Distribution of authigenic albites in a limestone succession of the Helvetic Domain, eastern Switzerland

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Key words: authigenic albite, Glarus Nappe, diagenetic processes

ABSTRACT

A new occurrence of authigenic albite was found in a Jurassic sedimentary succession of the Glarus Nappe near Walenstadt (eastern Switzerland). The euhedral shape and the chemical purity of these albites are evidence for their authigenic origin. The crystals are irregularly distributed in the sediment, highlighting the importance of the host rock composition for albite authigenesis. The crystals occur exclusively in limestones with a carbonate content higher than 80 wt-%. A diagenetic or hydrothermal origin of albite authigenesis is discussed for the studied region. Clay mineral transformation appears to be an important source of ions for albite formation.

RESUME

Un nouveau gisement d'albite authigène a été découvert dans une succession sédimentaire du Jurassic, situé dans la nappe de Glaris près de Walenstadt (Est de la Suisse). La pureté chimique et la forme euédrique de ces albites atteste de leur origine authigène. La répartition irrégulière des cristaux dans le sédiment souligne l'importance de la composition de roche hôte pour la formation des albites. Les cristaux sont présents uniquement dans les calcaires contenant plus de 80 %m de carbonate. La possibilité d'une origine diagénétique ou hydrothermale est discutée pour la région étudiée. La transformation des minéraux argileux paraît être une source importante d'ions pour la formation de l'albite.

1. Introduction

Authigenic albites grow during the low-temperature burial of sedimentary rocks. They differ from detrital albites by their euhedral crystal shape, their high chemical purity, and their uncommon crystallographic properties (Füchtbauer 1948). The occurrence of these albites is scarce and the exact processes controlling their formation remain unclear.

The specific features and the growth conditions of authigenic albites in carbonates were summarized by Kastner & Siever (1979), and more recently by Spoetl et al. (1999). They showed that authigenic albites preferentially grow in carbonate rocks, forming well-crystallized grains, with a common size of 60 to 120 µm. Their size increases with increasing burial temperature, and can reach up to 10 mm in rocks that experienced epizonal conditions (Richter 1978; Brauckmann 1984; Spoetl et al. 1999). Contrary to albites of igneous origin, authigenic albites show a peculiar twinning called "Roc Tourné", which is a combination of X-Carlsbad and albite twins (Rose 1865; Füchtbauer 1948).

Electron microprobe analyses show that authigenic albites tend to be nearly pure end-members of the plagioclase feldspar series with only very low anorthite and orthoclase component (Kastner & Siever 1979). This high chemical purity is responsible for the lack of light emission in cold cathodoluminescence microscopy. However, by using a hotcathode equipment, Richter et al. (2002) were able to show a weak luminescence emission, due to the activation of Mn^{2+} and Fe^{3+} .

In this study, we document for the first time the occurrence of authigenic albite in a Jurassic limestone of the Glarus Nappe (eastern Swiss Alps). We describe the albite distribution from a regional to a thin section scale, and we discuss a possible diagenetic or hydrothermal origin of these albites.

2. Geological setting

Authigenic albites were found in two sections situated in the Lüsis Slice (Glarus Nappe s.l.), which belongs to the Helvetic

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Fig. 1. Location of Nissibach and Milchbach on the tectonic map of Walensee, Switzerland (after Herb and Franks-Dollfus, 2003).

Domain of the eastern Swiss Alps (Herb and Franks-Dollfus, 2003)(Fig. 1). This study focuses on the section Nissibach, situated near the town of Walenstadt (Swiss coordinates: 742.930/221.940). The second section, Milchbach, is located along a stream, 2.8 kilometers east of Nissibach (744.940/ 219.880).

The Nissibach and Milchbach sections are composed of sediments accumulated along the northern Tethyan margin during the Jurassic. The sediment succession containing albite starts in the Middle Jurassic with Bajocian limestones and ends with the deposition of Oxfordian marlstones (Late Jurassic) (Fig. 2). The age of the sections is well constrained by ammonite- (Kugler 1987) and carbon isotope stratigraphy (Padden et al. 2002; Rais et al. 2007). The sediments were deposited on the continental shelf, in an open marine environment episodically affected by strong oceanic currents (Rais et al. 2007).

During alpine orogeny, the studied part of the Glarus Nappe experienced deep burial diagenesis to low-grade metamorphism. The burial history of the region was reconstructed by Wang et al. (1996) using illite crystallinity, the presence of index minerals, and the ordering of illite/smectite. In the studied area, clay mineral assemblages indicate deep burial diagenesis close to the anchizone boundary, suggesting burial temperatures around 200°C (Merriman & Frey 1999).

3. Methods

Albites were studied using a combination of optical microscopy, cathodoluminescence microscopy, back-scattered electron microscopy, and electron microprobe analysis.

The counting of albite crystals was carried out on a 6.25 mm² surface of thin-sections under the optical microscope. Albite concentration is expressed in number of albite crystals per square centimeter (Ab/cm2). Cathodoluminescence analyses were carried out on a cold-cathode device, using polished thin-sections.

The elemental composition was determined using a JEOL-JXA 8200 electron microprobe. Analytical conditions were 15 kV acceleration voltage at a current of 20 nA. The spatial resolution attained is about 2 µm. Standard used were synthetic and natural oxides and silicates.

The carbon content was measured with a UIC CM5012 Coulometer. Powders were extracted from the slab used for thin-section preparation by using a micro-drill. The inorganic carbon was obtained by dissolution of the sample in perchloric acid, and total carbon by combustion at 950 °C. Organic carbon contents were calculated by difference and are expressed in wt-%. Inorganic carbon results are expressed in wt-% carbonate. Analytical precision is ± 0.1 % for carbonate carbon and ± 0.3 % for organic carbon.

Nissibach

Fig. 2. Stratigraphy and albite distribution of the section Nissibach.

4. Albite crystal distribution

Albite crystals found in the studied sections are of authigenic origin, as indicated by the euhedral shape and the chemical purity of the crystals. To assess the influence of the host rock composition on the development of authigenic albites, their distribution is described from a regional to a thin-section scale.

4.1. Regional scale

The difference in albite abundance in the two studied sections indicates important regional variations. The section of Nissibach contains abundant crystals of large size (200 µm in average), whereas at Milchbach they are smaller (max 130 μ m and 80 µm in average) and only occur in the 2 m-thick limestone bed of the Schilt-Kalk. The presence of authigenic albite is not reported in previous studies of the region (Kugler 1987; Wang et al. 1996), suggesting a geographically very restricted occurrence.

4.2. Outcrop scale

The Nissibach section is described in more detail because of the numerous and well developed authigenic albites. The section is 9 m-thick and comprises four lithologies (Fig. 2). The "Reischiben-Serie" is a grainstone of Bathonian to Bajocian age mainly composed of echinoderm fragments with minor amounts of serpulids, shell fragments, quartz grains and reworked clasts of micritic limestone. The Reischiben-Serie contains only few albites.

The "Blegi-Oolite" is a 80 cm-thick hardground of Callovian to Early Oxfordian age consisting of an iron-rich micritic limestone. It is a mudstone to packstone containing iron-ooids and bioclasts (cephalopods, echinoderms, sponges, and microfauna). This bed does not contain albite.

The "Schilt-Kalk" is a Middle Oxfordian member of the Schilt Formation. It is composed of a 6.2 m-thick succession of micritic nodular limestones containing a typical pelagic fauna (ammonites, belemnites) surrounded by a micritic to microsparitic matrix (Fig. 3D). Between the nodules, bioclasts are scarce and usually unidentifiable. The nodules have been interpreted to be the result of reworking of early-cemented mud pebbles at the sea floor (Kugler 1987; Rais et al. submitted). In the Schilt-Kalk, albites are large and abundant, but crystals only occur in the nodules (see below).

The "Schilt-Mergel" is a succession of monotonous grey to beige marls containing rare fragments of cephalopods, sponges or echinoderms. The detrital fraction consists of clay, and no quartz crystals or authigenic albites were observed.

4.3. Bed scale

At Nissibach, the amount of albite is variable from bed to bed (see Fig. 2). The variation is random, with no systematic increase or decrease along the section. The maximum concentra-

Fig. 3. (a, b) Euhedral authigenic albites. Roc Tourné and polysynthetic twinning (Thin-section, crossed polars) (c) Cathodoluminescence. The nonluminescent albites contrast with the dark orange micritic nodule (N) and the bright orange color of the surrounding matrix (M). (d) Picture of a thin-section from the Schilt-Kalk: the albites are concentrated in the nodules (N). (e) Cut albite crystal at the edge of a nodule (Thin-section, crossed polars). (f) The top albite is reduced by pressure solution (S), and the lower albite is cut by a calcite vein (V) (electron microprobe back-scatter image). (g) Albite growth is disturbed at the contact with a bioclast (Thin-section, crossed polars). (h) Intergrowth between authigenic albite and pyrite (Thin-section, crossed polars). Scales: a,b,c,e,f,g,h: 100 µm. d: 5 mm.

tion is reached in samples Ni 12 and Ni 19 with respectively 624 Ab/cm2 and 656 Ab/cm2. In contrast, sample Ni 15 has a maximum of 96 Ab/cm².

4.4. Thin-section scale

Thin-section observations of the Reischiben-Serie and the Schilt-Kalk reveal that authigenic crystals growth is limited to specific zones. In the Reischiben-Serie, albites are especially abundant in the micritic clasts (up to 400Ab/cm2). In the grainstones, they occur mostly between bioclasts, only a few grew on bioclasts. Albites are less abundant in the grainstone (in average 200 Ab/cm²) than in the micritic clasts, but they are larger (up to 400 µm).

In the Schilt-Kalk, the albites are confined to the nodules. In most of the samples, the crystals are scattered throughout the nodules in a regular pattern, and are not concentrated at the edge of the nodule, and rarely along joints or veins. Al-

Fig. 4. Ternary diagram of the plagioclase feldspar series. The authigenic albites are almost pure end-members (DPlot program from John (2004)).

bites never occur in veins. Some crystals appear to be cut at the limit between nodule and matrix (Fig. 3E). Overgrowth of albite in the matrix has not been observed.

The amount of albite varies from a nodule to another within the same sample. For example, the albite concentration in the sample Ni 19 varies between 656 Ab/cm² and 368 Ab/cm2.

5. Petrography and geochemistry

Authigenic albites are euhedral and on average 200 μ m long (range from 50 µm to 400 µm). The crystals show Roc Tourné twinning (Füchtbauer 1948; Füchtbauer 1950) (Figs. 3A, B, G, H) and contain varying amounts of solid and fluid inclusions which are mostly residues of the host rock (calcite, dolomite, clay). Unfortunately the scarcity and small size (4 to 5 µm in average) of fluid inclusions exclude an analysis of crystallization temperatures and fluid compositions. Under coldcathodoluminescence microscope, albite crystals are nonluminescent (Fig. 3C), where the dark albites contrast with the bright orange of the surrounding carbonate.

The relation of the albites with other components of the sediment provides information on their genesis. Whereas bioturbation structures do not have an impact on crystal formation, the presence of a bioclast slightly disturbs their growth (Figs. 3C,G). Albites never grew beyond the border of the nodules, and in many cases, the crystals are clearly cut by dissolution seams (Figs. 3E,C). Dissolution seams are commonly observed around or within nodules, as shown in Figure 3F, where the upper albite crystal is dissolved on one side by pressure solution. Some albites formed across a first generation of calcite veins, whereas in other places late generation calcite veins cut trough them (Fig. 3F). Figure 3H shows a late-diagenetic pyrite crystal, intergrown with albite.

Fig. 5. Relation between carbonate content and albites abundance. Authigenic albites occur in sediment with a carbonate content higher than 87 wt%.

Microprobe analyses show that albites are nearly pure, with on average 98.4 mol% albite component (Table 1, Fig. 4). Very little variations were observed between the center and the rim of a crystal, indicating the absence of a detrital core as nucleation center. No detrital feldspars were found in the samples.

6. Albite abundance and carbonate content

The variation of albite abundance from one lithology to another indicates that the chemical composition of the host rock is determinant for albite authigenesis. In contrast the presence of authigenic crystals in micritic and sparitic limestone suggests that the texture of the host rock does not significantly influence albite formation.

A plot of carbonate content vs. the relative abundance of albite crystals (Fig. 5) shows that albite only occurs in sediments with more than 87 wt-% carbonate, whereas no albite was observed when the carbonate content is lower than 80 wt- %. Above 87 wt-% carbonate, albite abundance is not correlated to carbonate content. Organic carbon contents were lower than 0.1 % in all samples.

Table 1. Results of electron microprobe analyses of authigenic albites. Albite (Ab), orthoclase (Or), and anorthite (An) percentages were calculated using the general formula NaAlSi3O8 for albite.

Sample	SiO ₂	Al_2O_3	Na ₂ O $(wt \%)$	K_2O	CaO	Total $(wt \%)$	Ab	Or $(mod \%)$	An	Comments
Ni 10	69,21	19,43	11,33	0,08	0,10	100,15	99,05	0,48	0,47	1 Centre
Ni 10	68,44	19,31	11,07	0,27	0,29	99,38	97,07	1,53	1,40	2 Centre
Ni 10	67,65	20,09	10,63	0,92	0,23	99,52	93,57	5,30	1,13	3 Edge of 2
Ni 10	68,78	19,54	11,22	0,21	0,10	99,85	98,30	1,20	0,50	4
Ni 10	69,19	19,50	11,29	0,15	0,08	100,21	98,74	0,89	0,37	5
Ni 10	69,29	19,16	11,22	0,09	0,08	99,83	99,12	0,51	0,38	6 Centre
Ni 10	68,74	19,56	11,10	0,24	0,21	99,85	97,61	1,38	1,01	7 Edge of 6
Ni 10	68,30	19,38	11,06	0,21	0,16	99,10	98,02	1,21	0,77	8
Ni 10	68,44	19,36	11,26	0,10	0,42	99,58	97,40	0,57	2,03	9
Ni 15	69,24	19,30	11,38	0,11	0,08	100,11	98,97	0,64	0,38	11
Ni 15	69,51	19,39	11,36	0,10	0,21	100,57	98,44	0,56	1,01	12
Ni 15	69,37	19,08	11,45	0,03	0,07	99,99	99,53	0,15	0,31	13
Ni 15	69,21	19,33	11,23	0,10	0,09	99,96	98,98	0,59	0,44	14
Ni 15	69,29	19,39	11,25	0,12	0,07	100,12	98,96	0,68	0,36	15
Ni 15	69,06	19,45	11,23	0,11	0,11	99,96	98,83	0,63	0,54	16
Ni 15	69,76	19,13	11,44	0,03	0,08	100,44	99,46	0,18	0,36	17 Centre
Ni 15	69,69	19,30	11,08	0,09	0,09	100,25	99,01	0,54	0,45	18 Edge of 17
Ni 15	69,17	19,38	10,91	0,17	0,25	99,88	97,74	1,01	1,25	19 Small
Ni 15	68,65	19,44	10,92	0,21	0,12	99,34	98,18	1,25	0,58	20 Small
Ni 20	69,28	19,50	11,16	0,21	0,06	100,21	98,50	1,20	0,30	21 Centre
Ni 20	70,01	19,20	11,31	0,04	0,15	100,71	99,04	0,24	0,72	22 Edge of 21
Ni 20	69,75	19,35	11,31	0,08	0,08	100,57	99,15	0,47	0,38	23
Ni 20	69,71	19,34	11,14	0,06	0,06	100,31	99,34	0,35	0,31	25
Ni 20	68,79	18,99	11,16	0,10	0,56	99,60	96,77	0,54	2,69	27
Ni 20	70,11	19,22	11,45	0,04	0,04	100,86	99,60	0,22	0,17	28
Ni 20	69,56	19,34	11,16	0,10	0,06	100,22	99,10	0,61	0,29	29
Ni 20	69,63	19,39	11,32	0,19	0,21	100,74	97,91	1,08	1,01	30
	69,18	19,36	11,20	0,15	0,15	100,05	98,38	0,89	0,73	Average

7. Discussion

7.1. Albite characteristics and growth environment

The euhedral shape, the Roc-Tourné twinning and the chemical composition of the studied albites testify of their authigenic origin. The 98.4 mol% albite content is consistent with other occurrences where compositions mostly above 99 mol% were reported (Spoetl et al. 1999; Richter et al. 2002). The lack of chemical zoning in Nissibach samples (Table 1) indicates that albite growth initiated without a preexisting core, in contrast to authigenic albites developing as overgrowths around detrital plagioclases described by Turner et al. (1982). The growth of albites across bioturbation structures, and their interrupted growth at the contact with a bioclast are also in agreement with an authigenic origin.

The nodular structure of the "Schilt-Kalk", is related to reworking of mud pebbles at the sediment surface (Kugler 1987; Rais et al. submitted), and must predate albite authigenesis. The texture of the limestone does not influence the albite development, whereas the strong correlation between albite abundance and carbonate content suggests that the chemistry of the host rock is an important control. This relation is surprising as the difference of carbonate content between sediments containing albite and those without albite seems to be too low to create extremely different chemical conditions.

7.2. Distinctive features of the albites from Nissibach

Kastner & Siever (1979) estimated that most authigenic albites fall in the 60 to 120 µm range. In Nissibach, crystals range from 50 to 400 µm with an average of 200 µm. Previous studies have shown that the size of the albite crystals is closely linked to the maximum thermal grade of the host rock (Richter 1978; Brauckmann 1984). Thus, the albites from Nissibach are relatively large compared to other occurrences of similar burial grade. Brauckmann (1984) reported authigenic albites from Bramsche and Vlotho (Germany) of a few micrometers to 294 µm. In the northern England Pendleside Formation albites size is of 120 to 150 μ m in average in packstones, and up to 400 µm in grainstones. Authigenic albites can reach several millimeters in sediments which experienced epizonal conditions, like in Triassic dolostones of Crete (up to 3.5 mm) (Kastner and Siever 1979), or in Roches des Amoureux and Roten Turra (up to 10 mm) (Rose 1865; Jakob & Neher 1951).

The albites from Nissibach have the typical nearly-pure chemical composition of authigenic albites, although the albite

Fig. 6. Relative sequence of the sedimentologic and diagenetic events.

component is slightly lower than previous reports, with values ranging from 96.77 to 99.60 mol% (except 93.57 mol% measured on the edge of a crystal), and an average of 98.36 mol%. Richter et al. (2002) reported albite contents of 98.78 to 99.87 mol% (99.35 in average) from outcrops in Germany and Greece, whereas Spoetl et al. (1999) concluded that most of authigenic albites have an albite content typically above 99 mol%. They also showed that, in contrary to crystal size, albite composition is not related to the grade of thermal alteration of the host rock.

7.3. Albite formation models

Two principal models have been proposed to explain the provenance of the chemical components required for albite authigenesis: a model based on isochemical transformations (Kastner & Siever 1979), and a model including migration of deep saline fluids (Kastner & Siever 1979; Schedl et al. 1992; Spoetl et al. 1996).

The isochemical transformation model is a simple but pertinent model, where all the components necessary for albite authigenesis are supplied by the host rock. Sodium is concentrated in seawater trapped in the sediment pores or can be released by clay mineral transformation during diagenesis. Silica can also be supplied by clay minerals, or by the dissolution of biogenic siliceous fossils (sponge spicules, diatoms, etc). Aluminum can be released by the decomposition of Al-rich smectite (montmorillonite), which is a common detrital constituent of hemipelagic sediments.

The deep-brine migration model proposes a chemical exchange between the components of the host rock and migrating saline fluids. The chemical composition of the brines depends on their origin (meteoric, seawater, or hydrothermal), and on the lithology of the rocks through which they have passed. According to Kastner & Siever (1979), the most likely origin of these brines are hydrothermal waters, which migrate upward carrying high concentrations of H_4SiO_4 and Na^+ . In this model, aluminum species, which are hardly soluble, are supplied directly from the sediment.

The albite formation model applied to the studied sediments has to be consistent with the geological history of the region. The components required for albite formation according to the isochemical model, are present in the sediments of Nissibach and Milchbach. Moreover, deep burial transformations of the clay minerals during Alpine orogenesis (Wang et al. 1996) could have provided the chemical components for albite authigenesis. However, this model does not explain the restricted occurrence of authigenic albite in the region and its general scarcity. If clay diagenesis in limestones were the only requirements for its formation, albite would be expected to be a common mineral of these rocks. Contrary to the isochemical model, the brine migration model could explain the local occurrence of authigenic albite, forming along fluid migration paths, and evidence for hydrothermal fluid migration is reported from the Gonzen Mountain, situated a few kilometers away from Nissibach. Fluids having a temperature of about 100 °C migrated through a Jurassic fault system affecting the continental basement and overlying sediments. They generated a massive Fe-Mn ore body deposited in alternation with Early Kimmeridgian limestones (Pfeifer et al. 1988). This model implies an albite formation approximately 5 Ma after sediment deposition, whereas in the isochemical model, albite grew under deep diagenetic burial conditions during Alpine orogenesis. Therefore, the burial conditions at which albite formed are a key to the understanding of albite origin.

7.4. Constraining the depth of authigenesis

The textural relation of the albites with other components of the rock gives insight on the relative sequence of the events (Fig. 6). In Nissibach, albites grew after a first generation of calcite-filled veins and in association with secondary pyrite (Spoetl et al. 1999). Albite and calcium carbonate are dissolved at the edge of nodules by pressure solution (Kastner & Waldbaum 1968; Frey et al. 1973; Beach 1979), indicating that pressure solution processes were still active after albite formation. A last generation of calcite veins cut through the albite crystals. These observations support a formation of albite during deep burial diagenesis.

Another argument in favor of a diagenetic origin is that the maximum burial rate of the host rock has an effect on the size of the crystals, which suggests a continuous albite growth during burial diagenesis (Richter 1978; Brauckmann 1984; Spoetl et al. 1999). In Nissibach, the crystal size varies around 200 µm, which is consistent with deep diagenetic burial conditions.

8. Conclusions

The albites found in the sedimentary rocks of the Glarus Nappe are of authigenic origin. The chemical composition of the host sediment is determinant in albite formation, as illustrated by the irregular distribution of albites in the Nissibach section. Albite crystals apparently formed during deep burial diagenesis conditions, in association with other diagenetic processes. Clay mineral transformation is a potential important source of ions for albite growth. The apparent local distribution of albite remains enigmatic and would deserve further investigation.

Acknowledgements:

This project was supported by the Swiss Science Foundation, grant No. 2- 77549-04. We thank Thomas Pettke for the interesting discussion on fluid inclusions.

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Manuscript received July 5, 2006

Revision accepted July 1, 2007

Published Online First September 22, 2007