Phase relations among smectite, R1 illite-smectite, and illite

HAILIANG DONG,1 DONALD R. PEACOR,1 AND ROBERT L. FREED2

1Department of Geological Sciences, University of Michigan, Ann Arbor, Michigan 48109-1063, U.S.A.
2Department of Geology, Trinity University, San Antonio, Texas 78212, U.S.A.

ABSTRACT

A variety of smectitic and illitic clays were studied by TEM and AEM, following expansion by L.R. White resin, to define phase relations for clay minerals undergoing diagenesis and low-grade metamorphism. Samples included a prograde shale sequence from the Gulf Coast, a hydrothermal bentonite from Zempleni, Hungary, and shales from the Nankai Trough, Japan, Michigan, and the Welsh sedimentary basin. All samples were dominated by various proportions of only three kinds of clay minerals: smectite having no 10 Å layers, R1 illite-smectite (I/S) (50% illite), and illite with only small proportions of smectite-like interlayers; mixed-layer phases with intermediate ratios of I/S were observed only as minor components of other layer sequences. Lattice fringe images show that the common layer spacing of R1 I/S is 21 Å and that it has 0.7–0.8 K pfu; its properties are not an average of those of smectite and illite, which is consistent with the uniqueness of the R1 I/S structure.

A prograde sequence of clay mineral transitions in the studied samples can be characterized by five stages with different combinations of the three major phases (i.e., smectite, R1 I/S, and illite) corresponding to different grades. The sequence from low to high grade is (1) pure smectite, (2) smectite with small proportions of discrete R1 I/S and illite, (3) R1 I/S with small proportions of smectite and illite, (4) illite with some R1 I/S and smectite, and (5) illite. Common exceptions to this scheme are inferred to be caused by the inherent metastability of all phases, in occurrences that are rate or path dependent.

INTRODUCTION

It is generally accepted among clay mineralogists, in part from the influence of the classic paper of Hower et al. (1976), that the sedimentary smectite-to-illite transition occurs through a sequence of illite-smectite phases (commonly denoted I/S), including smectite-rich R0 I/S, R1 I/S, R2 I/S, and R3 I/S, and illite-rich I/S, with continuously variable ratio in the proportions of smectite and illite-like layers. According to this model, the relative proportion of illite interlayers increases until no expandable layers remain, during progressive prograde diagenesis and low-grade metamorphism of pelitic rocks. This model implies that all I/S with relative numbers of illite layers from 0 to 100% are likely to occur. This concept led in part to the implication inherent in the work of Hower et al. (1976) that the smectite-to-illite transition could proceed layer by layer, i.e., individual smectite layers may be progressively replaced by illite layers with loss of Si, Na, and H₂O and gain of K and Al during the transformation process.

However, several recent TEM studies indicate that R1 I/S with 50% I is relatively abundant, whereas other kinds of mixed-layer I/S phases (e.g., R0 I/S, R1 I/S with >50% I, R3 I/S, etc.) are rarely observed (Ahn and Peacor 1989; Veblen et al. 1990; Jiang et al. 1990a; Dong and Peacor 1996; Sears, personal communication). Ahn and Peacor (1989) observed only R1 I/S, but not R2 or R3 I/S, in a Gulf Coast sample (5500 m depth), using the overfocus conditions recommended by Guthrie and Veblen (1989a, 1989b, 1990). The presence of R1 I/S at the 5500 m depth is consistent with XRD data on the same sample (Ahn and Peacor 1989). They further concluded that caution should be exercised in estimating the relative proportion of R1 I/S based on TEM images only, because even though ordering exists over the whole image, it may be observed in rather limited areas with great difficulty, largely because of variable orientations of layers relative to the electron beam. This conclusion implies that R1 I/S observed in TEM images represents a minimum of that actually present in original rocks.

Ahn and Peacor (1986) proposed a dissolution-precipitation model for the smectite-to-illite transformation. This model suggests that a reactant, smectite, is structurally disarticulated and individual chemical components are transported through fluid and reorganized and precipitated as a new phase. Jiang et al. (1990a) noted that the chemical composition of R1 I/S is not a weighted average of those for smectite and illite, indicating that R1 I/S is a unique phase. The observation by Jiang et al. (1990a) is consistent with neoformation of all layers, i.e., even the 2:1 structure units associated with expandable interlayers must have compositions richer in Al and poorer in