American Mineralogist, Volume 99, pages 1996–2007, 2014

Temperature-induced amorphization of Na-zeolite A: A view from multi-nuclear high-resolution solid-state NMR

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Abstract

Despite efforts to understand the amorphization mechanisms of zeolites upon heating and subsequent dehydration, little is known about the extent of Si-Al disorder and topological variations in both crystalline and amorphous phases during amorphization. In this study, we investigated the atomic structure and the extent of configurational disorder (e.g., Si-Al ordering) in Na-zeolite A and other dehydrated phases during their temperature-induced amorphization using multi-nuclear solid-state NMR. We also report the first multi-nuclear (^{17}O, ^{29}Si, and ^{27}Al) NMR spectra of the intermediate amorphous phases. ^{29}Si MAS NMR results confirm the prevalence of amorphous phases up to ~1073 K and variation in Q-species for the crystalline phases. The ^{27}Al quadrupolar coupling constant of the $^{[4]}$Al peak in Na-zeolite A and the intermediate amorphous phases increase with increasing temperature, which suggests an increase in the topological disorder associated with the structural distortion around $^{[4]}$Al. 2D ^{17}O 3QMAS NMR spectra resolve the crystallographically distinct Si-O-Al sites in Na-zeolite A and three types of oxygen linkages namely, Si-O-Al, Si-O-Si, and Al-O-Al in the intermediate amorphous phases, which provides an unambiguous experimental evidence for an increase in the Si-Al disorder during the amorphization of zeolite. The detailed structural changes in Na-zeolite A and other dehydrated phases at various temperatures provide insights into the structural changes of other aluminosilicates during amorphization, thereby highlighting the changes in Si-Al ordering.

Keywords: Amorphization, 3QMAS NMR, Na-zeolite A, Si-Al disorder