"The physicochemical influence of the inorganic phase on the aging and performance of asphalt pavements in Finland"

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The need to understand the reasons behind the deterioration of asphalt pavements led to the research focused on the relationship between molecular interactions within this composite material and its macroscopic properties. The aim of the work was to establish the risks arising from the interaction between bitumen and the inorganic compounds, namely fine particles passing the 125 μm sieve.

The first case study, the forensic analysis of failed asphalt pavement, Ring Road II in the Helsinki Metropolitan Area, revealed that a substitution of the asphalt concrete component occurred between the mix design and production, mass by mass. The mineral filler used contained fly ash in lieu of using pure limestone as was designed. This work investigated how physicochemical properties, such as the altered density and chemical composition of the filler, affect the properties of asphalt during recycling in Finland.

First, the iron compounds located on the surface of the filler grains, in addition to the differences in the volume of the reinforcing phase, correlated well with the increase in the softening point of an asphalt mastic. It was found that with the decreasing density of the filler and increasing iron content the stiffness of the mastic increased significantly (by up to 15°C increase in softening point of the mastic at equal filler to bitumen mass ratio). In the context of material strength, this is desirable. However, in the context of recyclability the problems with grain separation in the reheating stage of the process, as well as with the window of compaction exist. The project also theoretically explored on a basic level the effect of the altered volumetric composition in the mastic on the process of rejuvenation or fresh bitumen diffusion during the recycling, finding that the increase of temperature should be required to assure good blending between those components at the higher volumetric content of the filler.

However, this mostly physical effect inflicted by the composition of the filler was extended to the chemical interaction of the filler with the deicing chemicals commonly used in the northern climate. Namely, the use of calcium chloride ($CaCl_2$) poses a risk of creating the iron (III) chloride ($FeCl_3$) upon contact with the mineral fillers of high iron content. The correlation between chloride content in the fine material, as witnessed by x-ray fluorescence spectroscopy, and iron content was determined based on the materials collected from the analysis of hot in-place recycling works conducted on the second case study – Highway number 1.

During the recycling of an asphalt concrete, the accumulation of chlorides could prove dangerous, if those inorganic compounds catalyzed the bitumen oxidation. This work used a novel methodology of the in-situ Fourier transform infrared spectroscopy performed in the Attenuated Total Reflectance mode at 163°C (the temperature typically used in the asphalt plant and during the laboratory aging of bitumen) to study the interaction between bitumen, air and chlorides. The iron (III) chloride, which can be formed from fines in acidified deicing brines, was confirmed as a catalyst of oxidation. However, the typical deicing chemical, calcium chloride, left the reaction mechanism and its rate unaltered.

What is more important, the novel time-resolved infrared spectroscopy analysis provided insight into pure bitumen oxidation as well, revealing that the reaction at high temperatures occurs step-wise i.e., thiol oxidation followed by ketone formation. Controlling the reaction by blocking the thiol oxidation via interaction with copper successfully hindered the ketone formation. The novel approach to asphalt concrete engineering on a molecular level is promising, but the effect of such control on the physical properties of asphalt pavements still needs to be confirmed.

The effect which oxidation of bitumen, followed by blending with fresh organic material, has on the molecular weight distribution, change in polarity, and rheological characteristic was studied in laboratory conditions to simulate four cycles of reuse. Increasing molecular weight distribution correlated well with an increase in the elastic response of the material. The loss of the ability to dissipate energy by flowing was postulated to cause the declining performance in the cold climate regions.

In conclusion, the mineral fillers with high iron content are posing a risk during the hot in-place recycling process. Especially, in terms of the chemical influence, the high iron content raises the potential of the asphalt concrete to accumulate the chlorides upon contact with deicing chemicals, which may lead to catalytic aging during the reheating of the asphalt concrete during the recycling. Despite the found influence by the inorganic phase, the importance of rejuvenation and recovery of the flow properties for good performance in cold climate is still underlined.

The project was sponsored by the Finnish Transportation Agency. The above summary of one of the aspects studied within the project “Pavement Life Cycle Research Program, Recycling and Rejuvenation” comprises the main content of the doctoral dissertation by the author. However, the project was a collaborative action between researchers of Aalto University, contractors, local bitumen provider and Finnish Road authorities. All of the other publications of the project are referred to in the dissertation by the author and in the body of the final report.