

# Thermodynamic model of formation of carbonates and uranium mineral phases in lakes Namshi-Nur and Tsagan-Tyrm (*Cisbaikalia*)

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## Abstract

The Tazheran lakes are located compactly in the small Tazheran steppe area. Their bottom sediments are predominantly various calcite–dolomite carbonates, and their waters are rich in uranium. The studies have shown that the main process in these lakes is chemogenic carbonate precipitation with the participation of carbon dioxide formed through the bacterial destruction of organic matter. For thermodynamic modeling of the composition of bottom sediments, we chose two lakes with different basic parameters. Calculations were made for the 15-component heterogeneous system  $\text{H}_2\text{O}-\text{Na}-\text{Ca}-\text{Mg}-\text{K}-\text{Sr}-\text{Ba}-\text{Si}-\text{Al}-\text{Cl}-\text{C}-\text{S}-\text{Fe}-\text{U}-\text{Mn}$  including particles in the solution, minerals, and gases at 25 °C and 1 bar. As starting information, we used the obtained analytical data on the natural composition of waters and bottom sediments. The results show that calcite–dolomite carbonates are predominant in the bottom sediments and the destruction of organic matter results in reducing conditions. This confirms the hypothesis of the formation of mineral phases of U(IV) during diagenetic processes in the bottom sediments of the studied lakes.

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**Keywords:** bottom deposits of lakes; mineralogy and geochemistry of authigenic carbonates; mineral phases of uranium; thermodynamic modeling; Cisbaikalia

## Introduction

Formation of the chemical composition of authigenic minerals of bottom sediments of water reservoirs is an object of great researcher's attention. Many of its issues, however, are still unclear because the process is controlled by many factors. Of special interest are biogeochemical processes of precipitation of suspensions in waters, followed by the formation of authigenic minerals with variable-valence elements, which can serve as indicators of genesis conditions (Lein and Ivanov, 2009; Lisitsyn, 2014). The microbiologic factors governing sedimentation in limnological systems are also yet to be elucidated (Gas'kova et al., 2017; Gaskova et al., 2017; Zavarzin, 2004).

Many researchers associate the chemical precipitation of uranium during sedimentogenesis with its reduction below the oxygen saturation zone (Evseeva et al., 1975; Vinogradov, 2013). However, available data on the stability of carbonate complexes of uranyl ion in reducing conditions in the presence

of hydrogen sulfide and ferrous iron raise doubts as to the possibility of this process. Today, many scientists (Fredrickson et al., 2002; Komlos et al., 2008a,b; Sani et al., 2004; Ulrich et al., 2009; Wang et al., 2013) believe that the solid phase  $\text{UO}_{2+x}$  cannot be stable in lacustrine bottom sediments in the presence of oxygen, manganese, and iron, because Mn(III/IV) and Fe(III) oxides must be its effective oxidants, as follows from the redox potential of U(IV)/U(VI). Other researchers (Burgos et al., 2008; Schofield et al., 2008; Sharp et al., 2011) report on U(IV) oxidation involving bacteria. It was also shown that oxidized Mn and Fe species and reduced U species can coexist in bottom sediments for a long time (Bernier-Latmani et al., 2010; Moon et al., 2009; Qafoku et al., 2014; Sharp et al., 2011). The fact is that the mobility of uranium species is determined not only by the redox conditions and pH of water but also by biogeochemical processes (i.e., the presence of organic matter (OM) and its interaction with iron and manganese oxides/hydroxides) (Andersson et al., 2001; Fredrickson et al., 2002; Koch-Steindl and Pröhl, 2001). Sorption of uranium complexes on iron hydroxides is significantly reduced in the presence of OM and/or competing cations, such as  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$  (Belli et al., 2015). Other

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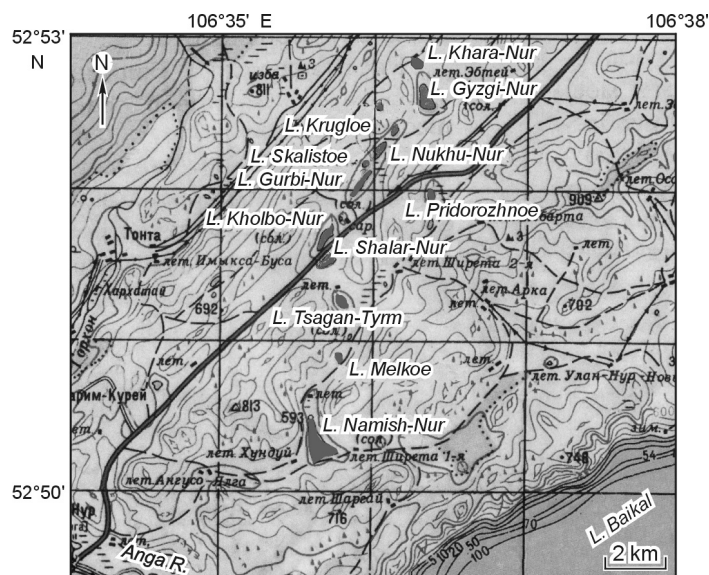


Fig. 1. Map of the location of the Tazheran lakes.

studies showed that the concentration of U(VI) in the aqueous phase drastically decreases during bacterial processes because of the formation of insoluble U(IV) compounds (Newsome et al., 2014, 2015). It was established that U(IV) in the geologic environment forms molecular complexes associated with biomass (Bargar et al., 2013; Bernier-Latmani et al., 2010). In almost all the above articles, the object of study was pore solutions but not the mineral phases of bottom sediments. Elucidation of geochemical reactions responsible for the formation of uranium phases in lacustrine bottom sediments requires thermodynamic calculation, which must be based on the natural contents of elements and the real mineral composition of the sediments.

A thermodynamic approach has been successfully used for several decades both to elucidate genesis of minerals in particular lake systems (Gaskova et al., 2011, 2015, 2017) and to elaborate theoretical models (Li et al., 2016; Ryzhenko et al., 2016) and models for strategic environmental management (Kwok et al., 2008).

The aim of this research is: (1) to carry out isotope studies of lacustrine bottom sediments and confirm the biochemical (with the participation of microorganisms) mechanism of sedimentation; (2) to perform a thermodynamic modeling and to substantiate the quantitative relationship between the compositions of the lake waters and the bottom sediment minerals; and (3) to elucidate the mechanism of formation of authigenic minerals of U(IV) in the bottom sediments.

### Objects of study and methods

The Tazheran lakes (Cisbaikalia) have been studied for many years and provided much factual information about the composition of their components (Chabaux et al., 2011; Sklyarova et al., 2002; Solotchina et al., 2011; Strakhovenko et al., 2015a; Vosel et al., 2015) (Fig. 1). The systems of small

lakes located compactly in the area are convenient to use as model natural objects, because this permits minimization of the number of factors determining the geochemical specifics of the water/bottom sediments/rock interaction in the watershed areas. The hydrocarbonate or hydrocarbonate–sulfate sodium–magnesium composition of the waters is the result of the weathering of rocks containing feldspars as the main component (gneisses, massive-crystalline alkaline rocks, and granites) from the watershed areas. The amounts of a particular mineral relative to the total mass of the terrigenous fraction can be different in the lakes, whereas its chemical composition is constant and corresponds to the composition of this mineral in the rocks of the watershed areas. The high alkalinity of the lake waters favors the mobility of sodium uranyl–carbonate compound ( $\text{Na}_4[\text{UO}_2(\text{CO}_3)_3]$ ).

We performed a comprehensive study of the inorganic and organic materials of the lacustrine bottom sediments, i.e., their morphology and phase and chemical compositions, using scanning electron microscopy (SEM) (MIRA 3 TESCAN microscope), X-ray diffraction (XRD) (ARL X'TRA diffractometer with  $\text{CuK}\alpha$  radiation), and IR spectroscopy. The contents of U isotopes in the samples were determined by high-resolution semiconductor gamma spectrometry with a well semiconductor detector (SCD). The elemental composition of the lake water and bottom sediments was determined by flame and electrothermal atomic-absorption spectroscopy (AAS) (Solaar M6, Thermo Electron Corporation). All AAS analyses were carried out according to the certified NSAM techniques. The accuracy and validity of the obtained analytical data were confirmed by analysis of standard samples. Its results agree with the certified contents of their elements.

Analysis for stable  $^{18}\text{O}$  and  $^{13}\text{C}$  isotopes in carbonates was carried out on a Finnigan MAT 253 mass spectrometer at the Analytical Center for Multielement and Isotope Analyses of the Institute of Geology and Mineralogy, Novosibirsk, using the Continuous Flow technique and a GasBench II sample

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