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The structural changes of silica gel by compressions up to 10 GPa

A. Arasuna (1), M. Okuno (1), X. Xue (2), T. Mizukami (1), M. Akaogi (3), M. Nozawa (3), H. Okudera (1), and S. Arai (1)

(1) Dept. of Earth Sciences, Kanazawa Univ, Kanazawa, Japan, (2) Inst. for Study of the Earth's Interior, Okayama Univ, Tottori, Japan, (3) Dept. of Chemistry, Gakushuin Univ, Tokyo, Japan

Structural change of TEOS-derived silica gel by compression up to 10 GPa has been investigated by XRD, Raman, IR, and ^{1}H and ^{29}Si MAS NMR spectroscopy, in order to gain insight into the compression behavior of silica gel. Silica gels were compressed under 5 and 10 GPa for 1 hr at room temperature by 6–8 Kawai-type multi anvil apparatus. All the measurements have been performed on the recovered samples.

Raman spectrum of silica gel has a broad band at around 460cm^{-1} assigned to a symmetrical Si-O-Si stretching mode. For samples compressed above 5GPa, this broad band loses its intensity in region of about 480cm^{-1} and becomes much sharper than that before compression. This observation indicates depolymerization of the network structure, decrease of the average Si-O-Si angle and narrowing of the angle distribution after compression. Such distinct spectral changes of silica gel could be related to the presence of Si-OH groups, which causes depolymerization of the structure. Silica gel contains a large quantity of molecular water and silanols. Both the Raman and ^{1}H MAS NMR spectra of silica gel showed decreasing water content with increasing pressure. ^{1}H MAS NMR spectra of silica gel contain a prominent narrow peak near 4.9 ppm and a very weak tail to higher frequency. The former may be largely attributed to relatively mobile molecular H_2O, and the latter to more strongly hydrogen-bonded SiOH/H_2O groups. For the compressed samples, the narrow peak near 4.9 ppm becomes much less intense, whereas the high-frequency shoulder (extending to about 10 ppm) becomes prominent, suggesting increased proportion of hydrogen-bonded SiOH/H_2O groups after compression.

Furthermore, ^{29}Si MAS NMR spectra for both the uncompressed and compressed silica gels revealed three peaks near -110, -101 and -92 ppm, which may be, respectively, attributed to Si of Q^4, Q^3 and Q^2 speciation (Si bonded to 0, 1 and 2 OH groups). The relative proportion of Q^3 vs. Q^4 is higher in the sample compressed at 10 GPa than the uncompressed sample, despite significantly lower total water content. This suggests that some of the molecular H_2O might have been converted to OH groups during compression, resulting in further depolymerization of the network structure.

In conclusion, this study has revealed significant irreversible structural changes for silica gels compressed at high pressure. These include significantly decreasing bulk water content and increasing proportion of strongly hydrogen-bonded water species, overall depolymerization and changes in the Q^n speciation distribution, and narrowing of the Si-O-Si angles and their distribution. These structural changes are expected contribute to the compression behavior of silica gels.