# THE BARITES GENESIS OF THE DERYUGIN BASIN (THE SEA OF OKHOTSK)

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In order to reveal the genesis of barites, the authors identified content and distribution regularities of rare-earth elements in barites from the Deryugin Depression, the Sea of Okhotsk. The obtained data show that a hydrothermal component does not affect the formation of the barites. Sea water is the source of the rare-earth elements, and their composition apparently corresponds to the composition of cold seeps.

*Keywords: barites, rare earth elements, the Deryugin Basin, the Sea of Okhotsk*

## INTRODUCTION

Authigenic marine barite formations ($BaSO_4$) occur in various geological settings, and their formation is associated with different physical and chemical, as well as biological processes (Bogdanov, 2006; Gonzalez-Munoz et al., 2012; Hein et al., 2007; Monnin et al., 2003; Vanneste et al., 2013). In the marine environment, there are several different ways to form barite, which are reduced to the interaction between water masses enriched with barium (fluid) and sulfate ion (sedimentation conditions). The barium sulfate precipitate is resulted from the solution supersaturation. (Hein et al., 2007; Paytan et al., 2002). Fluids (sea water, pore waters or hydrothermal solution) and medium (water column, bottom surface, sediments, cold seeps or hydrothermal ore genesis conditions) affect geochemical characteristics of barite (Griffith, Paytan, 2012). There are four general regimes of barite formation: (1) hydrogenic or pelagic — in which barite is formed in the water column during decomposition of the barium enriched organic matter; (2) hydrothermal — in which barite precipitates due to mixing of the Ba-enriched hydrothermal fluids and sea water near the bottom; (3) diagenetic — in which barite is formed in thick sedimentary layer in the result of post-sedimentation diagenetic processes; (4) in which barite is resulted from evolution of fluids or Ba−enriched cold seeps in sedimentary basin, which

are delivered to the bottom surface by tectonic and hydrological processes not associated with volcanic or hydrothermal activity (Binns et al., 1997; Brumsack, 1986; Griffith, Paytan, 2012; Hein et al., 2007; Naehr et al., 2000; Torres et al., 1996).

Variations of chemical microelement and isotope compositions of carbon, oxygen and sulfur in sea barites are used to make paleoceanography reconstructions for determining primary productivity in the past; the role and potential value of bacteria in barite deposition, as quantitative parameters that control the barite preservation in sediments; to define effect of diagenesis on the barite geochemistry (Griffith, Paytan, 2012; Martin et al., 1995). However, all the conclusions are possible only after accurate definition of these formations genesis. At present, in terms of the genesis, the barite mineralization of the Deryugin Basin (the Sea of Okhotsk) is the topic to discuss.

Barites from the Deryugin Basin for the first time were dredged in 1981 by B.I. Vasilyev and identified by M.I. Lipkina in 1987 (Astakhova et al., 1987). Later, significant amount of expeditions and laboratory studies were performed (Derkachev et al., 2000; Greinert et al., 2002). The obtained data allow estimation of the area of the barite-bearing surface of the bottom in the Deryugin Basin as a potentially large barite deposit, with predicted resources of 5 million tons (Aloisi et al., 2004). The question of the

Okhotsk Sea barite structures origin and its forming matter sources is still open. Some researchers believe that the barites formation is associated with low-temperature hydrothermal activity (Astakhov et al., 2017; Akhmanov et al., 2015), others associate their formation with cold gas-fluid emanation (cold seeps) (Derkachev et al., 2002; Aloisi et al., 2004; Greinert et al., 2002). In authigenic mineral formation, the composition of new formations must inherit features of concentration and distribution of various elements corresponding to one of these processes.

At present, during the data on rocks genesis interpretation, we use the data on rare-earth elements composition (REE, lanthanides). Due to its chemical properties this group of metals is the unique one and under various physicochemical parameters of the marine environment it forms special composition reflecting formational conditions. This ability is resulted from the REE chemical properties in natural processes, and allows inhering lanthanide compositions of the initial material.

This paper reveals the REE distribution in the Deryugin Basin barites in order to define the source of the matter (hydrothermal or cold seep) composing these formations.

## MATERIALS AND METHODS OF INVESTIGATION

The results were obtained using the data on the barite samples dredged by a bottom grab in the Deryugin Basin (the Sea of Okhotsk) (fig. 1) in 54th voyage of the R/V «Akademik MA Lavrentiev» in 2011 at the stations Lv54−7−4 (53.988° N, 146.310° E, depth — 1460 m), Lv54−35 (54.004° N, 146.292° E, depth — 1492 m). During interpreting the data on the REE composition in barites we used these chemical elements' content in bottom seawater, sampled at the 1444 m depth at Lv54−7−1 station (53.985° N, 146.309° E) in the same voyage. Mineral composition of barites was studied in the Center for Collective Use (CCU) «Primorsky Center of Local Elementary and Isotope Analysis» by FEGI FEB RAS using the powder microdiffractometry method (analysts N.V. Gruda) on a MiniFlex II diffractometer by RIGAKU (Japan) and scanning electron microscopy ( SEM) (analysts A.V. Poselyuzhnaya and A.E. Krasnenko) on an electron microscope Tescan Lyra3 (the Czech Republic).

The REE concentrations in barites were defined using the inductively coupled plasma mass spectrometry (ICP−MS) method in the above-mentioned CCU. This method is known as the most accurate method for determining of the low REE concentrations in various geological samples. The analysis was carried out using liquid samples in order to obtain their maximum representativity and homogeneity. It turned out that barites consisting of more than 95% of barium sulphate ($BaSO_4$) refer to

sparingly soluble minerals. The main ways of their decomposition include fusion and agglomeration with various agents.

Methods for the REE in barites determination are presented in several papers dealing with elemental and isotopic analysis (Guichard et al., 1979, Martin et al., 1995). For example, the neutron-activation method may not correspond to the required accuracy, which is so important in interpretation of the REE composition data, and also does not allow to determine the entire spectrum of elements. In determination of the REE in barites using the ISP−MS method, the analytical isotopes Sm, Eu and Gd are subjected to the barium strong influence, due to multiple excess of Ba concentration over the REE content, which can lead to data corruption and the results erroneous interpretation. For example, the paper of (Baioumy, 2015) on barites of the Oasis of Bahariya (Egypt), contains the data on anomalously high concentrations of these three REE, which, in our opinion, highly likely does not correspond to reality.

The main problem to be solved for the accurate REE and especially Eu determination, using the ISP−MS method, is to overcome the effect of Ba high concentrations in analyzed solutions. This was achieved by sample decomposing in Teflon crucibles and subsequent precipitation of Ba in the form of its sulphate by adding a stoichiometric amount of $H_2SO_4$ to remove the Ba excess from the solution (analyst Shcheka Zh.A.) The REE definition was carried out
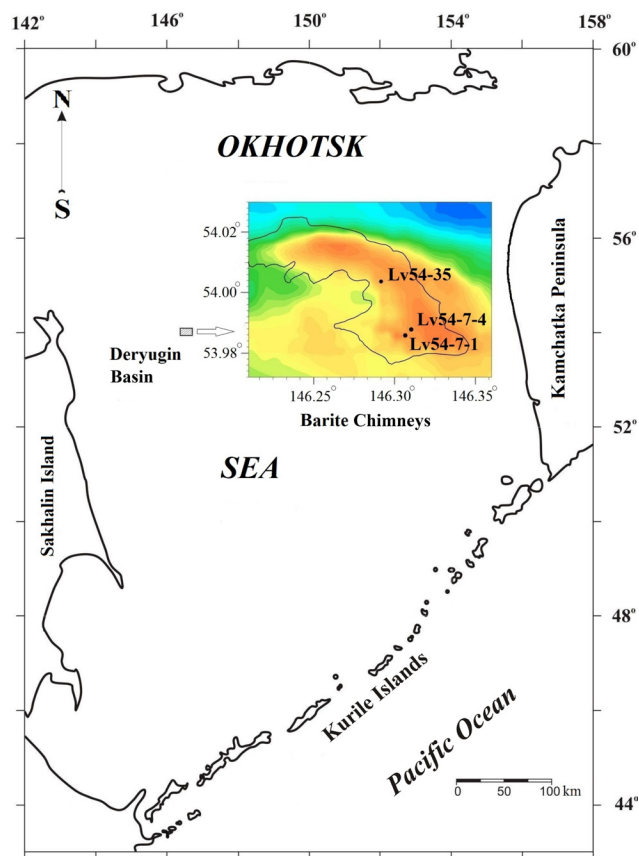


**Fig. 1.** Map of sampling area for barites and seawater.

on the Agilent 7700x mass spectrometer (Japan) using the device hardware (a collision cell for eliminating interference, filled with helium) and, if necessary, by the obtained results mathematical correction (Elovsky, 2015). The main advantage of this technique is a multiple decrease in the Ba/Eu ratio, as well as a low dilution factor of 150 (in routine methods, this factor is usually 5000 or even more), which did not significantly reduce the concentration of the detectable elements in the solution. At the same time, the process of precipitation of barium could cause an underestimation of the obtained results related to the sorption of REE from the solution on newly formed barite crystals.

We verified the possible REE sorption under these conditions using the «introduced-found» technique. The paper shows that sorption does not occur and the technique can be applied to quantitative estimation of REE (Blokhin et al., 2016, 2017).

The REE in a sea water sample were also determined using the ICP−MS method on an Agilent 7700x mass spectrometer with pre-concentration of rare-earth elements on an Lewatit® TP 207 ion exchange resin (Elovskiy and Mikhaylik, 2016).

## RESULTS AND DISCUSSION

Based on the data on powder microdiffraction and SEM (fig. 2, tab. 1), the body is composed of nearly pure barite. The rhomboid morphology of barite crystals (fig. 3) is typical for diagenetic bodies in various regions of the World ocean (Griffith, Paytan, 2012). The obtained data show that the REE content for barites varies from n $10^{-1}$ mg/kg for light to n $10^{-3}$ mg/kg for medium and heavy REE in barites from the Deryugin basin, while in the seawater sample the content varies from ng/l for light REE to n $10^{-2}$ ng/l for medium and heavy REE (Table 2). The distribution of slate (PAAS) of normalized REE barites and the benthal (depth of 1444 m) seawater in the Deryugin basin (fig. 4) shows the change in the ratio of light REE to heavy $LaSn/LuSn$ (cH — normalized by slate) within 0.54−0.96, which is close to benthal water in the Deryugin basin (0.63).

In hydrothermal barites, the index value is greater than 1 and the increase in heavy lanthanides in relation to the light lanthanides has not been revealed (Dubinin, 2006). Such an REE composition in the barites, which we studied, reflects the distribution of dissolved REE forms in the benthal seawater in the Deryugin basin.

The value of the cerium anomaly (Ce*), calculated as $Ce/Ce_{paas}/((0.5*La/La_{paas}) + (0.5*Pr/Pr_{paas}))$ is less than 1, also inherited from seawater (fig. 4). The depletion of cerium, which we studied in the Okhotsk baritic bodies, most likely gives evidence for a rapid growth rate of these bodies and is inherited from solutions (seeps?), which form these ore bodies. A positive (> 1) cerium anomaly in the ocean is characterized by hydrogenogenic ferromanganese bodies in which cerium accumulates due to oxidative sorption just in the water column on suspended Fe−Mn oxyhydroxides (Dubinin, 2006).

The magnitude of the europium anomaly (Eu*) varies from 0.20 to 0.59 ($Eu* = Eu/Eu_{paas}/((0.5*Sm/Sm_{paas}) + (0.5*Gd/Gd_{paas}))$). This shows that the hydrothermal substance is not involved into the formation of barites, because the vast majority of marine bodies
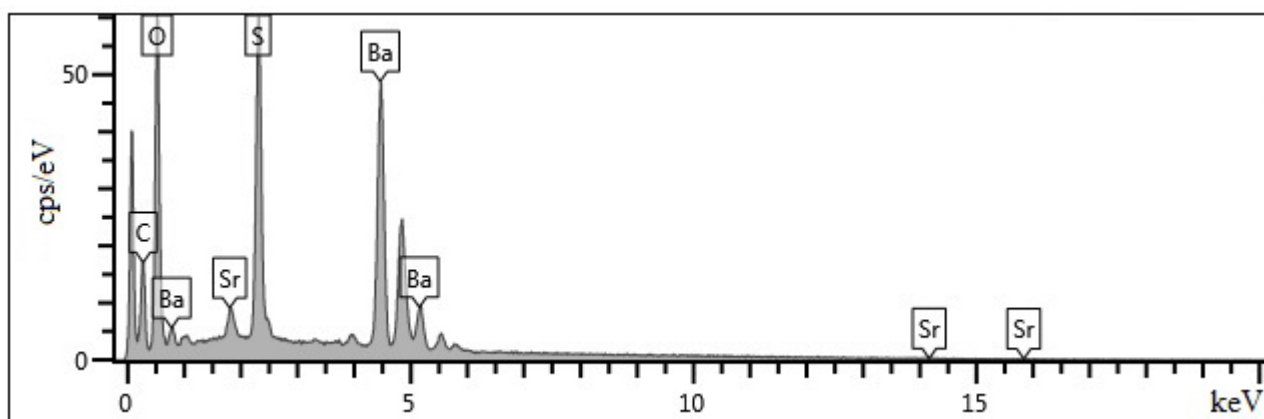


**Fig 2.** Qualitative chemical composition of barite mineralization in the Deryugin Basin on SEM data.

**Table 1.** Quantitative chemical composition of the Deryugin Basin's barite mineralization on SEM data.

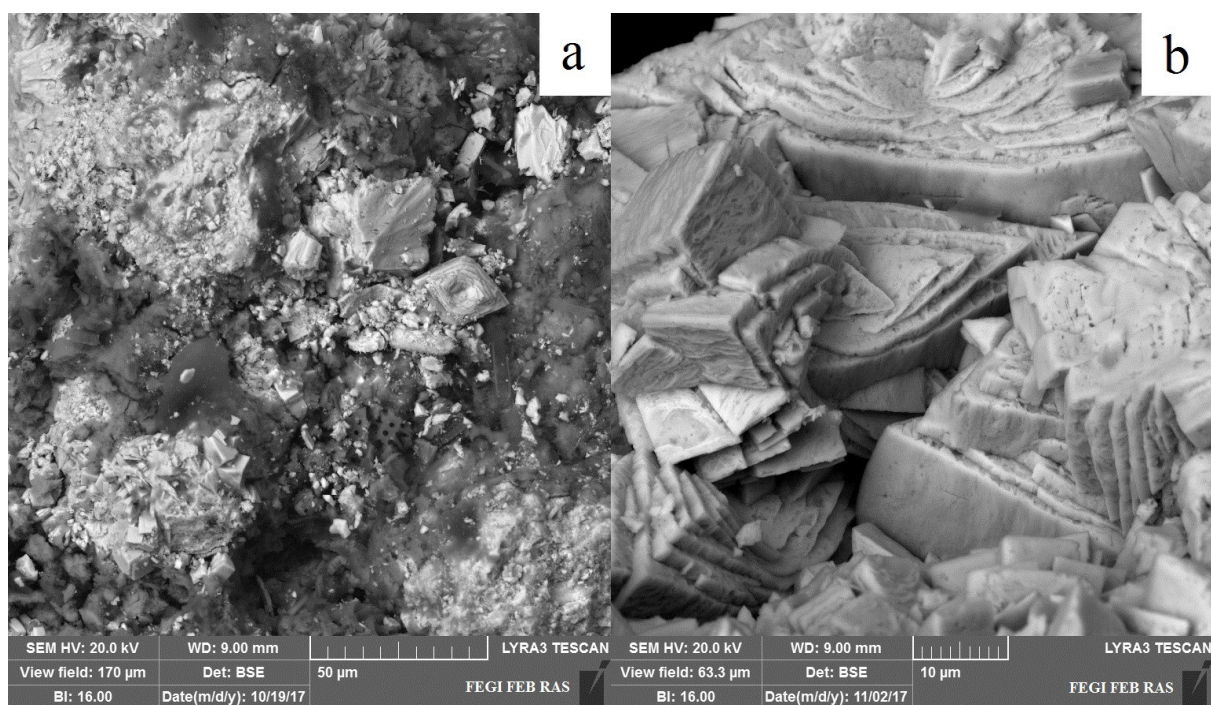| Element | Wt.% | Sigma Wt.% | At. % | Oxide | Wt.% oxide |
|---|---|---|---|---|---|
| O | 29.15 | | 67.27 | | |
| S | 15.00 | 0.11 | 17.27 | $SO_3$ | 37.45 |
| Sr | 2.84 | 0.14 | 1.20 | SrO | 3.36 |
| Ba | 53.01 | 0.20 | 14.25 | BaO | 59.19 |
| Total amount: | 100.00 | | 100.00 | | 100.00 |

**Fig. 3.** Barite mineralization of the Deryugin Basin on the increased SEM

**Table 2.** The REE content in barites (mg/kg) and concentration in sea water (ng/l) from the Deryugin Basin.

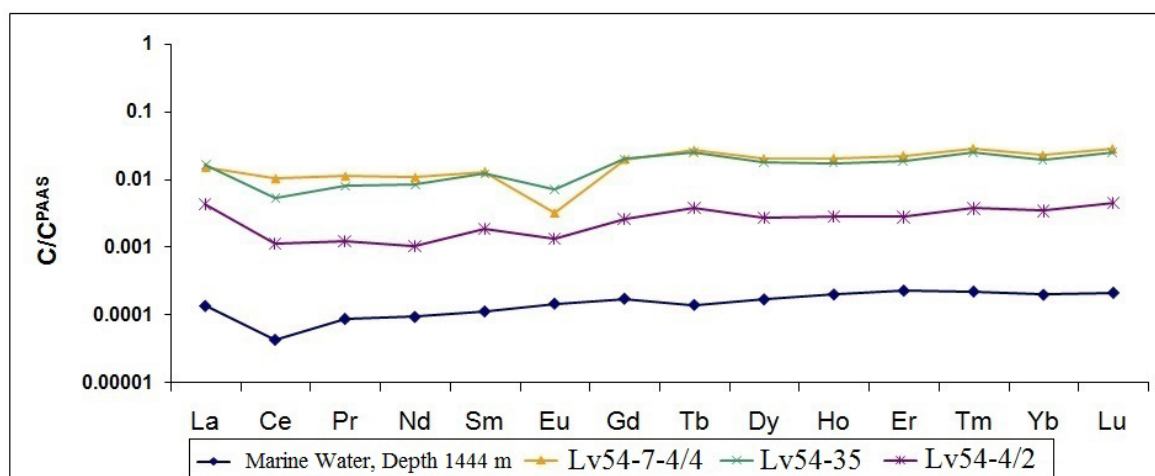| Element | Lv54−7−4/4 | Lv−54−35 | Lv54−4/2 | Lv54−7−1 |
|---------|-----------|----------|----------|----------|
| La | 0.574 | 0.626 | 0.163 | 5.220 |
| Ce | 0.809 | 0.417 | 0.090 | 3.376 |
| Pr | 0.097 | 0.069 | 0.011 | 0.770 |
| Nd | 0.359 | 0.279 | 0.035 | 3.173 |
| Sm | 0.072 | 0.067 | 0.010 | 0.628 |
| Eu | 0.003 | 0.008 | 0.001 | 0.160 |
| Gd | 0.091 | 0.093 | 0.012 | 0.819 |
| Tb | 0.020 | 0.019 | 0.003 | 0.110 |
| Dy | 0.093 | 0.084 | 0.013 | 0.804 |
| Ho | 0.020 | 0.017 | 0.003 | 0.202 |
| Er | 0.063 | 0.053 | 0.008 | 0.661 |
| Tm | 0.011 | 0.010 | 0.002 | 0.091 |
| Yb | 0.063 | 0.056 | 0.010 | 0.568 |
| Lu | 0.012 | 0.011 | 0.002 | 0.093 |



**Fig.4.** The slate distribution (PAAS on (McLennan, 1989)) of normalized REE in barites and bottom sea water of the Deryugin Basin (depth 1444 m, station Lv54−7−1).

formed due to hydrothermal activity are characterized by the magnitude of Eu* > 1 (Dubinin, 2006).

We suppose that the formation of the negative europium anomaly can be caused by two factors. The first one is that the continental lithogenesis is characterized by an intensive supply of terrigenous substance from feeding provinces (Bezrukov, Lisitsin, 1957).

The study of the mineralogical and geochemical features of the sediment from the well MD01–2415 (Levitan et al., 2007), drilled in the central part of the Sea of Okhotsk, has shown that the formation of the negative europium anomaly here is related to the granitic material in the Okhotsk volcanic belt, as well as the existence of remains of diatom organisms (Levitan et al., 2007). Moreover, smectite, which is one of the main clay minerals in the sediments of the Sea of Okhotsk (Astakhov et al., 2008, Volokhin, 2012), is also characterized by the negative europium anomaly (Dubinin, 2006; Fagel et al., 1997).

Under conditions of high sedimentation rates the barites are contaminated with both allotigenic and authigenic materials. At low REE concentrations in barites, the formation of their bulk composition depends on the impurity component.

In our case this leads to the appearance of the negative europium anomaly. However, the composition of REE of surface sediments (0–30 cm) in the Deryugin basin that accumulates simultaneously with the growth of barite formations has the positive europium anomaly (Eu * up to 1.4) both in the bulk sample (Sattarova et al., 2014) and the sample with removed authigenic mineral forms and, moreover, in the clay fraction of the sediment (Mikhailik et al., 2016). This is due to travelled material chiefly delivered by the Amur River (Derkachev et al., 2004; Mikhaylik et al., 2016; Sattarova et al., 2014).

The second reason that leads to the formation of a REE composition depleted in europium, the barite from the Deryugin basin, may be explained by the depletion of the fluid (cold seep) caused by europium. Restoration of europium to oxidation state +2 (the most mobile form of this element) is possible at elevated temperatures (Sverjensky, 1984). In the Deryugin basin, in the region of barite mineralization, high concentrations of methane are observed in the sediment column[3] (from the surface down along the column: 0–400 cm — 40–70 μl/l,500–1800 cm — 30 000–50 000 μl/l, at depths of 1100 cm from the bottom surface, the sediment are being enriched with methane up to 80,000 .mu.l/l, background 10–30 l/l) while in the bottom layer of water the concentration reaches 1943 nL/l, while background values are equal to 40–50 nl/l (Cruise ..., 1999; Cruise ..., 2000; Cruise ..., 2002). Isotopic composition of sulfur and oxygen ($\delta^{34}$S: 21.0–38.6 ‰ CDT; $\delta^{18}$O: 9.0–17.6 ‰) in barites clearly indicates biological sulphate reduction process and $\delta^{13}$C value in (> -43.5 ‰) carbonates within this area gives evidence for biogenic methane source (Greinert et al., 2002). These data are consistent with the results of a study of the geochemistry of the pore waters from the Ge 99–32 column, sampled in the immediate vicinity of the distribution of barite bodies in the Deryugin basin, which confirms that there is no any affect from the hydrothermal source of the substance (Cruise ..., 1999). Generation of the fluid enriched with barium occurs at a depth of 2 km at temperatures of 60°C under the conditions of clay minerals catagenesis (Bollwerk, 2002). This is confirmed by the results from Na–Li and Mg–Li geothermometers (Derkachev, Nikolaeva, 2007). Under these conditions, the europium is restored, and its solid phase of the clay substance is absorbed. When there is no high-temperature hydrothermal fluid influence, characterized by a highly positive europium anomaly (Dubinin, 2006), the REE composition of cold seeps depleted in europium is formed, which is inherited during barite formation.

## CONCLUSIONS

The obtained data on the distribution of REE in the barites from the Deryugin basin (low REE contents and the negative europium anomaly), as well as the features of the chemical and isotope compositions, the lack of temperature anomalies in the barite mineralization zone, the geochemistry of the pore waters of the enclosing sediment and, in addition, the morphology of the crystals show that cold seeps play the leading role of in the formation of the barite deposit. The hydrothermal component does not affect the formation of barite structures. This agrees with the data of V.V. Satarova et al. (2014), who showed that the total composition of the REE of the surface sediments in the Deryugin basin is formed by terrigenous demolition, and the effect of the hydrothermal component is not revealed. It is useful to remember about the microorganisms that play a role in the formation of marine authigenic minerals. More accurate investigation of the genesis of the barites from the Deryugin basin requires detailed identification of both mineral and geochemical criteria and in-depth microbiological study.

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