

## Supporting information

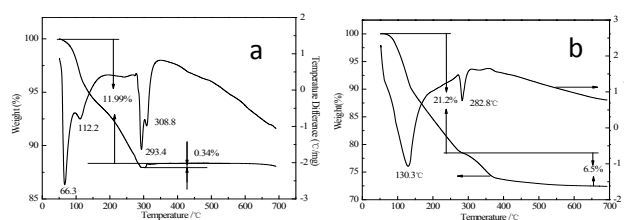


Fig.1 TG-DTA curve of pure NaOH (a) and ZrSiO<sub>4</sub> during the NaOH fusion process at the molar ratio of 6:1 (b)

For further study on the effect of OH<sup>-</sup> in the reconstruction of silicate network, TG-DTA analysis is performed. As is shown in Fig.1 (a), there are four endothermic peaks located at 66.3 °C, 71.6 °C, 125.7 °C and 285.2 °C on DTA curve of pure NaOH. The corresponding weight loss can be observed on the TG curve. Weight loss at 66.3 °C and 112.2 °C is due to volatilization of physically adsorbed water and crystal water. The weight loss rate of this process is 11.99 %. The weight of reaction system maintains constant when the temperature is over 300 °C, which means that NaOH is not volatile in optimum reaction temperature range.

The TG-DTA measurement of NaOH fusion reaction with molar ratio of 6:1 shows quite different curves compared with pure NaOH. The DTA peaks located at 130.3 °C are assigned to removal of water remaining in the mixture and crystal water. The ratio of weight loss was 21.2 % when reaction reaches thermal stabilization. The DTA peak at 282.8 °C is due to NaOH melting and alkali fusion reaction together. Because the NaOH is mixed with other compounds, the melting temperature is lower than the theoretical melting point. In contrast to pure NaOH, reaction system contains more H<sub>2</sub>O at higher temperature than 300 °C. A strong weight loss of about 6.5 % is visible between 280 °C and 600 °C. This decrease in weight is due to not volatilization of NaOH but the expulsion of water, which is generated by the condensation of neighboring silanol groups<sup>1-2</sup>. The evaporation of H<sub>2</sub>O obtained by dehydration condensation cause sustained weight loss at higher than 300 °C in the TG curve. Through the TG-DTA analysis we can know that the reaction of ZrSiO<sub>4</sub> decomposed by NaOH fusion produce H<sub>2</sub>O when the temperature is 300 °C above.

### The TG-DTA measurement of KOH fusion reaction

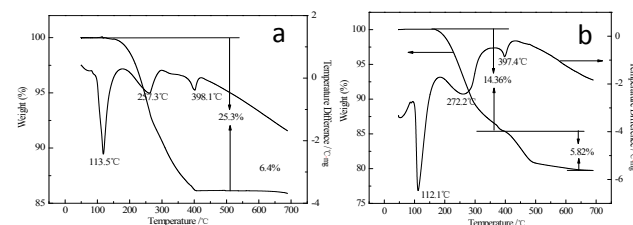


Fig.2 TG-DTA curve of pure KOH (a) and ZrSiO<sub>4</sub> during the KOH fusion process at the molar ratio of 6:1(b)

TG-DTA curves of pure KOH and ZrSiO<sub>4</sub> decomposed by KOH at the molar ratio of 6:1 are shown in Fig.2 (a). Three endothermic peaks located at 113.5 °C, 257.3 °C and 398.1 °C can be observed on the DTA curve and the corresponding weight losses appear on the TG curve. The peak at 113.5 °C and 257.3 °C are due to the volatile of free water in residual compounds and the removal of crystal water, respectively. Below 400 °C, a weight loss resulting from adsorptions of physically adsorbed water is observed. Because the air ratio of KOH more easily absorbs moisture than NaOH, the weight decrease continually without weight stable region at the corresponding position of TG curve and weight loss rate is 25.3 %. When the temperature rise to 398.1 °C, the KOH melt induce endothermic peak of the TG curve and no corresponding phenomenon appear on weight. The weight does not change again because KOH does not decompose or volatile in this temperature range.

TG-DTA curve of ZrSiO<sub>4</sub> during the KOH fusion process with molar ratio of 6:1 is shown in Fig.2 (b). The DTA peaks at 112.1 °C and 272.2 °C are due to the volatile of the free water in residual compounds and the endothermic effect caused by removal of crystal water. The corresponding TG curve shows continual weight decrease and weight loss rate is 14.36 %. Compared to pure KOH, the weight decrease continuously when temperature is over 397.4 °C and the corresponding TG curve corresponds to the weight loss rate of 5.82 %. On the basis of our previous results with NaOH treatment, this weight loss is attributed to condensation of the silanol groups<sup>3</sup>. Because KOH is not volatilized during the reaction, only evaporation of H<sub>2</sub>O obtained in the reaction of KOH and ZrSiO<sub>4</sub> induce weight loss.

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