microstructure of bitumen is used as a probe for the degree of blending of the RAP and virgin binders. Here it is assumed that the surface microstructural properties represent also the bulk material properties.

One of the dynamic modes of AFM, tapping-mode (23), is used to characterize surface microstructural morphology and to evaluate the interaction and extent of blending between the aged RAP-binder and virgin bitumen. It is known that microstructure strongly relates to a material's macroscopic mechanical response. Spatial variations in local material properties observed with AFM will also affect the material's macroscopic mechanical properties. The goal of this study is to image and understand the extent of blending and its transitional mechanical properties. It is expected that this can be used to establish improved and standardized mixing procedures for RAP and virgin binders. Eventually, this may lead to new reliable design parameters to enhance the durability of the recycled pavement materials. Finally it should be noticed that the present study only considers a better understanding of the degree of blending on the binder scale for one particular type of virgin binder. The effect of the choice of virgin binder and the impact of the extent of blending on the mechanical properties at the mastic and asphalt concrete level will be subject to further study.

MATERIALS & METHODS

Preparation And Rheology of the Binders

To derive bitumen from RAP common solvent extraction (EN 12697-1) and rotatory evaporation (EN 12697-3) methods were utilized. In this research, methylene chloride has been used as the solvent. The bitumen recovered from the RAP had a penetration of 21 (at 25 °C), a softening point of 60 °C and a mass density of 1.035 g.cm⁻³. A soft binder (penetration grade 160-220) was selected to represent virgin bitumen in the blending process. For this bitumen the penetration (measured in 0.1 mm units) was 144 (at 25 °C), the softening point of 43 °C and mass density of 1.020 g.cm⁻³. The rheological properties of these two binders, as well as their mixture, were characterized with a dynamic shear rheometer (DSR). To perform the DSR test, 100 gram of the RAP binder and the same amount of virgin bitumen (1:1 ratio) were blended for 5 minutes at 160 °C to achieve a homogenously blended bitumen. This blended binder was used to characterize the rheological properties in comparison with the other binders. The rheological properties of the three binders were characterized with DSR. G* and phase angles were shifted to 25 °C to obtain master curves as shown in Figure 1. From the DSR data one can observe that the rheological properties of the blend is in between those of the binder recovered from RAP and the virgin binder. However, only based on these rheology data it is difficult to draw conclusions about the extent of the blending process.

Imaging with Atomic Force Microscopy

Atomic force microscopy (AFM) is a scanning probe technique that allows to reveal surface topography and heterogeneity of materials with high spatial resolution (24,25,26,27). In AFM imaging, a cantilever with an extremely sharp tip (nominal tip radius of 8 nm) located on its free end is scanned over the sample surface utilizing a piezoelectric scanner (see Figure 2). The changes in tip-sample interactions result in deflection of the cantilever which is measured by an optical-lever detection system. In this system a laser beam is focused onto the back side of the cantilever and the reflected beam is detected with a position sensitive photodiode. While scanning, a specific operating parameter is kept constant by a feedback loop between the optical detection system and the piezoelectric scanners. Measurements are being recorded electronically. The data acquired build up a map of the surface topography, which is representative of variations in the tip sample interaction.

Tapping-mode AFM (23) was used to characterize RAP binder, virgin bitumen and their blending zone. In the tapping-mode the probe is modulated near its first resonant frequency while it is scanned across the sample. Thus the tip maintains an intermittent contact over the sample surface, keeping the tapping force low and the lateral forces negligible. This moderate force exerted on the surface leads to scanning in a non-invasive manner, which is ideal for soft material surfaces such as bitumen (23,26).

Since the probe is oscillating, it experiences attractive and repulsive forces depending on its position in the cycle. As the tip approaches the sample, the tip-sample interactions alter the amplitude, resonance frequency, and phase angle of the oscillating cantilever. During scanning, the amplitude at the operating frequency is maintained at a constant level, called the set-point amplitude, by adjusting the relative position of the tip with respect to the sample.

The oscillating cantilever dissipates various amounts of energy as it interacts with material heterogeneities on the sample surface. In tapping-mode the instrument provides three different types of data simultaneously: topography, phase-contrast, and amplitude error. Each of the image types provides specific information with respect to the sample surface. Topography images provide information of relative height of the various features as the probe tip is raster scanned across the sample surface. Phase Imaging creates images of the phase of the tapping response, which is a function of the forces that the tip is experiencing; in other words the relative damping of an oscillating cantilever tip as it experiences heterogeneity in material response of the surface. Error images provide a record of any variations from constant deflection of the cantilever as the tip is scanned over the surface, which represents the high frequency contrast of the topography signal indicating areas where topography is changing rapidly. All of these image types are obtained simultaneously and need all to be considered for the interpretation of tapping-mode AFM results.

AFM Instrumental Settings, Sample Preparation and Measuring Environment

The 'Multimode-V Atomic Force Microscope' from Bruker (Santa Barbara, USA) was used for this study. Steel sample disks (12 mm) were used as sample substrates for the AFM-measurements. Commercially available High-Resolution Tapping-Mode silicon cantilevers 'RTESPA' (Bruker) were used for tapping-mode AFM. These cantilevers have a nominal resonant frequency of 330 kHz and a nominal force constant of 40 N/m. The cantilever material is Antimony (n) doped Silicon which has 40 ± 10 nm aluminum reflective coating on its backside. The reflective coating helps to increase the laser signal as well as protects the bitumen sample from softening by laser exposure. The nominal cantilever dimensions were $120\times 35\times 3$ μm . The cantilever has a chemically etched rotated silicon tip on its edge to provide a more symmetric representation of features over 200 nm. The tip height is in the range of 15-20 μm and it has nominal tip radius of 8 nm.

For compositional imaging of a multiphase material like bitumen the most pronounced phase contrast is usually achieved at hard tapping. The probe scan rate was chosen to be 1.0 Hz (1 Line/s) and morphological details were recorded at 30×30 μm scan size with a pixel resolution of 512×512 . The study of the microstructure morphology of RAP-binder, virgin bitumen and their blending zone was performed at ambient conditions and in air.

The pure bitumen samples for the study with AFM were prepared by applying a film of liquid bitumen of 100 °C on a 12 mm metal sample disk. The film thickness was calculated by measuring the weights of the sample disks and using the known mass density of the materials. The layer thickness was found to be 0.4 ± 0.05 mm, so the thickness is about a hundred times larger than a typical microstructural dimension of the bitumen. The specimen for studying the blending of RAP and virgin bitumen was prepared by applying a bead of 15 mg RAP-binder by a spatula on one side of the 12 mm metal sample substrate (0.5 mm thickness) and the same amount of virgin bitumen from the other side. Then the specimen was heated for 40 seconds at 130 °C on a heater plate. In this case the heater plate conductively heated the steel substrate and thus the sample attached to it. The temperature and the heating time were sufficient to melt the bitumen from both sides, which allowed to form two drops of different bitumen to spread on the surface creating a thin film with a fused interfacial zone in the middle. At a constant temperature (130 °C) the rate of flow of soft-virgin bitumen was relatively faster than that of the RAP binder, but this didn't lead notable difference (observed macroscopically) in film thickness between these two zones. While probing with AFM, the

thickness difference at the fused interfacial zone was found to be lower than 100 nm. Prior to AFM imaging, the samples were allowed to equilibrate at the imaging temperature of 25°C for 24 hours.

EXPERIMENTAL RESULTS

The surface morphology of the thin film bituminous materials were characterized by AFM for samples prepared according to the procedure described above. A basic tapping-mode AFM experiment measures three datasets at a time: i) the height image measures the surface topography of the bitumen surface, ii) the phase image measures a mapping of the local mechanical properties of the surface, hence it will discriminate between regions with distinct mechanical properties (stiffness, adhesion etc.), thus different ‘materials’. Finally the iii) amplitude error signal detects high frequency features in the topography of the image. The latter will not be considered in the current study, because the features of interest are in the order of micrometers (spatial resolution along surface is about 50 nanometers and in the order of several nanometers in the z-direction perpendicular to the bitumen surface), which is much larger than the length scale monitored by the amplitude error signal. The scan speed (1.0 Hz) over the surface was slow enough to resolve all features of interest.

Qualitative AFM Results

The AFM images obtained for a representative (at least for the virgin and RAP binders) surface scan of 30×30 μm for the virgin bitumen, the RAP and the interfacial zone between the RAP and virgin binder are shown in Figure 3. The virgin and RAP binder images are from separately prepared samples. The interface between virgin and RAP binder has been found with a range of successive scans along a line perpendicular to the presumed interface between RAP and virgin binder, see Figure 4. The interface between the two binders could be identified by following the gradual change of microstructure along the scan-line (Figure 4). Initially the microstructure was found resembling qualitatively the images in Figure 3b, while at the end of the scanning line the microstructure looked very similar to those in Figure 3a.

There was just a single image that clearly showed a very distinct pattern, as in Figure 3c, which also happened to be in the middle part of the sample disc. Altogether this provides enough evidence to confidently identify Figure 3c as *the interfacial blending zone between RAP and virgin binder, observed here for the first time directly*.

Some immediate observations these images (Figures 3 and 4) are the following:

- All bitumen show a microstructure which is different;
- The topography and phase images are congruent, i.e. that clusters of different ‘material’ (phase) differ in topography as well;
- The microstructure consists of domains with a slightly elongated, oval shape (elliptical in first approximation), with height oscillations perpendicular to the surface and along the long axis of the microstructural unit (the phase outside these domains is referred to as continuous phase);
- The elliptical domains (white areas in the phase images) are the largest for the virgin binder and smallest for the RAP. The blending zone is more polydisperse and somehow intermediate to the two;

- The surface coverage by elliptical domains is largest for the virgin bitumen, i.e. the phase fraction of the continuous phase is smallest here.

Quantitative Comparison of the Microstructures

Comparing the observed morphologies between three different regions (Figure 4), provides information about the nature of the blending process. The software package Gwyddion (28) was used to derive objective measures for the size distribution, shape and phase fraction of elliptical domains, which together characterize the microstructure in a quantitative way. Statistical analysis was used on the distinct quantities, where N will signify the number of observations. In order to distinguish the microstructures from an amorphous background, images were converted to binary images and from that phase fractions were calculated; these are consistent with other ways of calculating the phase fraction (ellipsoid approximation).

The topography and phase images of pure bitumen show microstructures that in the approximation of ellipsoidal shape possess long axes that range from 460 nm to 4.77 μm , whereas this range for the RAP-binder is from 70 nm to 1.71 μm and for the blending zone 160 nm to 2.07 μm . Other quantities that could be derived were the short axes, surface coverage (or phase fraction ϕ) and the aspect ratio A (or eccentricity $e = 1 - A^2$) of the short to long axis. The closer the aspect ratio is to unity, the more the elliptic domain approaches a perfect circular shape, whereas an aspect ratio approaching zero signifies a needle-like shape.

The microstructural sizes (lengths of longer axis) were obtained from the AFM phase images of the three different binder regions and statistical quantities were derived from these lengths. Typical distributions that quantify the microstructure are presented in Figure 5. As mentioned earlier, the height profile shows regular topographical variations along the longer axis which reveals that micro-structural features exhibit a wrinkling pattern along that axis. Some examples of this are shown in Figure 6. The main features of the wrinkling profile are the wavelength λ and the maximum amplitude. The wavelength appears to be a very constant quantity for each bitumen type. This has been proven by connecting for each binder all profiles, and taking the Fourier transform of the connected profiles: a single reasonably sharp peak appears in the Fourier transform, which proves the constancy of the wrinkling wavelength. For virgin bitumen the wavelength is found to be around 0.55 μm (amplitude 16 nm), whereas it is around 0.30 μm (amplitude 11 nm) for RAP and 0.4 μm (amplitude 18 nm) in the interfacial mixing zone. All results derived from extensive statistical image analysis are summarized in Table 1. It should be noted that the (maximum) amplitude has a rather large uncertainty, reflecting the spread in measured amplitudes. Though, when plotting the amplitude results for a single material one observes a statistical spread, but the differences between individual amplitudes are very close to an integer multiple of a characteristic distance, a number between 4 and 6 nanometer. This is a remarkable finding which may relate to the molecular make-up of the microstructural features. It is too large for single molecular features, but could reflect an ordered (crystalline) structure with a typical repeat distance (lattice parameter) of 4-6 nanometer. This distance is in the range of the two smallest lattice parameters of typical paraffin (alkane) waxes, which are found to be between 5 and 8 nanometers (29). This is also in agreement with the hypothesis that the elliptical domains present in the microstructure of bitumen possess crystalline features that can be identified with wax (30).

DISCUSSION AND CONCLUSIONS

The case of mixing and blending of a soft virgin binder with a RAP derived binder has been studied by means of following the material's microstructure with AFM. The original binders display very distinct surface morphology and could thus be identified by their microstructure. The aspect ratios, sizes, areal coverage and profiles of the elliptical domains, that characterize the microstructure, have been derived by statistical means from the AFM-images, Table 1. From this it is concluded that these parameters can be considered as a fingerprint of the distinct bitumen grades. The presence of a blending zone at the interface of the two bitumen grades was found by consecutively recording AFM images in the lateral direction of the sample surface (Figure 4), while observing the change in microstructure. It is the first time that in this way the interfacial zone, where blending between two bitumen grades occurs, is observed in a direct way. The extent of the blending zone, d (Figure 7a), is in the order of tens of micrometers (estimated 50 μm). Thus, bringing two binders of very different stiffness (Pen-grade) into contact at about 130 $^{\circ}\text{C}$ will lead to a blending zone with a new microstructure; i.e. if two aggregates with a bitumen film of about 25 μm are brought into contact at 130 $^{\circ}\text{C}$ all material at both sides of the interface will be 'consumed' to form a new uniform phase that is termed as the blending zone. In this scenario one may speak of 'complete blending'. The extent of the blending zone, d , will most likely depend on parameters such as temperature, contact time and type of bitumen at either side of the interface, thus in first approximation one may write $d = d(T, t)$. The details of $d(T, t)$ would provide *valuable engineering information* on optimal process temperatures, residence times in the mixing drums. Also contact time and temperature during transport and storage should then be taken into account. In conclusion, when the average bitumen coating thickness D on the aggregates is known, a combination of temperature T and residence time t could be selected such that complete blending occurs before the material is applied to the road. Then complete blending will occur as long $D \leq \frac{1}{2}d(T, t)$.

The properties of the blended bitumen is found to be in between the values for the bitumen at either site of the interface (Table 1). This is the case for the microstructural parameters size, surface coverage, aspect ratio and wavelength of the profile. The only parameter that behaves different is the amplitude of the wrinkling profile: for a softer material a higher amplitude is expected than for the stiffer material. This has also been found in the current context: the softer, virgin binder was found to possess a larger wrinkling amplitude than the RAP binder. Therefore, one would expect the amplitude of the 'blended binder' to be in between the respective amplitudes of the virgin and RAP binders. However, it happened to possess the largest amplitude instead. This anomalous behavior might be explained by the long tail in the size distribution of the blended sample (Figure 5b). A scaling relation between microstructural size and amplitude may exist, but this has not been worked out any further in the context of this research.

From the DSR data of the three bitumen (Figure 1) it is evident that the mechanical properties of the blend are in between those of the non-blended binders. As corresponding behavior is found for the AFM derived microstructural parameters, this may hint to the conclusion that microstructure has a direct link to mechanical properties of the binder. This relation of microstructure to mechanical properties can also be inferred from the microstructural profile data (Figure 6). The 'wrinkles' could originate from a mismatch in coefficient of thermal expansion of both phases in the microstructure. Upon solidification stresses will lead to buckling of the elliptical domains: the observed wavy profile

pattern. Details of this pattern may eventually be directly translated into mechanical properties (31,32), though more research is needed to firmly establish this link.

Another surprising finding is that mixing of virgin and RAP bitumen results into a completely new material (from the microstructural point of view). If the process involved was merely mixing of two distinct colloidal fluids, the colloidal particles of both materials should be present in their mix as well. However, here a completely new ‘colloidal’ material is found; no traces of the original material’s microstructure is present anymore in the ‘mixed zone’. The two possible scenarios for mixing are sketched in Figure 7b; obviously scenario B is closest to the observations, hence one should speak about blending rather than mixing: the identity of the individual mixing components is lost, instead a new ‘material’ is found. This also sheds a new light on the proposed colloidal structure of bitumen (33): it may still be a colloidal system, but with unstable colloidal particles, which are prone to rearranging themselves into new assemblies.

We conclude that at the interface of RAP and virgin bitumen complete blending has been observed. However, this does not yet rule out the ‘black rock’ hypothesis (3), because the RAP bitumen was chemically extracted from the RAP. The challenge remains to proof the same for RAP coated aggregates that are mixed with fresh, soft bitumen. Finally, the results obtained in this study are also very useful to screen and understand the processes that occur when *rejuvenators* are used to enhance service life of aged asphalt. A similar approach could be used to explore the blending of RAP containing *Polymer Modified Bitumen* (PMB) with soft binders.

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REFERENCES

1. European Asphalt Pavement Association. *Asphalt in figures 2010, 2011*, Report.
2. Copeland A. *Reclaimed Asphalt Pavement in Asphalt Mixtures: State of the Practice*. 6300 Georgetown Pike McLean, VA 22101-2296: Office of Infrastructure Research and Development, Turner Fairbank Highway Research Center, Federal Highway Administration; 2011. FHWA-HRT-11-021.
3. Al-Qadi I.L., M. Elseifi and S.H. Carpenter. *Reclaimed Asphalt Pavement: A Literature Review*. University of Illinois at Urbana-Champaign; 2007.
4. Aurangzeb Q, I.L. Al-Qadi, W.J. Pine, J.S. Trepanier, and I.M. Abuawad. Thermal cracking potential in asphalt mixtures with high RAP contents. In: A. Scarpas, N. Kringos, I. Al-Qadi and A. Loizos, editors. *7th RILEM International Conference on Cracking in Pavements: Mechanisms, Modeling, Testing, Detection and Prevention Case Histories*. vol. 4 of RILEM Bookseries, RILEM, 2012, pp. 1271–1280.
5. McDaniel R.S., H. Soleymani, R.M. Anderson, P. Turner and R. Peterson. NCHRP Final Report (9-12). Transportation Research Board, Washington, D.C., 2000.
6. Tapsoba N., C. Sauzeat, H. Di Benedetto, H. Baaj and M. Ech. Low-temperature cracking of recycled asphalt mixtures. In: A. Scarpas, N. Kringos, I. Al-Qadi and A. Loizos, editors. *7th RILEM*

International Conference on Cracking in Pavements: Mechanisms, Modeling, Testing, Detection and Prevention Case Histories. vol. 4 of RILEM Bookseries, RILEM, 2012. pp. 1261–1270.

7. West R., J. Michael, R. Turochy and S. Maghsoodloo. Use of data from specific pavement studies experiment 5 in the long-term pavement performance program to compare virgin and recycled asphalt pavements. In *Transportation Research Record: Journal of the Transportation Research Board*. No. 2208, Transportation Research Board of the National Academies, Washington, D.C., 2011, pp. 82–89.

8. Copeland A., J. D'Angelo, R. Dongré, S. Belagutti and G. Sholar. Field Evaluation of High Reclaimed Asphalt Pavement–Warm-Mix Asphalt Project in Florida. In *Transportation Research Record: Journal of the Transportation Research Board*, No. 2179, Transportation Research Board of the National Academies, Washington, D.C., 2010, pp. 93–101.

9. Ma T., E. Mahmoud and H. Bahia. Estimation of Reclaimed Asphalt Pavement Binder Low-Temperature Properties without Extraction. In *Transportation Research Record: Journal of the Transportation Research Board*, No. 2179, Transportation Research Board of the National Academies, Washington, D.C., 2010, pp. 58–65.

10. Oliver J.W.H. The Influence of the Binder in RAP on Recycled Asphalt Properties. *Road Materials and Pavement Design*, Vol. 2, No. 3, 2001, pp. 311–325.

11. Bonaquist R. Can I Run More RAP? *HMAT: Hot Mix Asphalt Technology*, Vol. 5, No. 5, 2007, pp. 11–13.

12. Stephens, J. E., J. Mahoney and C. Dippold. *Determination of the PG Binder Grade to Use in a RAP Mix*. University of Connecticut, 2001.

13. Huang B., G. Li, D. Vukosavljevic, X. Shu and B. Egan. Laboratory Investigation of Mixing Hot-Mix Asphalt with Reclaimed Asphalt Pavement. In *Transportation Research Record: Journal of the Transportation Research Board of the National Academies*, No. 1929, Transportation Research Board of the National Academies, Washington, D.C., 2005, pp. 37–45.

14. Carpenter S.H. and J.R. Wolosick. Modifier Influence in the Characterization of Hot-Mix Recycled Material. *Transportation Research Record: Journal of the Transportation Research Board*, No. 777, Transportation Research Board of the National Academies, Washington, D.C., 1980, pp. 15–22.

15. Mohajeri M., A.A.A. Molenaar and M.F.C Van de Ven. Cracking resistance of recycled asphalt mixtures in relation to blending of RAP and virgin binder. In: A. Scarpas, N. Kringos, I. Al-Qadi and A. Loizos, editors. *7th RILEM International Conference on Cracking in Pavements: Mechanisms, Modeling, Testing, Detection and Prevention Case Histories*. vol. 4 of RILEM Bookseries, RILEM, 2012, pp. 1311–1321.

16. Loeber L. New direct observation of asphalts and asphalt binders by scanning electron microscopy and atomic force microscopy. *Journal of Microscopy*, Vol.182, No. 1, 1996, pp. 32–39.

17. Pauli A.T. Atomic force microscopy investigation of SHRP asphalts. *ACS Division of Fuel Chemistry Preprints*, Vol. 46, 2001, pp. 104.

18. Pauli A.T. and W. Grimes. Surface morphological stability modeling of SHRP asphalts. *ACS Division of Fuel Chemistry Preprints*, Vol. 48, 2003, pp. 19.

19. Pauli A.T., R.W. Grimes, A.G. Beemer, T.F. Turner and J.F. Branthaver. Morphology of asphalts, asphalt fractions and model wax-doped asphalts studied by atomic force microscopy. *International Journal of Pavement Engineering*. Vol. 12, No. 4, 2011, pp. 291–309.

20. Nahar S., B. Dillingh, S. Erkens, A.J.M. Schmets, H. Fischer, A. Scarpas and G. Schitter. *Is Atomic Force Microscopy suited as tool for fast Screening of Bituminous Materials? An Inter-laboratory Comparison Study*. Paper No. 13-4131, CD-ROM, Transportation Research Board of the National Academies, Washington, D.C., 2013.
21. Torquato S. Modeling of physical properties of composite materials. *International Journal of Solids and Structures*, Vol. 37, No. 12, 2000, pp. 411 – 422.
22. Kringos N., A. Scarpas, T. Pauli and R. Robertson, A thermodynamic Approach to Healing in Bitumen. In: Loizos A, Partl MN, Scarpas T, editors. *Advanced Testing and Characterization of Bituminous Materials*. Taylor & Francis, 2009. pp. 123–128.
23. Garcia R. and R. Perez. Dynamic atomic force microscopy methods. *Surface Science Reports*, Vol. 47, No. 6-8, 2002, pp. 197–301.
24. Binnig G., C. Gerber, E. Stoll, T.R. Albrecht and C.F. Quate. Atomic resolution with atomic force microscope, *Surface Science*, Vol. 189-190, 1987, pp.1-6.
25. Blanchard C.R. Atomic Force Microscopy, *The Chemical Educator*, Vol. 1, 1996, pp. 1–8.
26. Eaton P. and P. West. *Atomic Force Microscopy*. Oxford University Press, 2010.
27. Raghavan D., X. Gu, T. Nguyen, M. VanLandingham and A. Karim. Mapping Polymer Heterogeneity Using Atomic Force Microscopy Phase Imaging and Nanoscale Indentation, *Macromolecules*, Vol. 33, No. 7, 2000, pp. 2573–2583.
28. Necas D. and P. Klapetek. Gwyddion: an open-source software for SPM data analysis, *Central European Journal of Physics*, Vol. 10, 2012, pp. 181–188.
29. Dorset D.L. *Crystallography of the Polymethylene Chain: An Inquiry into the Structure of Waxes*, vol. 17 of International Union of Crystallography Monographs on Crystallography, Oxford University Press, 2005.
30. Schmets A.J.M., N. Kringos, T. Pauli, P. Redelius, and A. Scarpas. On the existence of wax-induced phase separation in bitumen. *International Journal of Pavement Engineering*, Vol. 11, No. 6, 2010, pp. 555–563.
31. Brochard-Wyart F. and P.G. de Gennes. How Soft Skin Wrinkles. *Science*, Vol. 300, No. 5618, 2003, pp. 441.
32. Cerda E. and L. Mahadevan. Geometry and Physics of Wrinkling. *Phys Rev Lett.*, Vol. 90, No. 7, 2003, pp. 074302
33. Lesueur D. The colloidal structure of bitumen: Consequences on the rheology and on the mechanisms of bitumen modification. *Advances in Colloid and Interface Science*, Vol. 145, No. 1-2, 2009, pp. 42 – 82.

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FIGURE 7 (a) Possibilities for the formation of a blended zone between RAP and virgin aggregates. (b) Scenarios for mixing or blending of microstructural properties at the interface of RAP and virgin bitumen.

TABLE 1 Quantitative details of the observed bitumen microstructures

	Domain size			Domain shape		Wrinkling	
	maximum (μm)	minimum (μm)	mean (μm)	aspect ratio	phase fraction (%)	λ (μm)	amplitude (nm)
virgin-bitumen	4.77	0.46	2.39 $\pm$ 1.0	0.68 $\pm$ 0.15	53.77	0.6	16 $\pm$ 5
RAP-binder	1.71	0.07	0.57 $\pm$ 0.4	0.81 $\pm$ 0.14	9.92	0.3	11 $\pm$ 3
blended-binder	2.07	0.16	0.66 $\pm$ 0.4	0.75 $\pm$ 0.17	26.43	0.4	18 $\pm$ 6

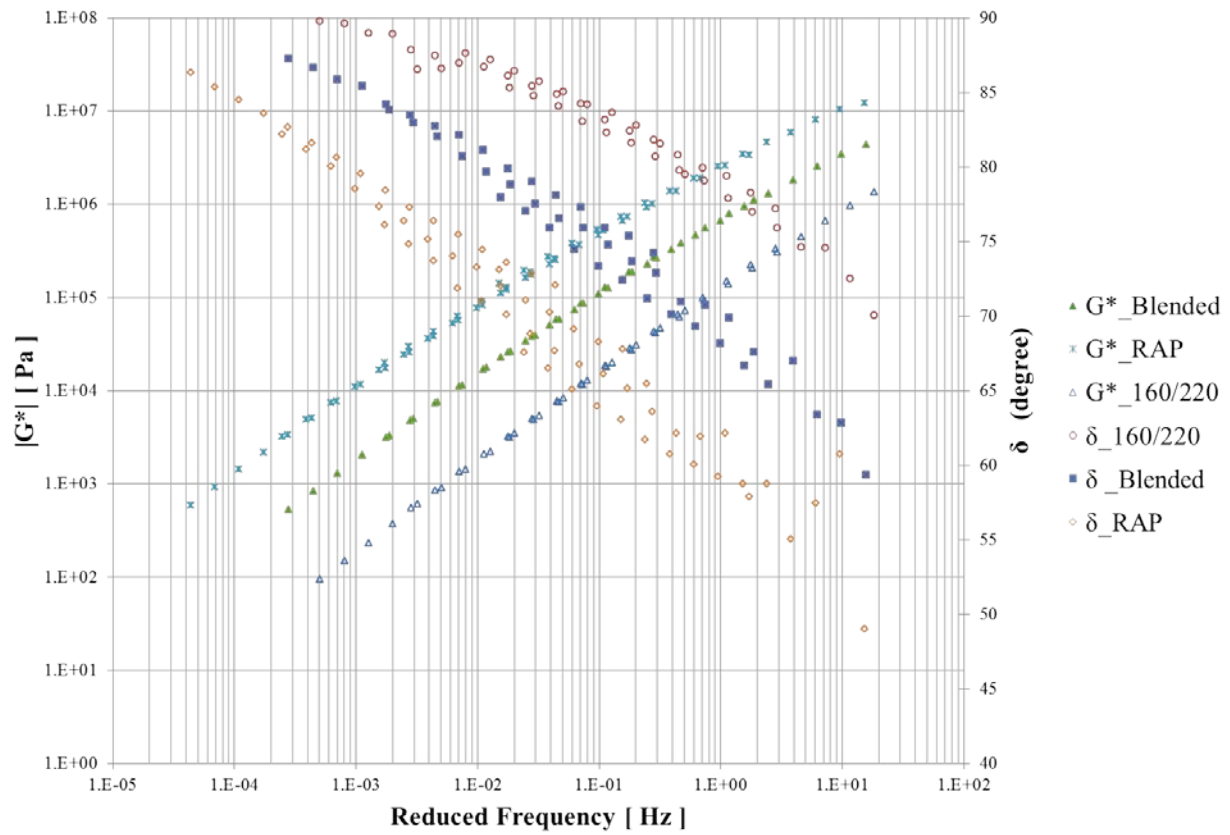


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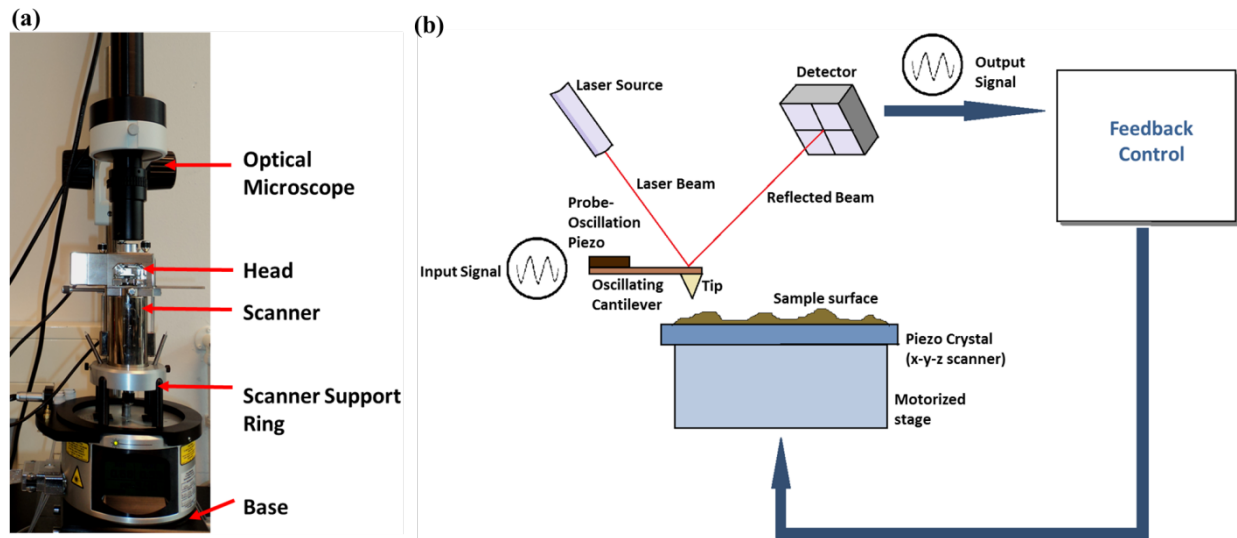


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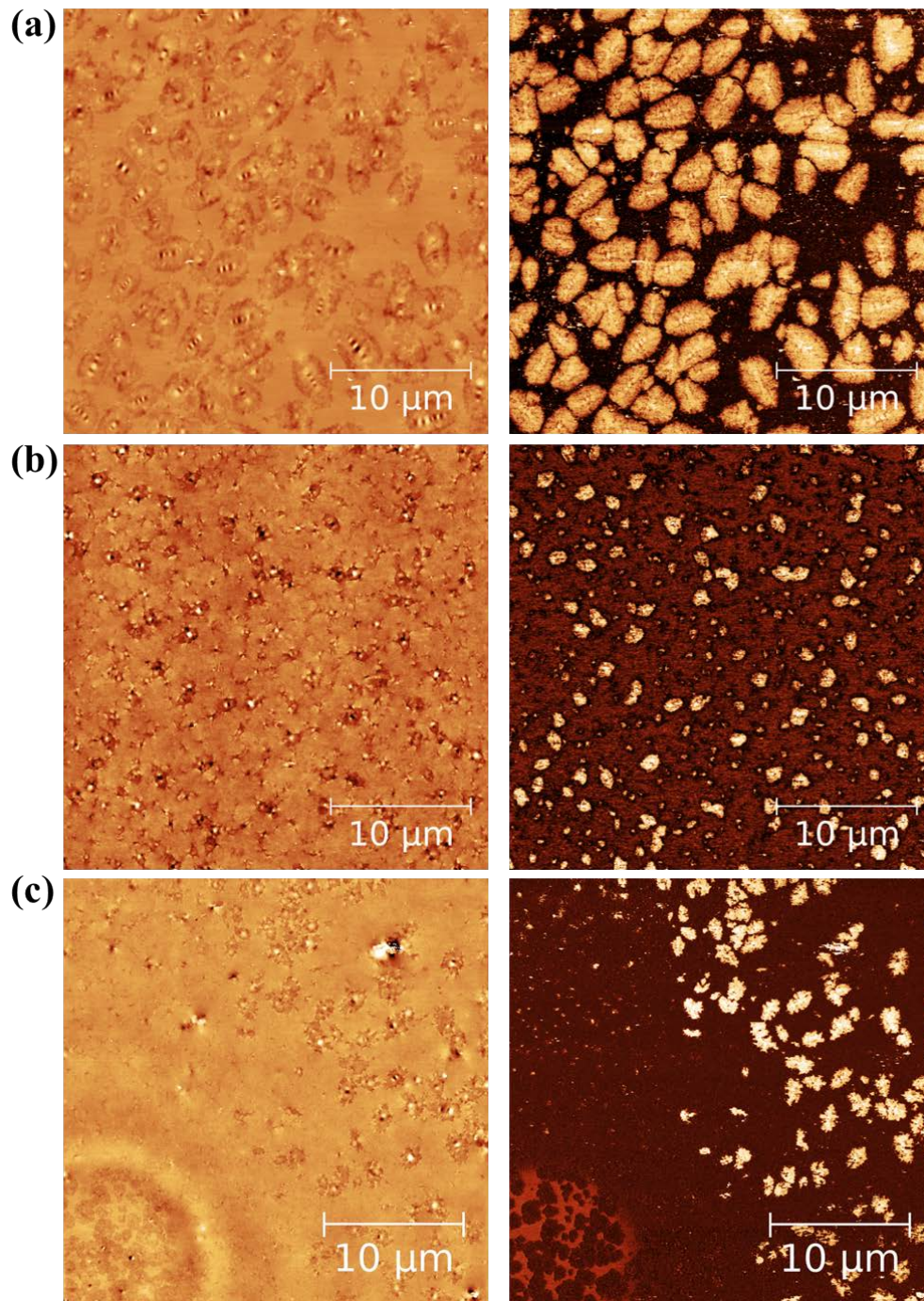


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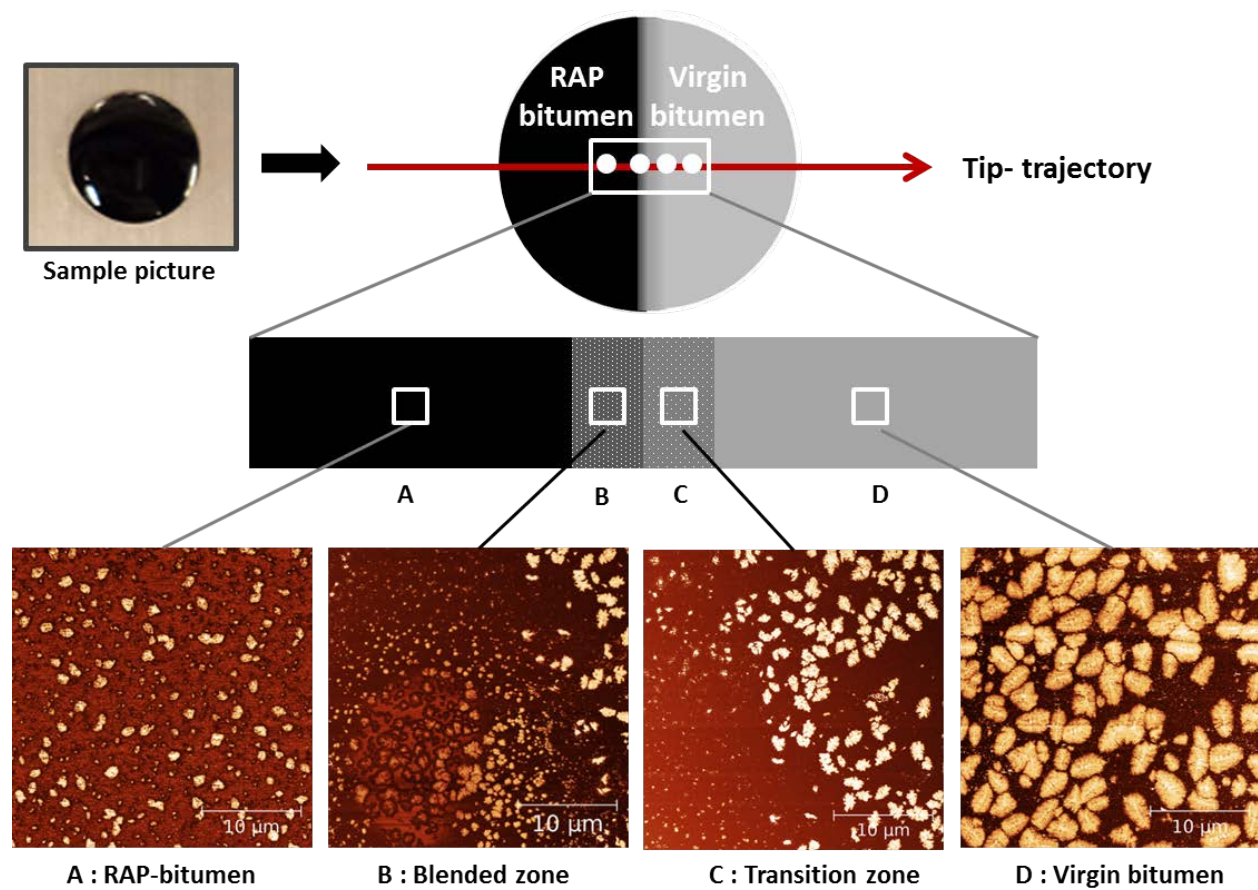


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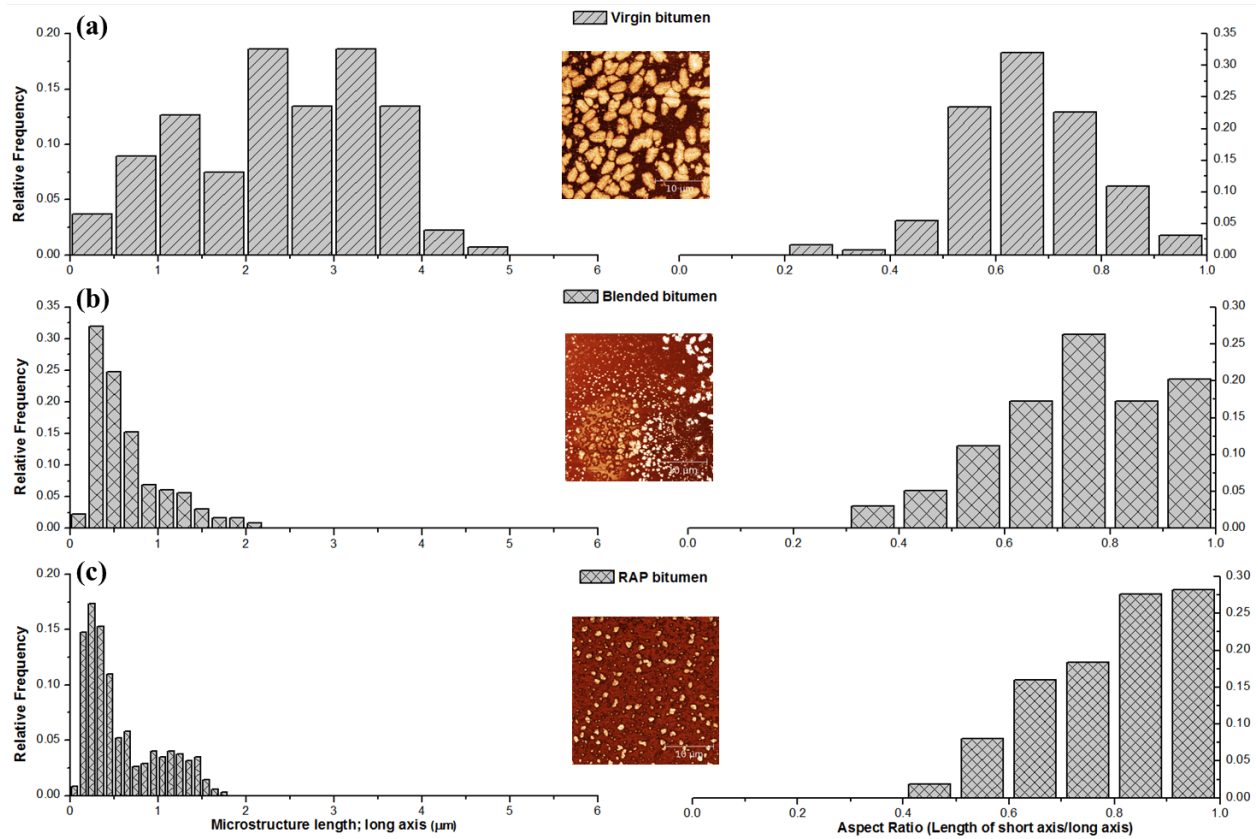


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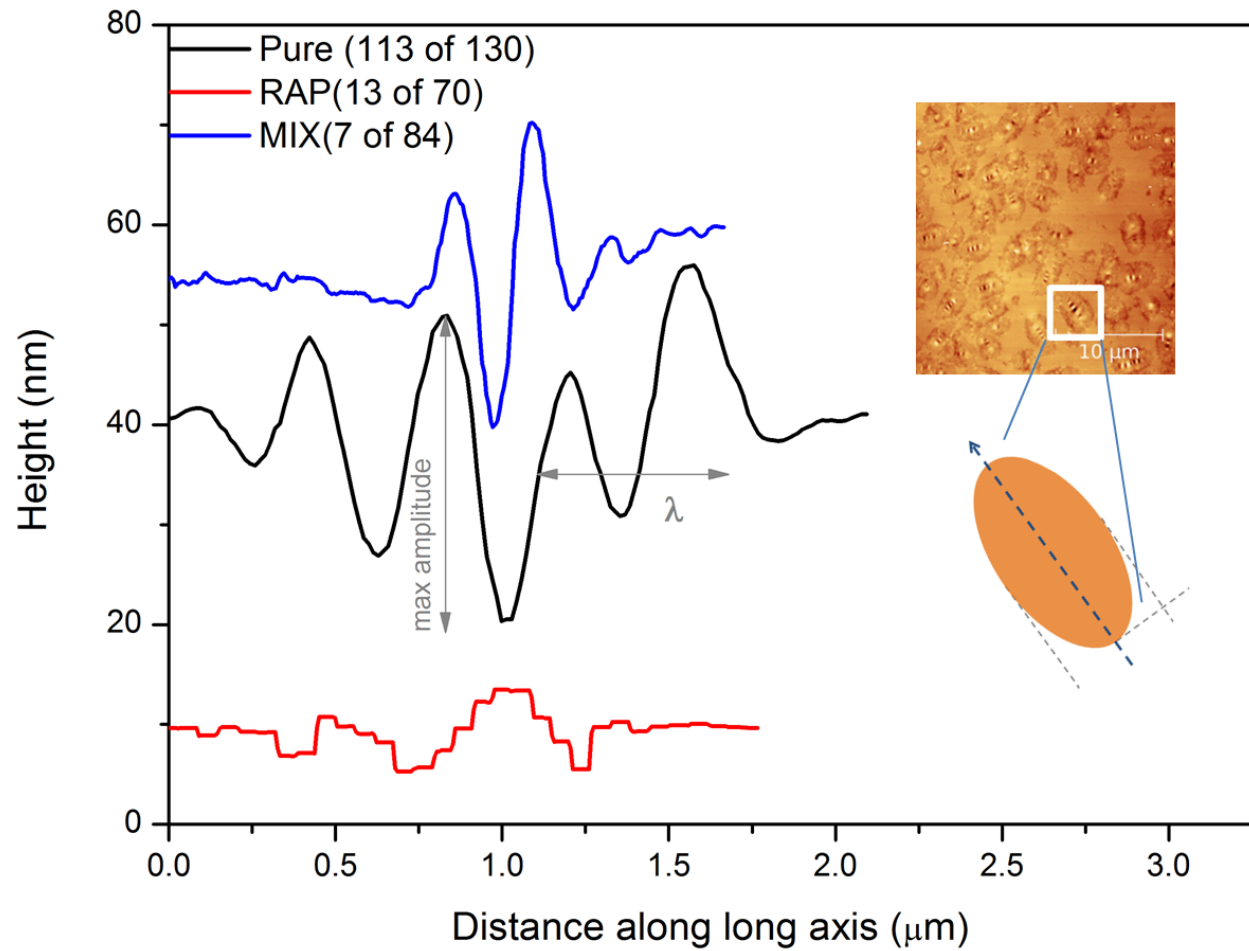


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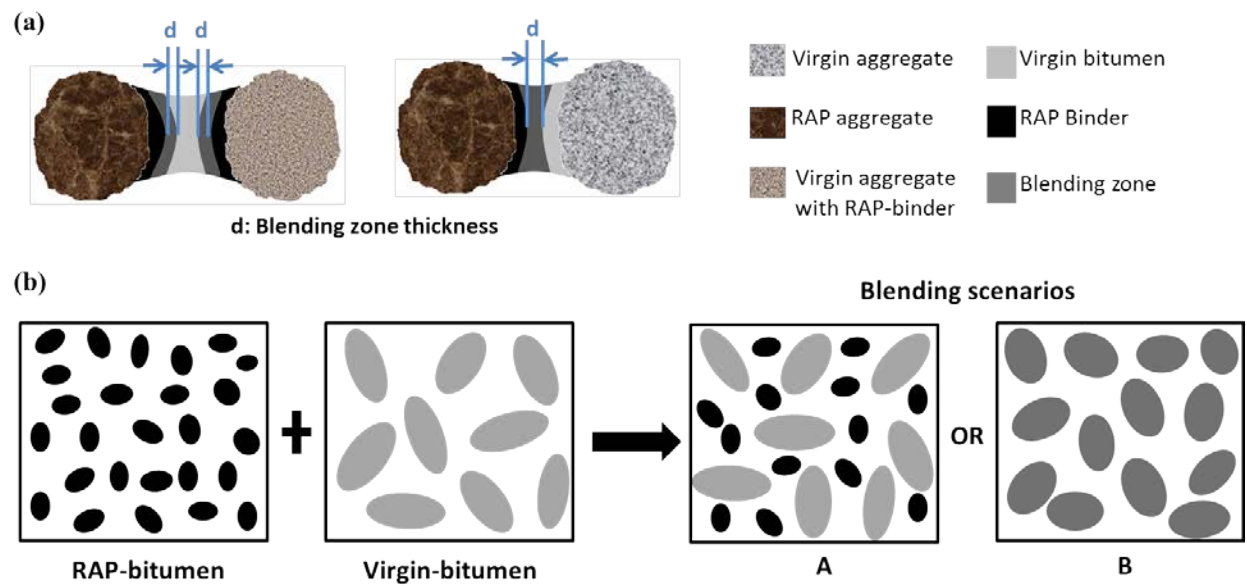


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