

AUTHIGENIC GYPSUM IN ROCKS AT THE BOTTOM OF JAPAN AND OKHOTSK SEAS

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Morphology of authigenic gypsum occurring at the surface of the Cenozoic rocks at the floor of Seas of Japan and Okhotsk is described. The mineral looks like unusual because of morphology of mineral aggregates, shape of crystals, and conditions of formation. The formation of this type gypsum was probably determined by high concentration of Ca^{2+} and SO_4^{2-} in porous water of cristobalite-bearing rocks, lithostatic pressure, and temperature gradient between porous and sea water. Abundance of gypsum in the rocks at the slopes of submarine morphostructures of the Japan and Okhotsk Seas allows consideration of this region as a province of authigenic gypsum mineralization.

1 table, 4 figures, 24 references.

Keywords: authigenic gypsum, morphological type, rocks at the bottom of Okhotsk and Japan Seas.

The study of the Cenozoic rocks obtained from dredging of various morphostructures of marginal seas in the northwestern Pacific in 1974–2006 revealed authigenic gypsum in them. In the Sea of Okhotsk, gypsum was found in the rocks picked up at the western (stations 2222, 2225, and 2227; depth of dredging 1350–2000 m), northern (stations 2356 and 2357; depth 2600–2900 m), and southwestern (stations 2361, 2363, 2364, 2367, and 2368; depth 2300–2900 m) slopes of the Kuril Trench. In the Sea of Japan, it was found at the Primorsk continental slope (stations 1076 and 1126; depth 1500–1930 m), uplands Pervenets (station 1747; depth 2350–2460 m) and Alpatov (stations 2047 and 2212; depth 2900–3300 m), ridges Okushiri (stations 1708 and 1713; depth 1300–2500 m), Oki (stations 1270; depth 1250–1300 m), and South Yamato (station 1434; depth 280–320 m). The similar gypsum was also found at the Pacific slope of the Kuril arc system in the Vityaz Ridge (station LV 41-16; depth 1200–1400 m) (Fig. 1). Further, seasonal (cellar) gypsum was formed at the section of some samples dredged from the Alpatov Upland (sample 2047-2; depth 2900–3300 m) and Oki Ridge (sample 1296-4b; depth 350 m). Authigenic submarine gypsum occurs as rounded (close to spherical) aggregates composed of mineral individuals, which were not previously described in literature¹. The aim of this study is examination of these aggregates and establishing their formation.

Analytical techniques

Gypsum (monomineralic sample) and cristobalite (bulk sample) were detected with a

DRON 2.0 diffractometer (CuK α radiation, graphite plate monochromator) operated at voltage 30 kV and current intensity 30 mA. Morphology and chemical composition of gypsum were examined on an EVO 50-XVP scanning electron microscope equipped with an INCA ENERGY 350 EDS. The samples were coated by carbon film EDWARDS E-306 high-vacuum system.

Results

Gypsum occurs as two varieties: (1) submarine variety formed under submarine conditions at the contact between Cenozoic rocks and sea water, and (2) cellar (seasonal) variety formed in storage at the section of the same rocks.

Aggregates of submarine gypsum are spheroids (0.1–0.5 mm in diameter, predominantly 0.2 mm) (Figs. 2, 3a, d, f) and occasional disks up to 0.2 mm thick and up to 1.5 mm in diameter (Fig. 3c). Spheroids cover the sample surface, whereas small disks are sporadic. The cellar gypsum occurs as spheroids (globe-shaper aggregates of 0.1 to 0.3 mm in diameter, predominantly 0.2 mm) which are scattered on the section of the samples (Figs. 3b, e).

The aggregates of submarine gypsum are composed of one type of mineral individuals as close aggregate of parallel lamellae of $1 \times 0.5 \times 70$ to $4 \times 1 \times 70$ μm in size (Figs. 4a, b, c). Globules of cellar gypsum are composed of abundant acicular ($1 \times 15 \times 70$ to $3 \times 15 \times 70$ μm) (Fig. 4d) and tabular ($2 \times 0.5 \times 4$ to $6 \times 1 \times 14$ μm) (Fig. 4e) crystals. Convex isometric ($5 \times 6 \times 4$ μm) segregations of gypsum are occasional between its needles (Fig. 4d).

¹ – Below the similar aggregates are named spheroids.

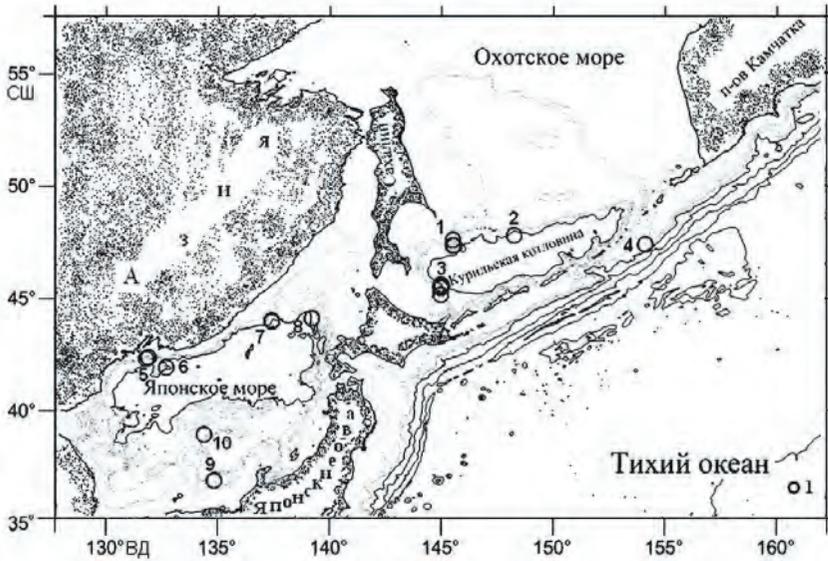


Fig. 1. Location of dredging stations. 1–3 – Slopes of Kurile Through, Sea of Okhotsk: 1 – western (stations 2222, 2225, 2227), 2 – northern (2356, 2357), 3 – southwestern (2361, 2363, 2364, 2367, 2368); 4 – Vityaz Ridge, Sea of Okhotsk (station LV 41-16); 5–10 – Sea of Japan: 5 – Primorsk continental slope (1076, 1126), 6 – Pervents Upland (1747), 7 – Alpatov Upland (2047, 2212), 8 – Okushiri Ridge (1708, 1713), 9 – Oki Ridge (1270), 10 – South Yamato Ridge (1434).

Thus, the habit of crystals of mineral individuals of submarine gypsum differs from that of cellar gypsum.

Submarine gypsum was formed at the surface of substrate that is the Cenozoic (Late Paleocene to Late Miocene) rocks. Authigenic gypsum as sporadic twins of $30 \times 10 \times 1 \mu\text{m}$ in size (Fig. 4f) and tables $4 \times 5 \times 1 \mu\text{m}$ (Fig. 4g) was found within the rocks.

The chemical composition of most part of the newly formed submarine (cores of lamellae) and cellar (surface of needles and tables) gypsum is usual (henceforward wt.%): 25.29–28.12 Ca, 21.57–23.7 S, and 46.84–48.0 O. Insignificant admixture of Si (up to 2.0), Cu (up to 0.87), and Al (up to 0.43) is frequent; Fe up to 0.53 is occasional; and Zn (0.78) and Ti (0.75) are in isolated compositions (see Table 1).

The complex composition of surface of the broad part of subsea gypsum lamellae is characterized by lowered content of Ca (16.96 wt.%) and S (16.12), elevated content of O (53.44), Si (4.98), Al (1.08), Fe (1.27), and presence of K (0.82), Na (2.49), Mg (0.59), Cl (1.42) (anal. 3). The composition of the surface of convex isometric cellar gypsum is similar. The absence of Cl in the cellar gypsum is the unique difference (anal. 6).

As aforementioned, two varieties of gypsum were found within the rocks which were basement for submarine gypsum. The chemical composition of tabular gypsum is close to theoretical, whereas that of gypsum twins is slightly different. With the same O content (48.33–48.78) and close Fe content (0.91–1.17), the latter is characterized by the lower concentration of Ca (19.66–20.35)

and S (19.12–19.37), and higher content of Si (8.2–8.94) and Al (1.56–1.62); in addition, K (0.32–0.56), Na (0.55), and Mg (0.39–0.57) were measured. This composition is similar to the complex composition of the surface of the broad part of submarine lamellae and convex cellar gypsum (anal. 3, 6).

The substrate is distinguished by high (in comparison with gypsum) content (wt.%) of Si (19.93–3.64) and O (24.68–64.05); Mg (0.32–1.97), Al (1.06–9.73), S (0.32–2.34), and Fe (0.38–11.07) are constant; K (0.37–4.36), Na (0.7–2.18), Ca (1.83–2.75), Cl (0.3–1.26), and Ti (0.4–1.15) are frequent; and Cu (0.68–2.35) and Zn (0.59–0.61) are occasional (see Table 1).

Discussion

Gypsum is known to be formed in marine sediments as indicated by its frequent findings in the core of Cenozoic biogenic-siliceous rocks raised from floor of Atlantic, Indian, and Pacific Oceans (Briskin and Schreiber, 1978; Muza and Wise, 1983). Many scientists suggest that gypsum is formed at the early diagenetic stage close to water/sediment interface. It precipitated (in most districts studied) in anoxic environment with involvement of anaerobic bacteria and significant amount of organic matter. The necessary concentration of Ca^{2+} is believed to be reached due to dissolution of carbonate microfossils composing sediments (Briskin and Schreiber, 1978; Muza and Wise, 1983; Schnitker *et al.*, 1980), whereas the link of SO_4^{2-} with sulfate-reducing bacterial activity is proved only theoretically. Common association

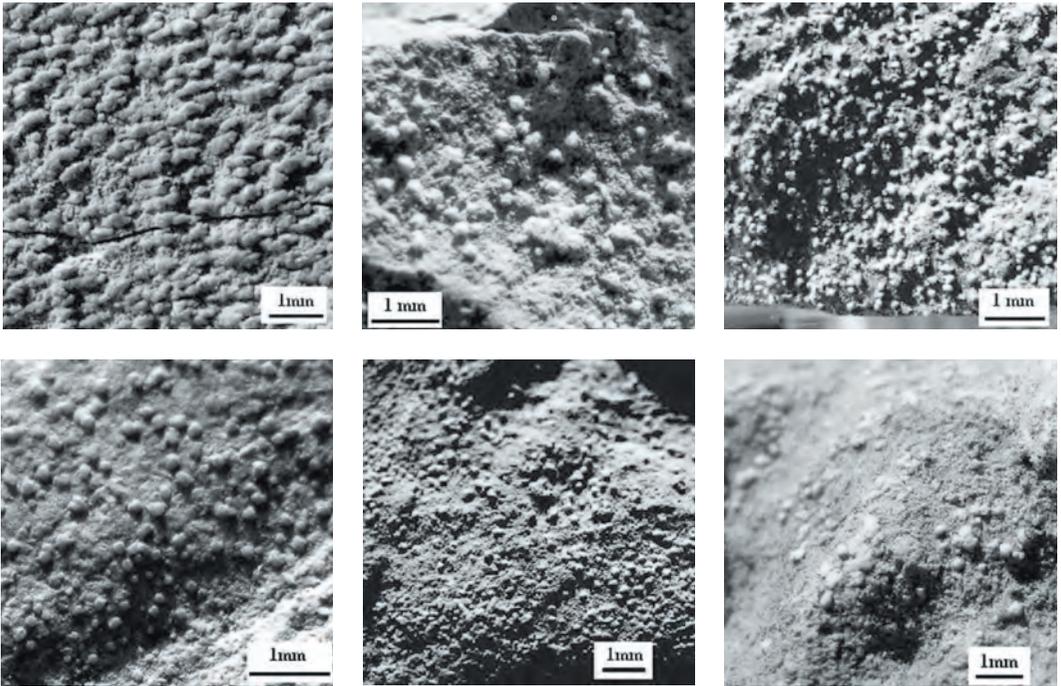


Fig. 2. Morphology of aggregates of submarine gypsum. Numbers of samples correspond to the sample locations shown in Fig. 1: a – cherty mudstone, sample 2227-a; b – diatomite, sample 2356-1; c – cherty silty mudstone, sample 2364-2; d – cherty silty mudstone, sample LV 41-16-1; e – silty mudstone, sample 1126; f – tuff stone, sample 1747-2t.

Fig. 3. Morphology of aggregates of submarine (a, c, d, f) and cellar (b, e) gypsum: a – cherty silty mudstone, sample 2047-3; b – diatom clay, sample 2047-2; c – cherty rock, sample 1713-8a; d – silty mudstone, sample 1713-9; e – diatom tuff, sample 1296-4b; f – tuffdiatomite, sample 1434.

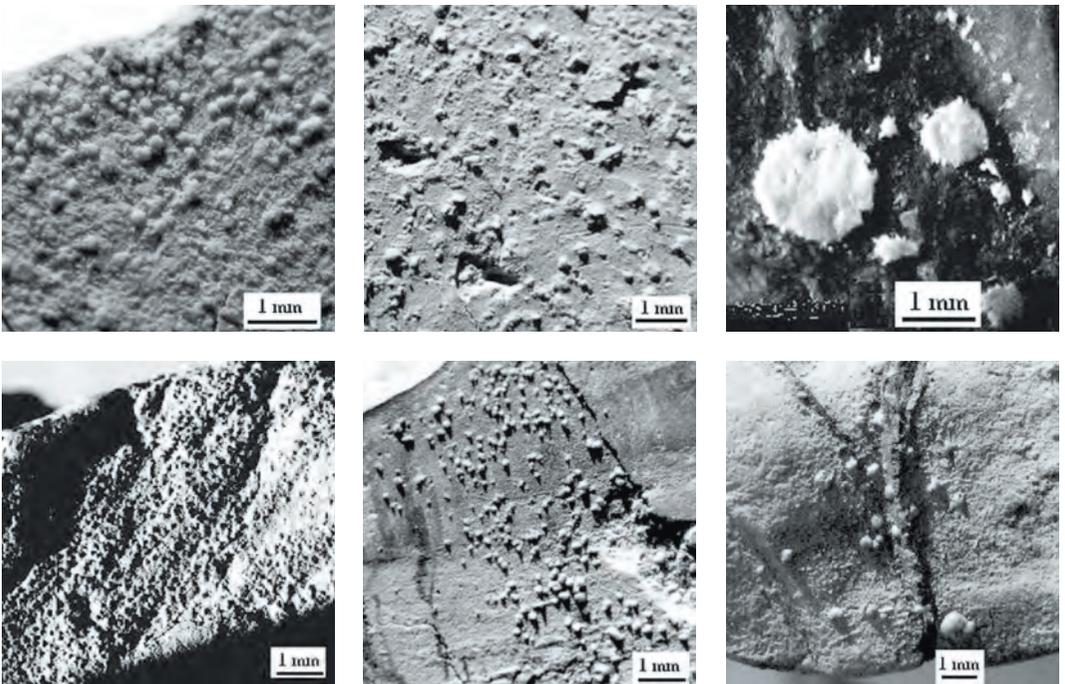


Table 1. Composition of gypsum and substrate of Cenozoic rocks in Japan and Okhotsk Seas

№ an.	№ sample	Location	Number of points	Content of elements (wt.%)												
				Ca	S	O	Si	Cu	Al	Fe	Zn	Ti	Mg	K	Na	Cl
Submarine gypsum																
1	2227-a	inner part of lamella	5	25.67	23.35	48.26	2.47	—	0.37	—	0.39	0.38	—	—	—	—
2	1713-8	inner part of lamella	8	25.11	24.02	46.86	0.14	0.58	—	—	0.53	0.38	—	—	—	—
		surface of lamella	2	28.64	23.62	46.98		0.36			0.39					
3	1713-8	surface of broad part of lamella	1	16.96	16.12	53.44	4.98	0.8	1.08	1.27	—	—	0.59	0.85	2.49	1.42
Cellar gypsum																
4	2047-2	surface of needle	2	25.77	23.58	47.74	1.45	0.93	0.43	—	—	—	—	—	—	—
5	2047-2	surface of plate	2	26.09	23.51	47.63	1.39	0.80	0.51	—	—	—	—	—	—	—
6	2047-2	surface of convex segregations of gypsum	1	22.75	20.05	47.59	5.54	0.81	1.47	0.46	—	—	0.42	0.37	0.55	—
Gypsum in substrate																
7	2227-a	surface of twined segregation	3	20.00	19.24	48.56	8.43	—	1.60	1.00	—	—	0.28	0.47	0.28	—
8	2047-2	surface of table	1	25.07	23.54	47.97	1.6	1.12	0.7	—	—	—	—	—	—	—
Substrate																
9	2227-a	substrate	2	0.92	1.85	55.58	30.65		4.36	2.13	—	0.2	1.29	0.76	1.62	0.62
10	2047-2	substrate	2	2.44	2.92	56.52	33.10	0.89	1.18	0.47	—	—	0.14	0.14	1.49	0.63
11	2047-a	surface of diatom	1	1.85	1.46	64.05	26.96	0.78	1.27	0.84	0.59	0.43	0.32	0.37	0.70	0.38

Notes: Scanning electron microscope equipped with integrated standards for qualitative (Point & ID) EDS analysis.

The sums obtained were normalized to 100%. N.N. Barinov, analyst.

High content of Si, Al, and Fe is resulted from rapid growth of gypsum, when it does not manage to release impurities trapped at crystallization.

of gypsum and pyrite suggests accumulation of SO_4^{2-} in interstitial water (involving gradient of diffusion) during the formation of pyrite SO_4^{2-} is probably accumulated as a result of dissimilatory sulfate-reducing to form H_2S followed by the transition of sulfide sulfur to sulfate ion by chemo- and phototrophic sulfur-bacteria and archaea (Grabovich, 1999). The available models of the SO_4^{2-} concentration in porous water do not explain the formation of authigenic gypsum (found in the rocks from drill cores deep-water holes) in oxidative environment. The model of concentration of sulfate ion in sediments as a result of its diffusion from sea-water is applicable to this sedimentation environment (Berner, 1970). SO_4^{2-} could be accumulated in both sediments enriched in oxygen and sediments enriched in H_2S .

In our case, authigenic (submarine) gypsum was formed on the rock surface rather than in sediments. Porous solutions were probable source of Ca^{2+} and SO_4^{2-} . As aforementioned, gypsum studied here is divided into two types in chemical composition: usual and more complex distinguished by the admixtures of Mg, K, and

Na and occasionally Cl. The qualitative chemical composition of the second type is close to the of substrate and sea water (Brownlow, 1984). The both types were found in both submarine and cellar mineralization in samples from three different districts. The geographical abundance of gypsum allows relict sea-water as common source of Ca^{2+} and SO_4^{2-} .

All samples containing gypsum are rocks displaying transformation of biogenic opal (A-opal) to CT-opal (cristobalite). In the Cenozoic rocks of the Japan and Okhotsk Seas, cristobalite was resulted from crystallization of silica of frustules dissolved in porous water (Mozherovsky *et al.*, 2001; Svinnikov, 2004). In the samples studied here, orals of diatoms are dissolved partially (semi-dissolved diatoms, sample 1296-4b) or completely (to form holes, samples 1713-9, 1747-2t, 2047-2). Crystallization of dissolved silica is shown as cristobalite that was established with the X-ray as insignificant impurity, initial transformation of A-opal (samples 1296-4b, 1434), rock-forming mineral (the process is appreciable) along with CT-opal (samples 1126, 1713-8b, 2047-3, 2227-a,

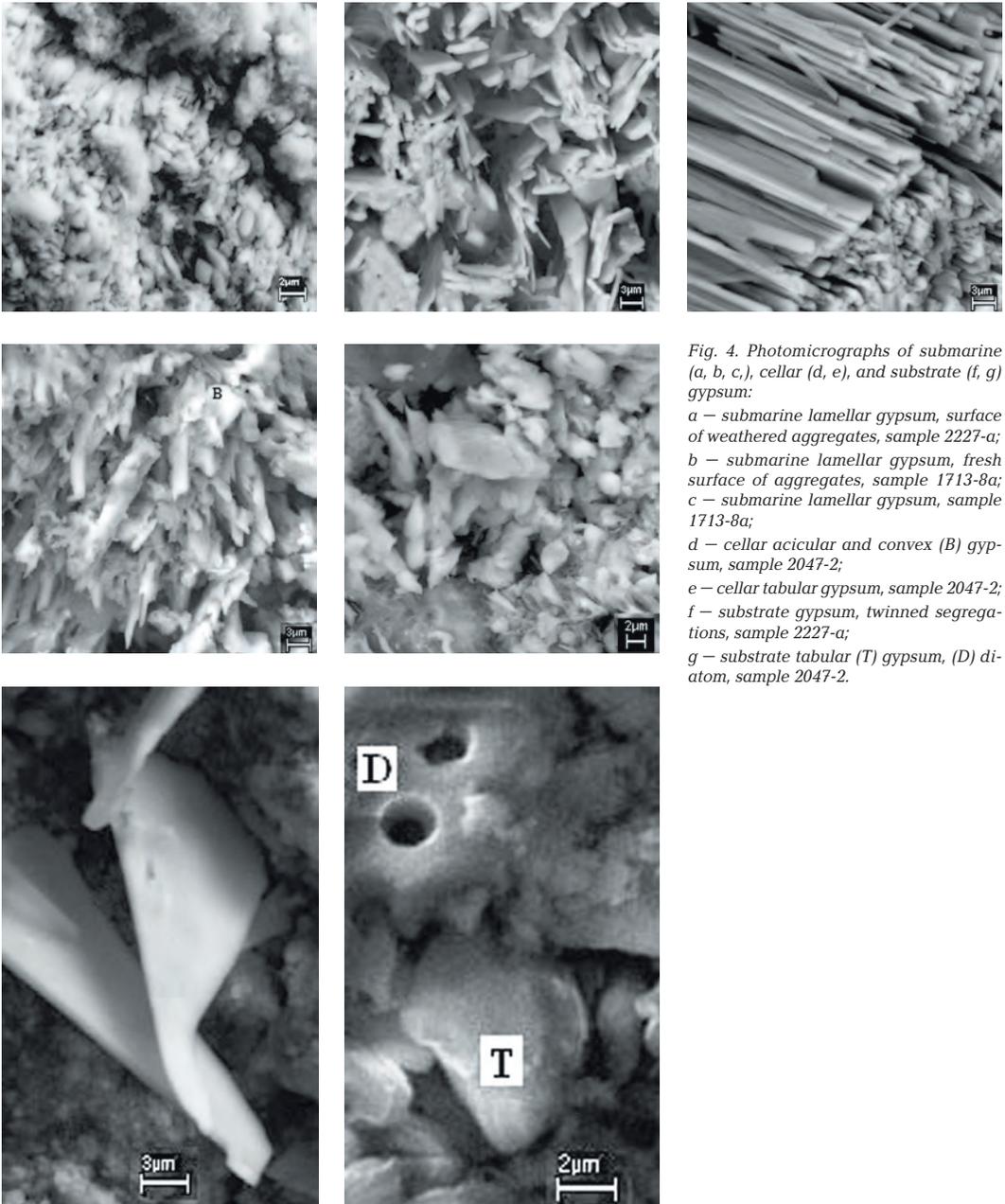


Fig. 4. Photomicrographs of submarine (a, b, c), cellular (d, e), and substrate (f, g) gypsum:

- a – submarine lamellar gypsum, surface of weathered aggregates, sample 2227-a;
- b – submarine lamellar gypsum, fresh surface of aggregates, sample 1713-8a;
- c – submarine lamellar gypsum, sample 1713-8a;
- d – cellular acicular and convex (B) gypsum, sample 2047-2;
- e – cellular tabular gypsum, sample 2047-2;
- f – substrate gypsum, twinned segregations, sample 2227-a;
- g – substrate tabular (T) gypsum, (D) diatom, sample 2047-2.

2356-1, 2364-2, LV 41-16-1), and predominant constituent (pronounced reflection at 0.413 nm, sample 1713-8a). Thus, in the Cenozoic sediments of the Japan and Okhotsk Seas, gypsum is spatially related to the transformation boundary A-opal/CT-opal (A/CT).

The process of transformation of silica is well studied in the core of Cenozoic biogenic siliceous rocks obtained with deep-sea drilling

in different districts of the World Ocean (Murray *et al.*, 1992). Some publications give data of distribution of the composition of interstitial water along sections of holes crossing the A/CT boundary. Cristobalite was found at depth (thickness of sediments) ca. 200 m, drill hole 469 and ca. 150 m, drill hole 471 when the Cenozoic (Miocene to Quaternary) cover was drilled in the region of the California continental borderland

(Eastern Pacific) (Grechin *et al.*, 1981). Cristobalite was established in the Mesozoic to Cenozoic siliceous rocks at the depth ca. 300 m (Eocene-Oligocene boundary) in the Nauru depression (drill hole 462, region of Caroline and Marshall Islands, Western Pacific) (Riech *et al.*, 1981). Cristobalite was found at the depth ca. 140 m in the core of the Upper Miocene calcareous mudstone from the Barbados Ridge (drill hole 672 m, region of Lesser Antilles, Western Atlantic Ocean) (Capet *et al.*, 1990). Content of SO_4^{2-} and Ca^{2+} in interstitial water of Cenozoic rocks in these morphostructures ranges from 27.9 to 18.9 and from 29.0 to 12.14 mmol/L, respectively above the A/CT boundary. Below this boundary, the content of these ions is 24.9 to 1.6 and 29.45 to 79.2 mmol/L, respectively. The content of SiO_2 below the boundary drastically (2–10 times) drops (from 1030–1116 to 584–93 $\mu\text{mol/L}$) due to the formation of solid phase that is cristobalite (Gieskes, Johnson, 1981; Gieskes *et al.*, 1981; 1990).

The maximal solubility of gypsum in water without regard for effect of other components is known to be 2.05–2.11 g/L that corresponds to 21 mmol/L for SO_4^{2-} and 51 mmol/L for Ca^{2+} within the temperature range of 20 to 50°C. At temperature above and below these values, its solubility drastically decreases (Great Encyclopedia..., 2008). The range of temperature 36.5 to 51.0°C within cristobalite is formed (Kuramoto *et al.*, 1992) is similar. According to Gieskes, and Johnson (1981), porous water in sedimentary rocks from all holes 10–27 m higher than the A/CT boundary is saturated in SO_4^{2-} and undersaturated in Ca^{2+} (to form gypsum at 20–50°C). Below (45–207 m) of the A/CT boundary, porous water (drill holes 469, 462, 672) is also saturated in SO_4^{2-} ; below 200 m, the content of SO_4^{2-} decreases (down to 1.1–0.1 g/L). The content of Ca^{2+} below the A/CT boundary gradually increase and below 200 m becomes high (drill holes 469, 672, and 462, at depth 190, 187, and 207 m, respectively). The concentration of Ca^{2+} continues to increase downward and reaches 3.1 g/L at 340 m in drill hole 672 and at 555 m in drill hole 471. At the depth below 200 m of the A/CT boundary, porous water is the maximum saturated in gypsum that is confirmed by the content of SO_4^{2-} 2.4 g/L and Ca^{2+} 2.3 g/L at 207 m in drill hole 462.

Thus, interstitial water of the Cenozoic cristobalite-bearing rocks of the Pacific and Atlantic Oceans have high content of SO_4^{2-} and Ca^{2+} at the depth ca. 200 m below the A/CT boundary. Frequent findings of gypsum resulted

from drying of porous solutions in core samples of deep-sea drilling in bank of core storage supports this (Briskin and Schreiber, 1978).

The similar situation is probable in the Cenozoic rocks of the floor of the Japan and Okhotsk Seas. The assemblage of submarine gypsum and cristobalite, presence of gypsum in substrate (cristobalite), and the formation of cellar individuals on the rock surface in stone storage room indicate the high concentration of Ca^{2+} and SO_4^{2-} in the porous water of the samples studied here.

The examined gypsum was formed on the surface of primary diatom-bearing sedimentary and volcanosedimentary rocks. The sediments of the Japan and Okhotsk Seas, where transformation of A-opal to CT-opal is weak (dissolved diatoms, absence or traces of cristobalite) are weakly lithified and high porous. In most samples, transformation of silica was terminated by the formation of lithified siliceous assemblages (chert, cherty siltstone and mudstone), where cristobalite is rock-forming mineral. The rocks similar in composition are highly fractured (diagenetic and tectonic nature); because of this fracturing the common hydrodynamic system occurs in the sedimentary system (Tectonics..., 1985). It is evident that under lithostatic pressure, porous solutions should be pressed off on the outcropped rock surface.

Submarine gypsum on the rock surface indicates its formation at the contact with sea-water. In the Sea of Japan, gypsum-bearing rocks were raised from the depth of 300 to 2900 m. The temperature of near-bottom water ranges from 0.14 to 1.5°C (Hydrometeorology..., 2003). In the Sea of Okhotsk gypsum-bearing rocks were raised from the depth of 1350–2900 m. The temperature of near-bottom water in the dredging area is 1.9–2.1°C (Hydrometeorology..., 1998). As aforementioned, cristobalite crystallizes at ca. 40°C. The similar bed temperature (42–62°C) was established in the sequences, where cristobalite is a rock-forming constituent (Tectonics..., 1985). The above data are testimony that there is a thermal gradient at the boundary between porous water (rock surface) and sea-water. Drastic drop of temperature at this boundary results in decreasing solubility of gypsum that appears to determine the mineral precipitation immediately on the rock surface.

Thus, in our opinion, the formation of submarine gypsum is determined by high concentration of Ca^{2+} and SO_4^{2-} in porous water of the rocks outcropped on the sea floor, lithostatic pressure of overlaying sequences, and thermal

gradient at the boundary between porous and sea water.

In the drill core raised from the floor of Atlantic, Indian, and Pacific Oceans, gypsum usually occurs as single or twinned crystals of selenite (up to 5 mm in diameter) and gypsum rosettes (from 2 to 7.5 mm) (Muza and Wise, 1983; Briskin and Schreiber, 1978). Tabular and prismatic to acicular (Briskin and Schreiber, 1978), prismatic, and flattened ($0.5 \times 1.0 \times 1.5$ mm) (Criddle, 1974) crystals of gypsum are very seldom. According to description and photos, all these crystal types differ from those studied here in both habit and significantly larger size. Radial aggregates (spherulites) of gypsum similar in size to spheroids of cellular gypsum are formed on the surface of pebbles in continental environment. In caves, gypsum is resulted from saturated solutions filtrated through porous carbonate substrate under effect of outer (hydrostatic) pressure. This model of gypsum formation at the contact of porous substrates saturated in mineralizing solutions and air (Maltsev, 1996) is close to that of the formation of submarine gypsum proposed in this study.

In addition to the conditions governed by natural environment, the process related to human factor — deteriorating environmental situation — is known. This is sulphatization that is «disease» of marble and limestone sculptures and constructions resulted in the formation of black patina by gypsum, other authigenic minerals, and moruloids. Lamellar crystals (up to 30 μm) forming «continuous carpet» of rosettes were found in these assemblages among numerous individuals of gypsum (Timasheva *et al.*, 2007). Some individuals of this gypsum are close in size to those of submarine gypsum, but frequently differ in curved shape and disordered relationship to each other.

Conclusions

Thus, the aggregates of authigenic gypsum from the Cenozoic rocks of the Japan and Okhotsk Seas interesting in their morphology were examined. These aggregates are unusual due to their morphology, shape of individuals, and conditions of formation. The occurrence of gypsum is determined by the following reasons: high concentration of Ca^{2+} and SO_4^{2-} in porous water in the Cenozoic rocks outcropped on the floor, lithostatic pressure, and thermal gradient between porous and sea water.

The abundance of gypsum in the rocks on the slopes of submarine morphostructures of the

Japan and Okhotsk Seas allows considering this region as a province (Frolov, 1992) of submarine authigenic gypsum mineralization.

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