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Correlation between Hinckley index and stacking order-disorder in kaolinite

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Abstract

Hinckley index (Hi) can be used to characterize the crystallinity of kaolinite. Stacking order-disorder in kaolinite can considerably affect its crystallinity. However, the correlation between Hi and stacking order-disorder in kaolinite has not been reported thus far. Herein, the correlation between stacking order-disorder in kaolinite and Hi was investigated via experiments (XRD, IR spectroscopy, TG-DSC), molecular simulation, and structure refinement. First, we experimentally discovered that the stacking order-disorder in kaolinite changed the relative position between two adjacent structural layers, mainly affecting the interlayer forces. When the kaolinite layers are orderly stacked, the interlayer force is higher and the stacking lattice energy is lower. The lattice energy of kaolinite in different stacking states was simulated and analyzed using first-principles calculation. It was determined that the kaolinite layers are orderly stacked when two kaolinite layers have zero shift and disorderly stacked otherwise. Finally, through structural refinements, we proposed a new crystallinity index based on stacking order-disorder in kaolinite (crystallinity index based on stacking, CIS). CIS was well fitted to Hi ($R^2 = 0.986$), indicating that kaolinite crystallinity, characterized by Hi, is essentially the ratio of orderly stacking to total stacking (the sum of ordered and disordered stacks). Furthermore, measuring Hi is difficult when kaolinite crystallinity is poor; however, CIS can be used alternatively. This study of the crystallinity of kaolinite will have important significance for its industrial application.

Keywords: Kaolinite, stacking, order-disorder, structure refinement, crystallinity index, first-principles calculation