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THERMAL BEHAVIOR OF CLAY MINERALS BEYOND THERMODNAMIC EQUILIBRIUM

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Detachment of glazed stoneware tiles from the bonded surface and further architectural ceramics as a consequence of concave curvature or moisture expansion is a recent problem of silicate ceramic industry. Theses phenomena seem to be directly related to fast firing process of modern kilns, where the whole firing cycle takes place in less than 40 minutes.

Within this context, rehydroxilation of dehydroxilated clay-related compounds such as metakaolinite, dehydroxilated montmorillonite or illite, is considered a main driving force. In order to get an insight into the sinterkinetic behavior, the relevant mineral reactions at high heating rates have been studied in more detail. Therefore, several in-situ DTA-MS/DTA-IR and ex-situ firing experiments using rates of 5, 30 and 50 K min^{-1} in the temperature regime between 1100 °C and 1250 °C have been performed on 1.) individual separated clay minerals (kaolinite, montmorillonite, illite) as well as on 2.) two different mixtures (consisting of quartz, feldspar, kaolinite and illite) close to industrial compositions.

Preliminary results show that compared to equilibrium studies all mineral reactions are shifted to higher temperatures by 10 to 30 K, in certain cases up to 76 K. With increasing temperature, the content of amorphous material increases according to XRPD evaluation. DTA experiments on kaolinite with a heating rate of 30 K min^{-1} showed a significant exothermal signal at ~ 1000 °C which is allocated to the formation of spinel. This phase, however, could not be identified using XRPD. Thus, an amorphous or at least highly disordered structure has to be considered. In montmorillonite, the formation of γ-cordierite was observed at 1200 °C, a phase first described by DIMITRIJEVIC & DONDUR (2007).

Further studies will include the quantification of the exact amounts of each phase and the total amorphous content using the Rietveld method. HT- in situ XRPD experiments with the separated clay minerals and the two aforementioned mixtures will be performed to obtain an overview of the structurally traceable reactions in the systems. In order to determine which phases (ordered and amorphous) contribute to the assumed rehydroxylation reactions, actual case examples of deformed tiles will be analyzed with XRPD and vibrational spectroscopy.

DIMITRIJEVIC, R., DONDUR, V., (2007): J. Serb. Chem. Soc., 72, 1351–1362.