Identification of interstratified mica and pyrophyllite monolayers within chlorite using advanced scanning/transmission electron microscopy

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ABSTRACT

Interstratified clay minerals reflect the weathering degree and record climatic conditions and the pedogenic processes in the soil. It is hard to distinguish a few layers of interstratified clay minerals from the chlorite matrix, due to their similar two-dimensional tetrahedral-octahedral-tetrahedral (TOT) structure and electron-beam sensitive nature during transmission electron microscopy (TEM) imaging. Here, we used multiple advanced TEM techniques including low-dose high-resolution TEM (HRTEM), high-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM) imaging combined with energy-dispersive spectroscopic (EDS) mapping to study interstratified layers in a chlorite sample from Changping, Beijing, China. We demonstrated an interstratified mica or pyrophyllite monolayer could be well distinguished from the chlorite matrix by projected atomic structures, lattice spacings, and chemical compositions with advanced TEM techniques. Further investigation showed two different transformation mechanisms from mica or pyrophyllite to chlorite: either a 4 Å increase or decrease in the lattice spacing. This characterization approach can be extended to the studies of other electron-beam sensitive minerals.

Keywords: Chlorite, interstratified layer, HRTEM, STEM

INTRODUCTION

Clay minerals are phyllosilicates with the basic structure of an octahedral sheet (O sheet, marked by red dashed box in Fig. 1) sandwiched by two tetrahedral sheets (T sheet). The main constituent of the two T sheets is Si4+, which sometimes can be substituted by Al3+. Mg, Fe, and Al are common O sheet cations with substitutions by Cr, Ni, Mn, V, Cu, and Zn, etc. (Bailey 1988). There are differences in the interlayer (I sheet, marked by the red solid box in the right portion of Fig. 1). Some clays have no interlayer cation such as talc and pyrophyllite. Some have interlayer cations K, Na, and others countering the charge imbalance caused by the Al3+ substitution for Si4+ in the tetrahedral sheets, such as mica. Other clay minerals such as chlorite have brucite-like sheets (B sheet, marked by the red solid box in the left portion of Fig. 1) in the interlayer. Due to the above differences, the lattice fringes of pyrophyllite, mica, and chlorite are 9.5, 10, and 14 Å, respectively.

Mixed layer clays, as intermediate products of clay mineral transition and weathering, are ubiquitously present in nature. Since Hendricks and Teller (1942) first described partially ordered stacking layers in micas and other similar minerals, interstratified clays such as illite-montmorillonite, chlorite-vermiculite, illite-chlorite-montmorillonite have been reported in sedimentary rocks (Weaver 1956; Reynolds and Hower 1970; Bettisonvarga 1997; Banfield and Murakami 1998). Changes in the weathering degree and climatic conditions generate the pre-existing clay phases to transform into other species through a sequence of intermediate interstratified phases. Consequently, mixed layer clay minerals indicate the pedogenic processes that have occurred in the soil and provide a progressive structural and compositional transformation sequence of the solid-state reaction (Hong et al. 2012). Both regular and random 1:1 interstratifications of 24 Å clays (referring to a combination of contiguous 14 Å lattice and 10 Å lattice) were reported in weathering reactions or transition process (Środoń 1999). Such mixed layer clays include chlorite/mica (Lee and Peacor 1985; Cruz 2001), chlorite/talc (Schreyer et al. 1982; Ahn et al. 1988), chlorite/pyrophyllite (Kong et al. 1990, 1992, 2000) and chlorite/biotite.