

## Changes in the structures of mixed-layer illite–smectite during flooding of terrigenous oil reservoirs

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### Abstract

A difference-spectrum method is proposed for the qualitative assessment of changes of illite–smectite structures accompanying the flooding of oil reservoirs. The method permits one to get an open system and reduce the application of procedures based on Markov's chain formalism. A computer simulation is made to obtain spectra by subtracting the spectrum of an ethylene glycol-saturated sample from the spectrum of an air-dried preparation throughout the entire range of concentrations of illite and smectite components with a short-range order factor  $R = 0$  or  $R = 3$ .

It has been established that only in the presence of filtration are the maximum and minimum of the spectra in the range of 12.5–9.4 Å complicated by a number of local extrema, whose position is specified by the structure of intermediate phases. The flooding process first involves mixed-layer phases with  $R = 0$ , leading to a partial segregation of the structures into phase with one and two networks of interlayer H<sub>2</sub>O. When the secondary mica particles break, phases with  $R = 3$  appear along the boundaries of nanoblocks, first only with 1 H<sub>2</sub>O and then only with 2 H<sub>2</sub>O in labile interspaces. Their coexistence with the phases  $R = 0$  in the sample proves the existence of percolation effects due to two-phase filtration in the porous medium. The fully flooded reservoir is always dominated by a mechanical mixture of illite–smectite phases of different nature with  $R = 0$  and with different ratios of components. Transformation of mica that can drastically reduce oil production begins long before the appearance of flooding zones, which are revealed by standard logging methods.

*Keywords:* oil production; mixed-layer illite-smectite; micas, X-ray diffraction; computer simulation

### Introduction

Long-term oil-field development in Tatarstan and other regions has revealed a number of phenomena not explainable by the conventional concepts of fluid dynamics. Injection of fresh water from near-surface sources into a reservoir usually leads to a reduction in the filtration capacity of terrigenous reservoirs, especially those of high salinity. In the producing Devonian horizon of Tatarstan, this process can have disastrous effects. It has been found that as the density of reservoir water reduces to 1.09 g/cm<sup>3</sup>, the oil-bearing reservoir of salinity > 5% sometimes stops producing oil (Muslimov, 2003). Similar phenomena are observed in a number of locations in West Siberia. It has been found that the dominant factor responsible for the reduction of oil extraction is not the swelling effect but the counter electroosmotic flow produced

by mica nanoblocks fixed in the pore space and having a high surface charge which cannot be compensated for in the absence of K<sup>1+</sup>. Such blocks are formed during the inverse transformation of secondary micas to the initial smectite with decreasing concentration of the reservoir solution under the influence of micflora, accompanied by the formation of mixed-layer illite–smectite phases (Krinari and Khrumchenkov, 2008a,b, 2011). Of greatest interest for applications is to identify the earliest stages of water flooding of reservoirs based on mineralogical criteria, and this was the main objective of the study. However, the kinetics of the process remains largely unexplored since no methods have been developed to analyze the real structure of the products of the inverse transformations of secondary micas. A number of methods have been proposed to prevent the reduction of filtration, but, without considering the specificity of man-made changes in the composition of the clay component, they are difficult to optimize for the development of particular locations; this requires different approaches and techniques for analyzing the structure of newly formed phases.

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## Experimental data and methods of studying illite–smectite structures

The experimental material was a fraction  $\leq 2.5 \mu\text{m}$  extracted from rocks and differently flooded producing reservoirs  $D_1$  and  $D_0$  of the Romashkino oil field of Tatarstan, and some locations in West Siberia. X-ray diffraction spectra were taken from oriented, air-dried, ethylene glycol-saturated, and sometimes heated preparations using the scale of inverse interplanar spacings of dimension  $1/\text{\AA}$  at intervals of  $0.0008 \text{ 1/\AA}$ . The recording range was from 50 to  $2.48 \text{ \AA}$ .

The alternation of layers or packets along the  $c^*$  crystallographic axis specifying the direction of the texture axis of the oriented X-ray diffraction sample is reflected in the recorded basal diffraction pattern—00L reflections. They are produced by packets or combinations of packets of different thicknesses corresponding to the basal interplanar spacings  $d_{00L}$  of mica, chlorite, and smectite in the natural, heated, or ethylene glycol-saturated states.

The main and well-developed method for studying mixed-layer phases is fitting, in which the parameters of theoretical basal diffraction spectra of clay minerals are chosen so as to fit experimental curves (Drits and Tchoubar, 1990), in particular, for different preparations of samples and composition of exchange cations. This allows identification of phases from alternations of different layers and packets (Drits et al., 2004; Sakharov et al., 1999). The fitting method is based on the theory of Markov chains, where the probability characteristics for any sequence of layers are uniform and have the same statistical dispersion. The conditions are completely fulfilled in objects formed during catagenesis in closed systems or poorly permeable sediments (Solotchina, 2009), where the mineral structure is in relative equilibrium with the environment. In this case, the statistical structural characteristics of the phases have a strict physical meaning, which cannot be universal for all sedimentary rocks. Since the inverse transformation of secondary micas causes enormous mechanical tension (Krinari et al., 2001) and since sandstones can undergo dispersion with removal of part of the products from the reaction zone, the statistical homogeneity of mixed-layer phases is violated, with partial transformation of the clay fraction to a mechanical mixture of layered silicates of nonequilibrium structure and different genesis. These minerals can no longer be considered as a set of phases with constant and statistically average parameters since the coefficients of hydrodynamic or statistical dispersion of these parameters are functionally related to the kinetic parameters of diffusion and filtration (Khrumchenkov, 2003). In studies of the inverse transformation, the fitting procedure may not always be correct because the system becomes open to one or another extent and does not remain constant and its final form depends on both the rate of decomposition and the rate of displacement of the new phases.

The direct decomposition of experimental diffraction patterns into theoretical ones, as proposed by Lanson (1997) is not quite correct for objects containing several different phases with 1  $\text{H}_2\text{O}$  and 2  $\text{H}_2\text{O}$  packets. Their basal diffraction in air

can produce a continuous maximum in a wide region of reciprocal space, but the spectra of the phases saturated with ethylene glycol become identical (Drits and Sakharov, 1976). There may be combinations of different heterogeneous structures that give very similar spectra. One more experimental parameter depending on both the structure of the phases and the number of  $\text{H}_2\text{O}$  networks is needed which can be modeled. It is also difficult to distinguish homogeneous structures of micas and chlorites from the occurrence of rare continuous labile gaps between layers, i.e., from the beginning of the formation of mixed-layer patterns.

## Method of assessing the structural appearance of illite–smectite in open systems

Our task does not require a numerical evaluation of the content of the individual phases or the probabilities of alteration of 2:1 layers with different interlayer spaces in them because the structural appearance of the object is continuously changing during the transformation. It is sufficient to fix the appearance or disappearance of certain structures that can be interpreted as indicators of certain stages of the transformation. Then, the object can be considered an assembly of particles capable of making a contribution to the diffraction and corresponding to the sequence of changes in the estimated characteristics of the structure in the section of the flooded reservoir.

A difference spectrum method is proposed in which the diffraction curves from an air-dry (Fig. 1a) and ethylene glycol-saturated samples are normalized to the  $\approx 7 \text{ \AA}$  reflections of the phases without swelling packets (see Fig. 1b), and the second spectrum is subtracted from the first (see Fig. 1c). The difference spectrum makes no contribution to the diffraction from mica, kaolinite, and chlorite, and contains information only on all mixed-layer phases with labile interlayer spaces, regardless of the ratio of their components. In addition, it better reflects the phase structure due to extraction of only the useful signal. Such a spectrum is almost independent of the composition of 2:1 layers and the apparatus function, and its main part is the straight line. The height of this “zero line”, denoted as  $L_z$ , with respect to the ordinate axis for the “air-dry minus ethylene glycol” spectrum in the range of  $0.05\text{--}0.15 \text{ 1/\AA}$  specifies the ratio of the intensities at two extrema points:  $d_{\text{max}}$  and  $d_{\text{min}}$ . Their position and height in as  $L_z$ , are determined by the concentration of mica (pM), smectite (pS), the number of networks of  $\text{H}_2\text{O}$  molecules in labile packets, and the short-range order factor  $R$ .

The Sybilla software developed by Chevron was used to calculate theoretical spectra of disordered (with a short-range order factor  $R = 0$ ) and ordered ( $R = 1; 2; 3$ ) illite–smectite phases for different ratios of components:  $10 \text{ \AA}$ , mica;  $12.4 \text{ \AA}$ , smectite with one water network (1  $\text{H}_2\text{O}$ ) between the layers;  $14.4 \text{ \AA}$ , smectite with two networks of water (2  $\text{H}_2\text{O}$ ); and  $16.8 \text{ \AA}$ , ethylene glycol-saturated smectite and. The values of  $d_{001}$  for smectite were taken, respectively, with allowance for that the prevalence of monovalent or divalent cation exchange.

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