PROBABLE EXISTENCE OF REWORKED OPAL-CT IN MIOCENE
SEDIMENTS FROM THE ASAMAI DISTRICT,
AKITA PREFECTURE, JAPAN

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Abstract The $d(101)$ spacing of opal-CT can be used as a detailed scale of silica diagenesis and as an indicator to infer the thermal history of sediments. However, it is naturally expected that the observed $d(101)$ spacing is constrained according to the coexistence of reworked opal-CT and diagenetically formed opal-CT. The same is also expected when two groups of opal-CT are formed at quite different stages of diagenesis.

In this paper, a probable example of coexistence of reworked opal-CT and diagenetically formed opal-CT is described, and geological implications of reworked opal-CT are discussed.

Introduction

Opal and opaline silica are classified into the three categories, opal-A, opal-CT and opal-C; opal-A is highly disordered silica near amorphous, opal-CT is low cristobalite unidimensionally disordered by randomly interstratified stacking of tridymite layers and opal-C is well ordered low cristobalite (JONES and SEGNET, 1971). In addition to these categories, possible presence of tridymite-like mineral has been suggested by several workers (MITCHELL and TUFTS, 1973; IJIMA and TADA, 1981). Opal-CT changes its internal structure to opal-C or well-ordered low cristobalite by heating. This ordering occurs at the expense of tridymite layers and results in the progressive decrease of $d(101)$ spacing of opal-CT (FLÖRKE, 1955; JONES and SEGNET, 1971). In this respect, there are no strict guidelines to distinguish opal-C from opal-CT.

A number of studies on the transformation of silica and silica diagenesis have verified

that opal-CT is an intermediate product in the transformation of opal-A to quartz and the $d(101)$ spacing of opal-CT progressively decreases from 4.12 to 4.04 Å (MIZUTANI, 1966, 1977; VON RAD & RÖSCH, 1974; MURATA & NAKATA, 1974; MITSUI & TAGUCHI, 1974, 1977; MURATA & LARSON, 1975; VON RAD *et al.*, 1978; KANO, 1979; KANO & TAGUCHI, 1980; IJIMA & TADA, 1981; PISCOTTO, 1981). The transformation rate of silica and the decreasing rate of $d(101)$ spacing of opal-CT are generally governed by temperature and chemical conditions (MIZUTANI, 1966, 1977; ERNST & CALVERT, 1969; KASTNER *et al.*, 1977; KANO, 1979; KANO & TAGUCHI, 1980).

In the previous studies, most of workers have had a tendency to consider simply that opal-CT in sediments is formed from opal-A in situ. However, IJIMA & TADA (1981) have found two groups of opal-CT in a type of siliceous sediments; one is progressively ordered opal-CT and the other is additionally cemented opal-CT (tridymite?) with a larger $d(101)$ spacing than that in the host sediments.

In addition to this finding, we recently found two groups of opal-CT, one of which may be of reworked origin, in a glauconite-bearing sandstone of the Miocene Sugota

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Fig. 1. Map showing the studied area and sampling point (o).

Table 1. Stratigraphy in the Asamai district (modified after OZAWA *et al.*, 1979)

Geological Age		Stratigraphy
Quaternary	Holocene	Alluvium
	Pleistocene	Terrace deposits
Neogene	Miocene	Funakawa Formation (150-400)
		Onnagawa Formation (150-450)
		Sugota Formation ? (100-300)
		Hatamura F. (100-400)
		Iizawa Formation (300-500)
Cretaceous		Kamuroyama Granites

() m in thickness

Formation in the Asamai district, Akita Prefecture, north-eastern Japan.

In this paper, we discuss the probable existence of reworked opal-CT in the sandstone sample and geological meaning of reworked opal-CT.

Geological setting

The Asamai district is located on the east side of the Dewa Hilly Land near Yokote city (Fig. 1). Detailed geology of this district has been reported by OZAWA *et al.* (1979). For the present study, the Iizawa, Hatamura and Sugota Formations are stratigraphically important (Table 1). The Iizawa Formation, the lowest strata composed of andesitic volcanics is overlain by the Hatamura Formation with local unconform-

ity. The Hatamura Formation consists of acidic and andesitic volcanics, sandstone, mudstone and conglomerate, and yields the Daijima-type Flora and silicified wood abundantly. The Sugota Formation consists mainly of sandstone and conglomerate with basaltic volcanics, acidic pyroclastics and mudstone. This formation overlies the Hatamura Formation with local unconformity and furthermore, overlies directly the Iizawa Formation in part. The Onnagawa Formation and the Funakawa Formation consist mainly of siliceous rocks (so-called hard shale) and of mudstone, respectively. The mudstone member of the upper part of the Sugota Formation is thought to be in intertonguing relationship with the lower part of the Onnagawa Formation for the similarity of lithology. However, this member can be differentiated from the Onnagawa Formation with the presence of a foraminifera assemblage of the Nishikurosawa stage (Middle Miocene) (OZAWA *et al.*, 1979). According to OZAWA *et al.* (1979), the lower and middle parts of the Sugota Formation are of littoral to neritic facies, and the upper part of the Sugota Formation and the Onnagawa Formation are of bathyal facies.

Description of sample

A glauconite-bearing sandstone sample was collected from the upper part of the Sugota Formation near Niiyama in the southwest Omori town (Fig. 1). Mineral constituents were identified through X-ray diffractometry, and the $d(101)$ spacing of opal-CT was determined referring to the internal standard peak of quartz($10\bar{1}0$). Scanning was done at a speed of 1/4 degree per minute. The pre-

cision of $d(101)$ spacing determined with this method is usually ± 0.005 Å.

The sandstone sample studied includes quartz, plagioclase, pumice(?), andesite or dacite rock fragments, glauconite, pyrite, diatoms, sponge spicules and other siliceous fossils, and the matrix consists of opaline silica and clays. Pyrite has globular shape. Glauconite closely associates with opaline silica and occasionally replaces siliceous fossils and pumice-like materials. Volcanic materials, plagioclase and most of quartz are evidently detrital. Authigenic quartz (chalcedony) is rarely found in siliceous fossils.

Interestingly, the sandstone sample also contains small fragments of silicified wood ranging from 2 mm to 5 cm in length and from 1 mm to 1 cm in width (Plate I). The fragments have an angular to subangular shape with fibrous texture and fractures of various dimensions. The fractures clearly cut the silicified fibers in vertical or oblique, or subparallel directions (Plate I). Large fractures are filled with very small fragments of silicified wood and other constituents of the host sandstone, and microscopically observable fractures are filled with silica cement, fine-grained silicified wood and clastic quartz. Usually, silicified wood fragments show a wood texture such as fibers and cells. On the other hand, some marginal part of the fragments does not have such textural characteristics and interlocks the sandstone matrix in places (Plate I-3). The width of the marginal part is usually tens μm and 200–300 μm in maximum. Opaline silica and authigenic quartz interlocking each other are common in the silicified wood fragments (Plate I-4). Globular pyrite and other opaque materials, and very rarely glauconite are also observed.

Opal-CT in the glauconite-bearing sandstone sample

In the glauconite-bearing sandstone sample, two modes of occurrence of opal-CT can be identified; one is in silicified wood fragments and the other is in the host sandstone.

The X-ray diffractogram of silicified wood fragments shows mainly quartz and opal-CT

Fig. 2. X-ray diffractograms of samples.

Q: quartz, C: opal-CT, T: tridymite peak of opal-CT, P: Plagioclase and Cl: clays.

peaks (Fig. 2). Opal-CT shows a (101) peak which has a broad top seemingly splitted into two. The width of peak top is about 0.02 Å and the center of the broad top is at 4.094 Å. On the other hand, the X-ray diffractogram of the host sandstone, that was taken after silicified wood fragments had been carefully removed by hand-picking, shows a rather sharp (101) peak of opal-CT at 4.102 Å (Fig. 2). Detrital quartz is much more abundant than authigenic quartz in the host sandstone, while quartz in silicified wood fragments is authigenic. Moreover, the X-ray diffraction peak-height ratio of quartz(1010)/opal-CT(101) of silicified wood fragments is about 3.0, much larger than 0.8 of the host sandstone. Thus, the ordering of opal-CT and the transformation of opal-CT to quartz in silicified wood fragments seem to be at more advanced stages than those in the host sandstone.

However, the coexistence of opal-CT with rather larger $d(101)$ spacing and abundant authigenic quartz in silicified wood fragments contradicts with an observation that quartz is abundantly formed from opal-CT, usually after the $d(101)$ spacing has reached about 4.06 Å (KANO, 1979). On the other hand, IJIMA & TADA (1981) reported that a mix-

ture of two opal-CT samples with quite different $d(101)$ spacings shows a rounded or shouldered peak at a place between the original positions of (101) peaks of two opal-CT samples. This evidence enables us to explain easily the above contradiction, by supposing that silicified wood fragments include two groups of opal-CT. In this case, the two groups of opal-CT must have a $d(101)$ spacing larger than 4.094 Å and that smaller than 4.094 Å, respectively. Probably, the latter value is smaller than 4.06 Å. Actually, the (101) peak profile of opal-CT in the silicified wood fragments has an undulating broad top which seems to show the appearance splitted into two. This profile resembles that of a mixture of two opal-CT samples rather than sharp (101) peak profile observed for the host sandstone. The magnitude of undulation at the peak top splitted into two is small within the range of potential fluctuation in X-ray diffraction intensity, namely within the twice standard deviations of the measured intensity that is statistically estimated according to the condition of X-ray diffractometry. Therefore, we cannot determine surely whether the splitted peaks show real peaks or not. However, in either cases, the $d(101)$ peak profile observed for silicified wood fragments would be of a mixture of two groups of opal-CT with different $d(101)$ spacings since other constituent minerals have no strong peaks around 4.1 Å.

As stated in the foregoing, fractures cut the silicified wood fragments. The fracture surfaces seem to be curved or edged relatively smoothly, though microscopically they appear somewhat irregular. Such fracture will be possible to be formed only when the original wood has become more rigid and brittle state, namely has been silicified. Large fractures in the fragments are filled with various sandstone constituents and such large fractures cannot be found in the other sandstone constituents (Plates I-1 and I-2). Therefore, the large fractures must have been formed before or at the time of deposition of the sandstone. Consequently, the wood fragments must have been silicified before the incorporation into the host sandstone.

The fact that glauconite minerals replace siliceous fossils and closely associate with silica cement in the sandstone indicates that a dissolution-reprecipitation reaction of silica has started at a glauconite formation stage, probably soon after the deposition of the sandstone. Glauconite is usually formed at the sea bottom or below that. Therefore, at that stage, silica must have precipitated in fractures and other minute interstices of silicified wood fragments, and the marginal part and fracture surfaces of the fragments would have reacted to interstratal solution. Actually, some marginal part of silicified wood fragments has no distinct textural characteristics while the main part has an original wood texture. In some cases, the margin of the fragments interlocks the sandstone matrix in microscopic scale and closely contacts with glauconite (Plate I-3). Microfractures are filled with silica, which seems to correspond to an extension of silica cement in the sandstone matrix. Authigenic glauconite in silicified wood, even though very rarely found, possibly demonstrates the permeation of chemical species into interstices during the silica reaction. The facts mentioned here indicate that some of silica was precipitated in the silicified wood fragments during the diagenesis of the host sandstone. Though, the first silicification of the fragments occurred before the incorporation into the host sandstone, as suggested from the mode of occurrence of large fractures in the fragments.

According to KASTNER *et al.* (1979), clays delay the transformation of opal-A to opal-CT. Such chemical effects may control the ordering rate of opal-CT and the transformation rate of silica. In this respects, even if all opal-CT in silicified wood formed during diagenesis of the host sandstone, the difference of chemical conditions between the silicified wood fragments and the host sandstone might have resulted in differences in the $d(101)$ spacing and the transformed amount of silica minerals. At present, of course, we cannot perfectly deny the above possibility in the formation of two groups of opal-CT in the studied sample. However, since the result of X-ray analysis of the silicified wood

fragments is reasonably interpreted to show the coexistence of two groups of opal-CT as stated before and since it is believed that the chemical environment of the fragments has not been so much different in places after the host sandstone was cemented with silica, it is considered that the formation stage of each group of opal-CT would have been quite different each other. It is very likely that opal-CT with the smaller $d(101)$ spacing in the silicified wood was formed first and somewhat ordered in the provenance sediments, and then it has been more ordered after its incorporation into the host sandstone, while the other one with the larger $d(101)$ spacing in the silicified wood fragments and the host sandstone have been formed and progressively ordered during the diagenesis of the sandstone. Opal-CT with the larger $d(101)$ spacing has been likely formed through silica addition from the host sandstone. Also, it may have been formed from opal-A remaining in the fragments if opal-A is the first precipitate in the fragments.

From the above mentioned, it is suggested that opal-CT with the smaller $d(101)$ spacing associated with the silicified wood fragments was incorporated into the glauconite-bearing sandstone.

Discussion

Since MURATA & NAKATA (1974) and MITSUI & TAGUCHI (1974, 1977) reported that the $d(101)$ spacing of opal-CT decreases progressively in the early stage of diagenesis, an increasing number of studies on this phenomenon have been presented (*e.g.* MURATA & LARSON, 1975; MIZUTANI, 1977; VON RAD *et al.*, 1978; KANO, 1979; IJIMA & TADA, 1981). The decreasing rate of $d(101)$ spacing seems to be expressed by the differential equation,

$$dS/dt = -kS \quad (S(A) = d(101) - 4.040) \quad (1)$$

where t is the elapsing time, and k is the rate constant which is expressed by Arrhenius' equation if the chemical condition is constant (KANO, 1979; KANO & TAGUCHI, 1980).

According to MIZUTANI (1977), the $d(101)$ spacing can be used as a detailed scale of

silica diagenesis and as an indicator to infer the thermal history of sediments. However, as stated before, reworked opal-CT itself has a smaller $d(101)$ spacing than that of in situ opal-CT in the host sediments. The coexistence of reworked opal-CT with in situ opal-CT formed diagenetically will also provide an apparent smaller $d(101)$ spacing than that of opal-CT formed diagenetically in situ but larger than that of reworked opal-CT. Therefore, reworked opal-CT leads to misinterpretation of silica diagenesis and thermal history of sediments unless the existence of reworked opal-CT can be ascertained. Though any sure methods to identify directly the existence of reworked opal-CT have not been established yet, the observed $d(101)$ spacing and other available geological informations may enable us to infer the existence of reworked opal-CT.

Then, the $d(101)$ spacing of reworked opal-CT, together with other geological informations can be used to infer the history of silica mineral changes in the provenance sediments and would offer the information of the thermal history of the provenance sediments.

A parameter S'_0 of $d(101)$ spacing of reworked opal-CT at the redeposition is estimated by using the integration of eq. (1) in a range from the redeposition ($t=0$) to the present ($t=t$). If opal-A is continuously converted to opal-CT without delay of the reaction since the beginning of diagenesis, the integration of eq. (1) is

$$\ln \frac{S'}{S'_0} = \ln \frac{S}{S_0} = - \int_0^t k dt \quad (2)$$

where S and S' are the S of authigenic opal-CT and reworked opal-CT at present ($t=t$), and S_0 and S'_0 are those at the deposition ($t=0$), respectively. KANO (1979) roughly estimated S_0 at 0.09 ± 0.01 Å. Putting this value and S and S' observed for a sample into eq. (2), S'_0 can be readily calculated. For example, S of the glauconite-bearing sandstone and S' of silicified wood fragments are 0.062 and a value smaller than 0.054 Å, respectively. If chemical conditions do not significantly affect the ordering of opal-CT, S'_0 is calculated at a value smaller than

0.08 Å. Then, the $d(101)$ spacing of reworked opal-CT was smaller than 4.12 Å at the incorporation into the sandstone. Furthermore, if the present $d(101)$ spacing of reworked opal-CT is 4.06 Å or smaller as stated before, this value was 4.08 Å or smaller. The calculated $d(101)$ spacing of opal-CT in the silicified wood at the time of deposition would indicate that opal-CT had been formed in silicified wood before the redeposition.

The glauconite-bearing sandstone sample includes andesite or dacite rock fragments which are common in the Hatamura and Iizawa Formations. Silicified wood is abundant in the Hatamura Formation. Moreover, the Sugota Formation overlies the Hatamura Formation with local unconformity and directly overlies the Iizawa Formation in part. Thus, the silicified wood fragments in the glauconite-bearing sandstone sample is believed to have been supplied from the Hatamura Formation around the Asamai district in the Dewa Hilly Land. Judging from the thickness of the formation, the sediments of the Hatamura Formation were buried to 100–400 m deep in maximum, at the beginning of deposition of the Sugota Formation. According to UTADA (1971), the Dewa Hilly Land seems to belong to (H) terrane, one of terranes classified with characteristics of zeolitic zoning in the Neogene pyroclastics, and the geothermal gradient of (H) terrane can be estimated in a range from 4.5 to 6°C/100 m. Then, using the surface temperature of 15°C and the above range of geothermal gradient, the maximum temperature which the Hatamura Formation had experienced before the beginning of deposition of the Sugota Formation will be estimated at 20–39°C. Moreover, the Hatamura Formation was deposited in the Daijima stage which ranges from 15 to 21 Ma or older (IKEBE *et al.*, 1973). Therefore, the time interval of deposition of the Hatamura Formation would have been 3–4 Myr or longer. On the other hand, according to PISCIOTTO (1981), the bulk of the silica transformations occurred during the last 3–4 Myr and within narrow temperature range (9–27°C for opal-A to opal-

CT and 35–61°C for opal-CT to quartz) in the Monterey Shale, California. Thus, presumably, opal-CT had been formed in the silicified wood of the Hatamura Formation before the beginning of deposition of the Sugota Formation.

Silicified wood samples in the Hatamura Formation which were collected from the Dewa Hilly Land near Ōmori town by ITANO (1980 MS) are mostly made of quartz. On the other hand, reworked silicified wood in the glauconite-bearing sandstone includes opal-CT and quartz. This difference of silica phases found in the silicified wood samples from the Hatamura and Sugota Formations, considering together with the geological history in the Asamai district, suggests the following sequence of diagenetic history of silicified wood: 1) Silicification of wood fossils occurred in the Hatamura Formation. 2) In the earlier time of opal-CT stage of silica transformation (probably during the time of deposition of the Sugota Formation), a part of the Hatamura Formation was eroded and some of silicified wood were incorporated into the sediments of the Sugota Formation. 3) Silicified wood in the Hatamura Formation had been buried to deeper part during continuing deposition and silica in the wood had been transformed to quartz. 4) On the other hand, reworked silicified wood in the Sugota Formation had been buried also, but not so deep. Thus, opal-CT in reworked silicified wood has not been transformed to quartz wholly.

If some geological conditions to result in the deposition of reworked opal-CT were present around the Dewa Hilly Land as suggested in this study, it is supposed that fine-grained reworked opal-CT may have been also incorporated into the Onnagawa Formation overlying the Sugota Formation. Although we have no exact evidence in this respect at present, the fact that opal-CT in some siliceous rocks collected from the Onnagawa Formation in the studied area shows a (101) peak profile similar to that observed for silicified wood fragments seems to support the above proposal.

Summary

The mode of occurrence and the X-ray diffraction analysis of opal-CT in a glauconite-bearing sandstone sample from the Sugota Formation suggest the incorporation of reworked opal-CT into the sandstone, even though incomplete points to ascertain the presence of reworked opal-CT still remain. The geological evidence also suggests that there had been the source area to provide reworked opal-CT around the Dewa Hilly Land during the deposition of the Sugota Formation.

Reworked opal-CT is geologically very important. If the presence is overlooked, reworked opal-CT leads to misinterpretation of silica diagenesis and the thermal history of associated sediments. On the other hand, reworked opal-CT can be used to infer the history of silica mineral changes in the provenance sediments and would offer the information of the thermal history of the provenance sediments.

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秋田県浅舞地方の中新世堆積物中の再堆積したと思われるオパール CT について

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(要 旨)

オパール CT の d(101) 値は、珪酸の続成作用の詳細な尺度や堆積物の熱史を推定するための指標として用いられる。しかしながら、続成作用でできたオパール CT と再堆積したオパール CT が共存する場合、観察される d(101) 値は、ゆがめられたものになると考えられる。続成過程にあって、かなり異なった時期に形成された2種類のオパール CT が共存する場合も同じことが起こり得る。

本論では、続成作用でできたオパール CT と再堆積したオパール CT が共存すると思われる例について報告するとともに、再堆積したオパール CT のもつ地質学的意義について議論する。

[地 名]

Asamai 浅舞

Niiyama 仁井山

Ōmori 大森

Yokote 横手

Dewa Hilly Land 出羽丘陵

[地層名]

Iizawa F. 飯沢層

Hatamura F. 畑村層

Sugota F. 須郷田層

Onnagawa F. 女川層

Funakawa F. 船川層

Explanation of Plate

Plate I

1. A sketch of glauconite-bearing sandstone indicating the areas of pictures 2, 3 and 4. Fractures cut silicified wood (horizontally ruled area).
2. Microphotograph of area 2. Fractured surface of silicified wood is relatively smoothly curved or edged.
3. Microphotograph of area 3. Small fractures cut silicified wood. The marginal part of silicified wood has no distinct textural characteristics and interlocks with the sandstone matrix. White arrow indicates glauconite replacing a diatom.
4. Microphotograph of area 4. 4a shows a fibrous and cell-like texture of silicified wood, in which opaline silica (dark part) and quartz (light part) are observed (4b; crossed nichols).