Synthesis of Pt/mesoporous SiC-15 and its catalytic performance for sulfuric acid decomposition

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SI cycle, one of thermochemical processes, has been considered a promising process to produce hydrogen from water [1]. Presently, the stability of catalysts for sulfuric acid (SA) decomposition in a wide temperature range of 550°C to 850°C is one of technological barriers in the SI cycle: $H_2SO_4 \rightarrow SO_2 + 1/2O_2 + H_2O$. The mixed metal oxide catalysts were attempted for SA decomposition, but they are disintegrated via metal sulfate formation at the reaction temperature lower than the decomposition temperature of their respective metal sulfate (< 750°C). Therefore, metal oxide catalysts or metal oxide based catalysts are not practical system in the SI cycle. On the other hand, SiC supported Pt catalysts are stable at relatively low temperature (< 750°C), but the corrosion of active Pt metal on SiC supports are a main cause of deactivation at 850°C [2]. Here, mesoporous SiC (mSiC-15) was prepared using SBA-15 templates. SBA-15 was prepared with P123 block copolymer as reported [3]. PCS (polycarbon methylsilane, M.W.= ~800) was dispersed in heptane (150 g) and 2-butanol (1.2 mL). The SBA-15 particles were dispersed in the mixed heptane solution with PCS. After evaporating solvent, mSiC-15 was obtained by calcining the dried samples at 1300°C in an Ar stream for 2 h. The average pore size of the mSiC-15 (3.9 nm) was smaller than that of the SBA-15 (6.1 nm). TEM analysis shows that the morphology of the mSiC-15 is similar to that of the SBA-15 (Fig.1-a). After loading 1 wt% Pt on the mSiC-15 by an impregnation method using chloroplatinic acid, the morphology of the Pt/mSiC-15 catalyst was not much changed (Fig.1-b,c). Pt amounts of the pristine Pt/SBA-15 and Pt/mSiC-15 catalysts were 0.73 wt% and 0.80 wt% (ICP-OES analysis), respectively

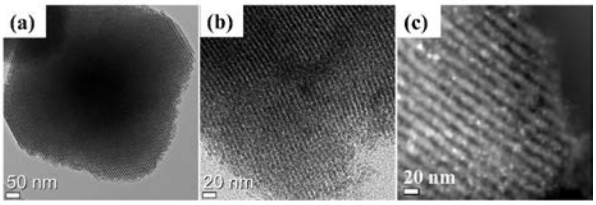

Fig. 1 TEM images of (a) mSiC-15 and (b) Pt/mSiC-15 and STEM image of Pt/mSiC-15.

The activity measurements of catalysts were performed with a quartz reactor (I.D.: 8 mm) at 650°C, 750°C and 850°C (GHSV: 76,000 mL/g_{cat}/h). The Pt/mSiC-15 catalyst was stable at all the reaction temperatures for 12 h, while the Pt/SBA-15 was steadily deactivated (Fig.2). After 6 h reaction at 850°C, the Pt amounts of the Pt/SBA-15 and Pt/mSiC-15 catalysts were 0.43 wt% and 0.69 wt%, respectively. After 12 h reaction at 850°C, the Pt amounts of the Pt/SBA-15 and Pt/mSiC-15 catalysts were 0.39 wt% and 0.68 wt%, respectively. The Pt components in mSiC-15 were a little corroded in the initial reaction time, but the Pt corrosion was prevented after 6 h at 850°C, while Pt in SBA-15 was steadily corroded. The loaded Pt components were less corroded at the lower reaction temperature.

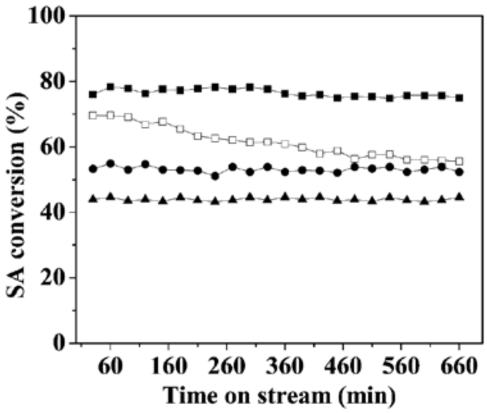

Fig. 2 SA decomposition with (□) Pt/SBA-15 (850°C), (■) Pt/mSiC-15 (850°C), (●) Pt/mSiC-15 (750°C), and (▲) Pt/mSiC-15 (650°C).

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