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Effect of zeolite properties on asphalt foaming

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HIGHLIGHTS

- Synthetic zeolites from fly ashes can be used in asphalt foaming technology.
- The efficiency of asphalt foaming depends on the type of zeolite structure/property.
- DTA/TG assesses amount and timing of water release from zeolite during foaming process.

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ABSTRACT

This study includes investigation of two zeolite additives to non-modified asphalt binder and associated foaming phenomena. Two zeolites differ in their crystalline structures, i.e. one was Na-P1 synthetic zeolite and second was clinoptilolite natural zeolite. Prior to asphalt foaming these materials were additionally soaked with water which resulted in total of four base materials for laboratory investigation. The amount of zeolite dosed to the asphalt binder was 5% with respect to asphalt mass. The foaming effect was examined in terms of dynamic viscosity and further analyzed as a function of physical and chemical properties of both zeolites. In conclusion it was noted that the foaming effect strongly depends on the amount of water in zeolite structure, mode of its release with time, type of exchangeable cations as well as silicone to aluminum ratio in zeolites and finally their texture properties.

1. Introduction

Changes of the environment and its degradation are important problems in the modern world. Extremely hazardous compounds which contaminate the environment include: carbon dioxide (CO_2) – which is one of the reasons behind greenhouse effect – as well as carbon monoxide (CO), sulfur dioxide (SO_2), nitrogen oxides (NO_x) – which cause acidification of the environment. Construction industry contributes to these emissions and one example is asphalt industry with HMA (Hot Mix Asphalt) plants in particular. Conventional HMA technology requires heating asphalt up to temperatures of 140–200 °C. Starting with these temperatures, asphalt achieves adequately low viscosity, allowing proper coating and mixing with aggregate batch [1]. On the other hand, high temperatures require high energy use, and lead to high emissions of CO_2, SO_2 and NO_x, as well as aerosols which pose a threat to the environment and workers health.

In order to mitigate these adverse effects, modern asphalt technology has been working on environment-friendly technologies. Over the last 20 years, several dozens of WMA (Warm Mix Asphalt) technologies were developed, allowing decrease in mixing and compaction temperatures by approx. 20–40 °C [2–4]. Temperature level decrease has resulted in significant reduction in the emission of harmful compounds [5–8]. Further, due to better workability WMA can contain higher RAP (Reclaimed Asphalt Pavement) content [9–13] as well as contain CRM (Crum Rubber Modifier) and recycled Asphalt Shingles (RAS) additives [14–16]. The construction of road surfaces using WMA technology also includes lower energy use and lower asphalt surface construction costs [17–22]. However, the range and the extent of observed benefits depends on the implemented WMA technology. The most common WMA technology utilizes asphalt foaming by adding a small amount of water into the hot binder or directly to the aggregate and asphalt mixer [23–28]. Several asphalt foaming technologies have been developed, which could be generally categorized as two groups [2].

- water-based,
- water-storing additives, e.g. zeolites.

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Zeolites are a group of aluminosilicates with diversified frameworks. The Structure Commission of the International Zeolite Association (IZA-SC) assigns a 3-letter framework type code to each zeolite unique structure (e.g. for clinoptilolite it is HEU, for Na-A zeolite it is LTA). This code is a part of the official International Union of Pure and Applied Chemistry (IUPAC) nomenclature for microporous materials [29].

Crystalline zeolite structure includes empty spaces in the form of chambers and channels which provides zeolites with unique properties that are particularly advantageous for a number of industrial applications, including WMA [30–32]. Channel dimensions of the zeolites are on the order of 3 Å up to 30 Å which makes them sufficiently large so that not only single atoms but also small chemical compound particles can diffuse and penetrate inside them. One particular distinctive property of minerals from this group includes presence of water particles in their composition, so-called 'zeolite water'. When heating material up to approximately 400 °C, zeolite water is released from the mineral structure in a continuous manner without changes in the crystalline volume. Consequently, during cooling a mineral in the humid environment, water particles are continuously absorbed by its structure.

Genetically, the zeolite material group includes natural and synthetic zeolites. At this moment, natural zeolite group includes approximately 100 different minerals. However, only some of them exist in the form of accumulations of so-called deposits that are economically feasible in terms of mining and processing. The most common zeolites creating deposits include: clinoptilolite, phillipsite, chabazite and mordenite [33]. In the second genetic group, synthetic zeolites are artificially created with strictly defined structure parameters which is dictated by a particular industrial application (e.g. selective molecular sieves). The most common zeolites obtained in the industrial synthesis processes include minerals of Na-X, Na-Y, Na-A, ZSM-5 type. Synthetic zeolites are obtained from the chemical reagents, mineral materials such as clay minerals and silica-group minerals as well as from some waste by-products of coal processing (such as fly-ashes) [34,35]. Up to date the WMA technologies include the use of synthetic zeolites manufactured using chemical reagents [6,36–38], synthetic zeolites from fly ashes [39], and natural zeolites such as clinoptilolite [38–42]. Studies available in the literature have examined the effect of zeolite additives on asphalt properties [43–46] as well as on the properties of WMA produced at lowered temperatures [9,47–50].

Regardless of the genetic group, zeolites allow for asphalt foaming due to the incremental release of water stored in their internal structure. Discharge of zeolite water from the crystalline structure is a long-term process. Therefore it is feasible to improve WMA workability during production, construction and compaction [5]. Study by Lai et al. focused on examining in details this phenomenon [51]. That study analyzed the release of zeolite water from the zeolite with LTA structure as a function of time and temperature. It was noted that zeolite gradually released water with time up to stabilization that occurred after 20–40 min. Stabilization time was highly depended on temperature and it was observed that the higher the temperature, the more water was released from zeolite structure within analyzed temperature range (from 76.6 °C to 121.16 °C).

Optimal amount of water needed for asphalt foaming spans between 2 and 4% with regards to the asphalt mass [52–54]. With insufficient amount of water the foaming is ineffective, whereas with the excessive water there is a significant risk of adhesion failure between asphalt and aggregate [55]. Therefore when using asphalt foaming technology with zeolites, it is necessary to recognize the topology of zeolite structure, the amount of zeolite water and its character, as well as water release characteristics as a function of time and temperature. In terms of the environment protection, it is desirable that the amount of water released from zeolite would be as high as possible, and temperature of its release as low as possible.

In the recent years there has been a number of studies assessing the effect of zeolites on asphalt and mix asphalt properties but only one has focused on zeolite properties. In that study [39], optimal amount of zeolite was assessed using compactability experiments in the gyratory compactor. In addition to optimal zeolite content, the results from the physical and chemical tests showed that synthetic zeolites with NaP1 structure type are feasible for WMA technology. However, that study [39] did not explain the mechanism of asphalt foaming with zeolite water. Other available studies in the literature do not contain any results allowing to associate the asphalt foaming effect with the type of zeolite structure. Thus it seems worthwhile to study the hypothesis that foaming efficiency is the results of zeolite water release determined by the zeolite structure. Having considered that, the aim of this study was to analyze the effect of Na-P1 (synthetic) and clinoptilolite (natural) zeolite type on asphalt foaming efficiency and attempt to explain the mechanism of this process through the topology of zeolite structures. The first part of this study presents a comprehensive property evaluation of both zeolites together with the base asphalt. Then foaming effect is assessed for all four zeolite-modified asphalts in terms of the complex shear modulus and dynamic viscosity. Final part includes discussion and identifies potential links between zeolite topology and foaming potential.

2. Materials and methods

2.1. Base materials

Zeolites used in this study represent two significantly different framework topologies (see Fig. 1). According to standard nomenclature [29] Na-P1 (synthetic zeolite) represents gismondine-like framework (GIS). In this structure, 8-membered channels of 3.1×4.5 Å and 2.8×4.8 Å are formed by two 4-membered rings. On the other hand, natural clinoptilolite characterizes heulandite topology (HEU) with two-dimensional channel system formed by 8-membered rings (4.1×4.1 Å) and 10-membered rings (5.5×3.1 Å) [56].

The Na-P1 was obtained in the conversion of F-class fly ash and aqueous solution of sodium hydroxide. The preparation of the zeolite was performed using patented semi-industrial scale device for zeolite synthesis. In that process, 20 kg of fly ash and 12 kg NaOH were mixed with 90 dm^3 of H_2O. The substrates were subjected to hydrothermal synthesis at the temperature of 80 °C and duration 36 h [34,58–60].

Natural zeolite clinoptilolite (ZN-C) originated from Ukrainian deposit (Sokyrnytsya) in a form of zeolitic tuff [61].

The foaming effect was studied using 35/50 asphalt binder. 35/50 parameter refers to European classification of asphalt binders that is based on penetration testing according to EN 12591:2010. Typical elemental composition and fraction composition for this asphalt are presented in Table 1. Chemical composition was determined using ED-XRF method (Energy Dispersive X-ray Fluorescence Spectroscopy, spectrometer Epsilon 3x). Fractional composition was determined using TLC-FID chromatography (Thin-Layer Chromatography Flame Ionization Detection) that identifies four typical asphalt groups – namely saturate, aromatic, resin and asphaltene (SARA) [62]. The TLC-FID chromatogram is presented in Fig. 2. Gaestel Index (I_c) calculated from the SARA results is equal to 0.37 which indicates colloidally stable microstructure of this base binder.