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論 文 要 旨(博士)

論文題目	Synthesis and Characterization of Sulfo-Functionalized Microporous and Mesoporous silica (スルホ基機能化ミクロ多孔性およびメソ多孔性シリカの合成と特性化)

Mesoporous silica has many physical properties that meet the requirements as an excellent separation material, catalyst and adsorbent. It provides a larger nanospace for reactions which allows the manipulation of larger molecules compared to zeolite. However, surface of mesoporous silica is covered with catalytically inactive siloxane and silanol groups. The surface can be functionalized with a wide range of functional groups through many methods to give desired surface properties for various applications. Surface functionalization with sulfo group gives mesoporous silica a strong acidic character. This hybrid can act as a solid acid to catalyze various reactions replacing hazardous liquid acids such as HF and H_2SO_4 to solve cost, environmental and technical issues. Besides, pore size is an important feature of porous materials especially in adsorption, separation and catalysis which involve specific shape and size selectivity and diffusion of molecules. Pore size control plays an important role in determining the characteristic and applications of a porous material. In vapor adsorption, adsorption potential can be enhanced by overlapping the adsorption potential in micropores. It is interesting to synthesize and characterization ordered mesoporous silica functionalized with sulfo groups due to its large potentials. To our best knowledge, no detail study about pore size control of sulfo-group functionalized mesoporous silica was reported yet, especially about the study of surface acidity of mesoporous silica by microcalorimetry of ammonia adsorption and the synthesis of microporous silica with ordered pore structure. In present study, synthesis and characterization of sulfo-functionalized microporous and mesoporous silicas with ordered pore structure was carried out. The samples were synthesized by the oxidation of thiol-group functionalized microporous and mesoporous silica which were obtained through hydrolysis and co-condensation of tetramethoxysilane (TMOS) and mercaptopropyl trimethoxysilane (MPTMS). The sulfo contents were controlled by regulating the amount of MPTMS whilst the pore size was controlled by the size of structure directing agents (surfactants). Surface acidities and chemisorptions of the samples were studied through adsorption microcalorimetry of ammonia. Both sulfo-functionalized microporous and mesoporous silica were successfully synthesized through this method and the pore sizes were effectively controlled in a desired range. Besides, a novel facile synthesis method of sulfo-group functionalized mesoporous silica was found during the study. This facile synthesis was carried out through 1-step surfactant removal and oxidation of thiol-functionalized mesoporous silica-surfactant complex by a temperature-controlled calcination. The sample was not only found to give better pore regularity, but also ammonia uptake and differential heats which are comparable to that of sulfo-functionalized mesoporous silica obtained by conventional method.