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Crystallinity indices of quartz in chert of Central Japan: their reliability and some geological applications

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(Abstract) Crystallinity indices of quartz (CI) determined by X-ray analysis have been reported in various types of chert of different ages. Reported CI's distribute from <1 to 10. The difference may result from the difference in sample state prepared for determination of CI or existence of essential variation in crystallinity of quartz. We determined CI of many euhedral quartz crystals and quartz in cherts in different sample states, and could conclude that CI does not depend largely on sample states prepared with different methods, but depends partially on euhedral macroquartz used as a standard material. Even after removing this disadvantage, there is still wide difference in CI of quartz, indicating that CI is not uniform but varies according to the mode of origin and the thermal processes having operated on the quartz.

Our data coupled with data described in literature show that 1) CI's in Cenozoic deep-sea oozes and cherts are lower than those in Mesozoic bedded cherts on land, 2) bedded cherts suffering thermal effect are characterized by higher CI, 3) CI's in lacustrine and fluvial cherts are higher than those of bedded cherts of deep-sea origin, 4) CI's of older cherts are generally larger than those of younger cherts of the similar origin.

This analysis suggests that CI can be used as a useful indicator deciphering origin of sedimentary siliceous rocks and their diagenetic and metamorphic stages.

Key words : crystallinity index, quartz, chert, diagenesis, provenance, thermal effect, XRD

1 Introduction

It seems undoubted that there is a wide range of crystallinity of quartz in chert on land and under the sea. The crystallinity index (CI) defined by Murata and Norman (1976) is widely applied to describe characteristics of quartz in chert. For example, Deutsch et al. (1989) showed a clear relation of DTA (Differential Thermal Analysis) peaks and CI between 1 and 10. CI is based on the degree of resolution of the *d* (212) X-ray reflection at 1.3820 Å (Fig. 1). **a** and **b** of a standard euhedral macrocrystal of quartz (hereafter referred to as $\mathbf{a}_s$ and $\mathbf{b}_s$, respectively) are measured and **F** is determined as ($\mathbf{b}_s/\mathbf{a}_s$). Crystallinity of other quartz characterized with **a** and **b** can be calculated as CI=10・**F**・(**a**/**b**), where **F** is called the scaling factor. Of course, the standard euhedral macrocryst gives CI of 10. CI is very convenient to know X-ray characteristics of quartz because it can be determined only through XRD (X-ray Diffraction) measurement. CI's often are reported from various types of rocks of many localities as listed in Table 1.

What implication does CI have in a geological sense? Difference in CI means that there are many variations in a mineral species called quartz. Do the variations result from lattice distortions and defects in quartz crystals? From impurities included in quartz crystals? From difference in grain-size of quartz crystals (large or small)? From difference in crystal shapes of quartz (euhedral or unhedral)? Does CI change through diagenesis and metamorphism, and with age?

CI has been recorded in the range of 67° to 69° of 2θ of XRD patterns (Murata and Norman, 1976). However, as occasionally pointed out, CI determined through Murata and Norman's method seems to be partly a function of

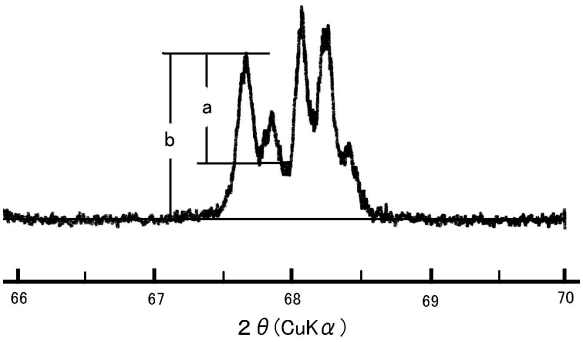

Fig. 1. XRD chart of quartz showing five peaks around 2θ =68°. Crystallinity index for quartz is determined from ratio, a/b, and scaling factor, **F**.

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Table 1. Crystallinity index of quartz described in different localities and different chert types. Data from chert cobbles in conglomerates are discarded.

Sample	Mineral or Rock	Host Formation	Age of Host Formation	Locality	**CI**	Reference and Notes	scaling factor
1	chert from magadiite	Rome beds (informal name)	Pliocene	Malheur County, Oreg.	1.3	Murata and Norman, 1976	F=1.67
2	bedded chert	Monterey Shale	Miocene	Temblor Range, Calif.	2.0-3.2 (8)		F=1.67
3	chert (black flint)	chalk	Cretaceous	England	<1.0		F=1.67
4	chert	Franciscan Formation	Jurassic and Cretaceous	Cerro Colorado quad., San Benito, Co., Calif.	8.0-9.0 (2)		F=1.67
5	bedded chert	Franciscan Formation	Jurassic and Cretaceous	Penitencia Creek, Santa Clara County, Calif.	8.5		F=1.67
6	replacement bedded chert	Calera Limestone, member of Franciscan Formation	Jurassic and Cretaceous	Permanente quarry, Santa Clara Co. Calif.	6.2 (2)		F=1.67
7	novaculite	Caballos Novaculite	Devonian and Mississippian	Marathon Basin, Tex.	2.2 (2)		F=1.67
8	novaculite	Arkansas Novaculite	Devonian and Mississippian	Hot Springs Region, Ark.	5.7-9.8 (7)		F=1.67
9	replacement bedded chert	dolomite in Allomoore Limestone	Precambrian	Van Horn, Tex.	4.8		F=1.67
10	bedded chert	Ironwood Iron-Fm	Huronian	Gogebic Range, Mich.	6.5-7.0 (2)		F=1.67
11	wood	Noroly Sandstone	Miocene	Tesla, Alameda County, Calif.	1.1		F=1.67
12	wood	Lamar River Formation	Eocene	Yellowstone Natl. Park, Wyo.	1.8		F=1.67
13	*Tempskya* trunk	----------	Cretaceous	Greenhorn, Oreg.	<1.0		F=1.67
14	wood	Chinle Formation	Triassic	Holbrook, Ariz.	2.2-3.5 (2)		F=1.67
15	*Osmundites* root sheath	----------	Triassic	Mt. Augusta and Fremouw Peak, Antarctica	1.4-1.7 (2)		F=1.67

16	silicified peat	----------	Permian	West Coalsack Bluff, central Antarctica	4.1		F=1.67
17	wood	Tradewater Formation	Pennsylvanian	Christian Co., Ky.	4.6		F=1.67
18	*Psaronius* with quartz druses	Monograhela Formation	Pennsylvanian	Shade, Ohio	8.9		F=1.67
19	*Calixylon*	----------	Devonian	Kettle Point, Ont., Canada	1.2		F=1.67
20	chalcedony rim	John Day Formation	Oligocene and Miocene	Madras, Oreg.	1.0		F=1.67
	macroquartz core				9.3		
21	chalcedony rim	volcanic rocks	Tertiary	Chihuahua, Mexico	2.4		F=1.67
	macroquartz core				9.2		
22	chalcedony rim	basalt	Triassic	Rio Grande do Sul, Brazil	<1.0		F=1.67
	macroquartz core				9.0		
23	chalcedony rim	basalt	Keweenawan	Hibbard Bay, Ont., Canada	3.3		F=1.67
	macroquartz core				9.0		
24	chalcedony rim	amygdaloid Island Flow	Keweenawan	Isle Royal, Mich.	4.7		F=1.67
	macroquartz core				9.2		
25	petrified wood	----------	ca. 20Ma	----------	ca.1.2	Stein, 1982	---------
		----------	ca. 35Ma	----------	ca.1.6		
		----------	ca. 50Ma	----------	ca.1.9		
		----------	ca. 170Ma	----------	ca.2.1		
		----------	ca. 200Ma	----------	ca.8.0		
			ca. 350Ma		ca.5.0		
26	diagenetic quartz	calcareous rocks in deep-sea sediments	late Miocene	equatorial Pacific near Cost Rica Ridge	<1.0-2.1 (21)	Hein, Sancetta, and Morgenson, 1983	F=1.67

27	nodular chert	limestone	lower Cretaceous	Carpathian Mts. Czech and Slovakia	1.7-3.0 (4)	Lintnerova and Petercakova, 1994	F=1.99
28	gray chert	chert in melange	Permian	Takayama, Mino Terrane, Central Japan	5.6	Ui and Mizukami, 1994	F=1.37
29	red and gray cherts	chert in melange	Jurassic to Triassic	Niitsu, Mino Terrane, Central Japan	9.2 (2)	Ui and Mizukami, 1994 1.5km from granite	F=1.37
30	red, bluish green, black and white cherts	bedded chert	Triassic	Kamiaso, Mino Terrane, Central Japan	3.9-5.2 (8)	Ui and Mizukami, 1994	F=1.37
31	red chert light gray chert	bedded chert	Triassic	Inuyama, Mino Terrane, Central Japan	3.7 7.3	Ui and Mizukami, 1994	F=1.37
32	gray chert gray chert greenish blue chert gray chert gray chert gray chert gray chert red and gray chert	bedded chert	Triassic	Kasuga, Mino Terrane, Central Japan	7.8-10.1 (8) 9.8-10.2 (4) 10.3 9.9-10.5 (4) 8.4-10.0 (2) 7.6-10.0 (3) 6.6-9.9 (3) 4.8-5.2 (3)	Ui and Mizukami, 1994 <1 km from granite 1-2 km from granite 1.4 km from granite 2-3 km from granite 3-4 km from granite 4-5 km from granite 5-6 km from granite 6-7 km from granite	F=1.37
33	white chert	chert layer in volcaniclastic rocks	Miocene	Niu Mountains, Fukui, Central Japan	2.44-4.56	Hattori et al., 1996	F=1.4
34	epigenetic chert	sandstone	Jurassic	Akatani, Nanjo, Central Japan	5.0-7.9 (12)	Hattori, 1999a	F=1.33
35	red chert	bedded chert in Mino Terrane	Triassic	NW of Mt. Fujikura, Nanjo, Central Japan	1.3-3.1 (11)	Umeda, 1999	F=1.4

	blue green chert				2.4-4.2 (11)		
	white chert				3.2-5.8 (4)		
	black chert				4.8		
	transparent chert				2.8		
36	red chert	bedded chert	Triassic	Fujikura-dani route, Nanjo, Central Japan	2.2-3.2 (5)	Umeda, 1999	F=1.4
	bluish green chert				3.3-6.4 (12)		
37	core of chert nodule	Mg-clay deposits	Miocene	Madrid Basin, Spain	1.2-2.6	Bustillo, 2001	
	outer shell of nodule				ca. 5.0		
38	red, gray, brown, and black chert	bedded chert	Triassic	Tamba Terrane, Central Japan	3.6-9.3 (56)	Mikami et al., 2002 sample from wide area of Tamba Terrane	F=1.31
39	black to gray chert	bedded chert	Triassic	Tamba Terrane, Central Japan	5.9-9.7 (16)	This paper, western part in Fukui Prefecture	F=1.46
40	white brown chert	chert layers in volcaniclastic rocks	Miocene	Yachi, Kaetsu Hill, Fukui, Central Japan	5.1-7.0 (11)	Umeda, 2003	F=1.25
41	agates	igneous and metamorphic rocks and tuff	Precambrian	Western Australia	6.6-9.0 (6)	Moxon et al., 2006	---------
	agates	----------	Paleozoic	worldwide	4.7-6.6 (40)		
	agates	----------	Mesozoic	worldwide	1.6-5.0 (14)		
	agates	----------	Cenozoic	worldwide	1.0-4.6 (37)		
42	quartz in metapelite	greenschist facies metamorphic rocks	Eocene and Pre-Tertiary rocks	Taiwan	4.47-10.00 (33)	Lee and Yu, 1994 increase in CI with metamorphism	F=1.37*
43	quartz in chert	cores of nodules	late Jurassic	south-central Poland	<1.0-3.7	Migaszewski et al., 2006	---------

* The maximum crystallinity of 33 samples was assumed to be CI=10.0

( ) after **CI**: number of samples treated.

grain size of sample powders and may be affected also by lattice distortions induced by mechanical stress. There is a deterioration in diffraction peaks when the samples are too fine (Herdianita et al., 2000). The diffraction peaks change according also to the packing state: mounted firmly or loosely on holder plates (Klug and Alexander, 1954). If these operational variations are related significantly to the resultant CI's, comparison and correlation of different quartz crystals on the basis of CI are not reliable, and it is necessary that CI is described with the grain sizes of sample powders and the packing firmness of mounted powders. However, the determinations of these factors are very difficult, especially for practical use of CI by general geologists.

In this experiment, crystals of euhedral macroquartz (rock crystals) were crushed and ground many times to determine **b**/**a**. The result showed that no clear difference in the ratio is detected in an identical sample at least when rock chips are hand ground to powders in an agate bowls and pestles. Also no significant difference is recognized among cut surfaces, surfaces of polished thin sections and mounted powders of chert samples. However, **F** (=$\mathbf{b}_s/\mathbf{a}_s$) varies according to standard rock crystals used. After the confirmation of the stability of **b**/**a**, we determined CI of many cherts in central Japan: some are bedded cherts in the Mesozoic Mino Terrane and chert cobbles of Cretaceous, Paleogene, and Neogene conglomerates.

The crystallinity index defined by Murata and Norman (1976) is determined soley by XRD measurements. Unfortunately, we have no basis to tell what relation the CI has to the crystallinity in the sense of mineralogy and crystallography. The crystallilnity index (CI) used in the following description is only from the viewpoint of XRD measurement.

2 Method

Samples used in this trial are rock crystals, and white, black, green and red cherts in the Nanjo Massif of central Japan. Change in grinding time makes powder of different grain-sizes; longer grinding time produces finer powder. Thus, CI's of chert and rock crystals were determined for powders of different grain size. X-ray measurements were done not only for powders of chert but also for surfaces cut by diamond-blade attached rock-cutters and thin section surfaces to determine the effect of surface firmness and smoothness of samples. Cut surfaces and polished thin sections should be firmer than powder samples mounted on sample holders.

There are two problems to be clarified on the occasion of inter-laboratory comparison of CI. One is whether every rock crystal gives identical CI (=10), and the other is whether CI depends on grain-size and firmness of sample powders on powder holders. If different rock crystals have different CI, CI of other quartz, depends on the crystals used as standard samples. We determined **a** and **b** for powders with different grain-size of three rock crystals in different conditions. The rock crystals were crushed, ground, and powdered in agate bowls with agate pestles by handwork. **a** and **b** were determined for every still coarse-grained powder (the size is about 20 μm) as the first trial, then the powder was re-ground and re-determined as the second trial, and the powder was re-ground and re-determined as the third trial. The powders of the third and the fourth trials are viscous and adhere to walls of agate bowls. The agate is composed of quartz (chalcedony). We cannot deny that very little amount of quartz from the agate could contaminate to the sample powders. As we crushed and ground chert chips of 1 to 2 gr, the effect of contamination is thought to be negligible.

The first determination of CI of each chert was done for the cut surfaces of about 1 to 2 mm-thick sample plates. Thin sections without glass covers were polished with #800 polishing powder (1/800 in. that is ca. 32 μm) and subsequently with 1/4 μm diamond paste. Second and third XRD measurements were done for these samples at each polishing step and CI's were determined. As another trial, the remainder of chert chips were crushed, ground to produce powders of chert. Sample preparation was done repeatedly as described in the case of rock crystals, and the routine determination of CI was applied for powders of chert of different grain size.

The X-ray diffractometer used is the Toshiba ADG-301 Diffractometer and the measurement condition is 40kV, 30mA. 65° to 70° interval of 2θ is scanned at 1/4°/min with nickel-filtered copper radiation (CuKα). The time constant is 1 sec, and the range is 500. The slits used in the goniometer are S_1=1°, S_2=0.15mm, and S_3=1°. The chart speed is 1 cm/min. Powder samples were mounted in non-reflective glass holders, and were packed and flattened with stainless plates. Although this packing and flattening may produce preferred orientation of quartz powder on sample holders (Zhang et al. 2003), the effect seems to be negligible. The mean grain size of coarse powders treated in the experiments is smaller than 20 μm.

Table 2. XRD result of clear euhedral quartz crystals (coarse powder) after first, third, and fourth to sixth days after grinding (Step A1). **a** and **b** in cm.

Sample	First Day				Third Day				Fourth to Sixth Day			
	a	**b**	**a/b**	**CI**	**a**	**b**	**a/b**	**CI**	**a**	**b**	**a/b**	**CI**
violet amethyst	4.72	5.92	0.80	10	6.1	7.65	0.80	10	-	-	-	-
transparent crystal	4.1	5.5	0.75	9.4	6.1	7.85	0.78	9.8	4.76	6.01	0.79	10
Miocene crystal	4.35	6.25	0.70	8.8	3.65	5.4	0.68	8.5	4.7	6.6	0.71	9.0

In general, reproducibility of the results seems to depend mainly on the consistency with which the finely-ground powder is mounted in the non-reflective glass holder: firmness of packing and flatness of the sample surface being especially critical factors (Klug and Alexander, 1954, pp. 299). The reproducibility of twice continuous measurement for identical samples is very good: the experimental errors in peak height in continuous measurement for same samples are less than two percents. Dispersion of this level seems due to back-ground noise.

3 Reliability of Crystallinity Index

3.1 Crystallinity of Clear Euhedral Quartz Crystals

We prepared three different rock crystals as standards. They were ground stepwise. After first grinding forming powder sizes less than 20 μm, the first XRD measurement was done two or three times: first (just grinding day), third, and fourth to sixth days after grinding (Step A1) (Table 2). Then the powder was ground again. After the second grinding, the second XRD measurement was done (Step A2). After the third grinding, the third XRD measurement was done (Step A3). Further grinding seems to give no effect to the grain-size of powder. However, last grinding was done (Step A4) (Table 3).

Two of the three rock crystals were derived from the collection of Department of Geology, Fukui University: one is a 2cm long violet amethyst crystal, the other a 3cm long transparent crystal. The last crystal is a 1cm long Miocene transparent crystal lodged in Fukui City Museum of Natural History. For each XRD measurement, heights **a** and **b** were measured (Fig. 1). Height **a** was not influenced but **b** was influenced according to scan speeds of XRD and chart speed of recorders. It was reconfirmed that 1/4°/min of the scan speed and 1cm/min of the chart speed are necessary. CI was calculated by comparing with the sample giving the largest **a**/**b** (0.80 of the first and the third days of violet amethyst, that is CI=10)(Table 2).

If grinding of quartz makes crystal distortion and defect, and irregular binding of elements on surfaces of powder, these irregularity may be recovered with duration. In order to see any dependence of CI on the duration after grinding of quartz, we measured CI of the identical samples on the first, third and fourth to sixth days after grinding. Table 2 shows that duration after grinding seems to give no significant effect to CI of each sample. In the table, we can recognize significant difference of CI among crystals. This implies that CI changes depending on standard crystals which are used in determining **F**. **a** and **b** change considerably but variation of **a**/**b** is very small, and **a**/**b** is irrelevant to **F**. Based on this observation, we can say that it is better to use ratio, **a**/**b**, instead of CI.

The next experiment was done to assure the relationship between grain-size of powder and **a**/**b**(Table 3). Although the variation in the ratio represented in Table 3 may include back-ground noise and errors in a millimeter level at reading XRD charts, this evidence indicates that discussion of CI of chert should inevitably involve this level of error. It can be said that the difference in grain-size of the powder produced by handwork grinding gives this level of error (up to 15%) on the ratio.

Table 3. XRD result showing ratio, **a/b**, of clear euhedral quartz crystals. Step A1 is for coarse-grained powder (generally used for XRD. Third or Fourth to Sixth Day of Table 2), Step A4 is for finest grained powder. **a** and **b** in cm.

Sample	Step A1			Step A2			Step A3			Step A4		
	a	**b**	**a/b**	**a**	**b**	**a/b**	**a**	**b**	**a/b**	**a**	**b**	**a/b**
violet amethyst	6.1	7.65	0.80	4.22	5.97	0.71	2.2	3.26	0.67	-	-	-
transparent crystal	4.76	6.01	0.79	5.4	7.0	0.77	3.03	3.9	0.78	1.88	2.6	0.72
Miocene crystal	4.7	6.6	0.71	4.24	6.41	0.66	2.26	3.4	0.66	2.68	3.75	0.71

Table 4. XRD result with respect to Mesozoic cherts in the Nanjo Massif and at Unuma, Mino Terrane, Central Japan.

sample #	Step B1 (cut)			Step B2 (#800)			Step B3 (1/4 μ)			Step B4			Step B5			Step B6			Step B7		
and memo	**a**	**b**	**a/b**	**a**	**b**	**a/b**	**a**	**b**	**a/b**	**a**	**b**	**a/b**	**a**	**b**	**a/b**	**a**	**b**	**a/b**	**a**	**b**	**a/b**
Chert 1 (Unuma)	2.1	4.8	0.44	-	-	-	-	-	-	2.1	4.8	0.44	1.28	2.53	0.51	-	-	-	-	-	-
Chert 2 (near	3.35	7.1	0.47	1.5	3.4	0.44	1.4	2.5	0.56	2.5	4.5	0.63	2.72	4.85	0.56	1.24	2.36	0.53	-	-	-
dolomite)	2.15	3.9	0.55	1.7	3.4	0.50	1.8	3.45	0.52	-	-	-	-	-	-	-	-	-	-	-	-
	3.33	6.1	0.54	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
	2.0	3.38	0.59	0.71	1.3	0.55	0.88	1.61	0.55	1.3	2.42	0.54	1.34	2.3	0.58	1.34	2.47	0.54	1.17	2.77	0.61
Chert 3 (white)	1.63	4.55	0.36	1.8	5.0	0.36	1.75	3.6	0.49	2.1	4.85	0.43	1.4	3.8	0.37	0.94	2.29	0.41	-	-	-
	-	-	-	-	-	-	1.6	3.8	0.42	-	-	-	-	-	-	-	-	-	-	-	-
Chert 4 (red 1)	0.77	3.55	0.22	0.85	3.7	0.23	0.65	2.9	0.22	1.1	3.2	0.34	0.8	3.0	0.27	0.5	1.6	0.31	-	-	-
	-	-	-	-	-	-	1.0	4.0	0.25	-	-	-	-	-	-	-	-	-	-	-	-
Chert 5 (red 2)	1.15	4.0	0.29	1.3	4.3	0.3	1.0	3.7	0.27	0.85	2.6	0.33	0.76	2.34	0.32	0.6	1.63	0.37	-	-	-
	1.2	3.65	0.33	1.4	3.95	0.35	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
Chert 6 (green)	0.95	3.75	0.25	1.0	4.0	0.25	1.6	4.9	0.33	1.0	3.45	0.29	1.15	3.58	0.32	0.49	1.66	0.30	-	-	-
Chert 7 (black)	1.4	3.8	0.37	1.5	4.6	0.33	0.8	2.0	0.40	1.2	3.65	0.33	1.3	3.73	0.35	0.48	1.75	0.27	-	-	-

Chert 1 is a Triassic chert from Unuma, Gifu Prefecture. Chert 2 is a white chert bed associated with a dolomite layer, Chert 3 a white chert, Cherts 4 and 5 red cherts, Chert 6 a green chert, and Chert 7 a black chert from Nanjo Massif. Peak heights, **a** and **b**, and the ratios, **a/b**, were measured by XRD for cut surfaces (Step B1), uncovered thin section polished with #800 polishing powder (Step B2) and with 1/4 μm polishing powder (Step B3), ordinarily ground sample powder (Step B4), additionally ground powder (Step B5), further ground powder (Step B6) and extremely fine powder (Step B7) which can be made by handwork. **a** and **b** in cm.

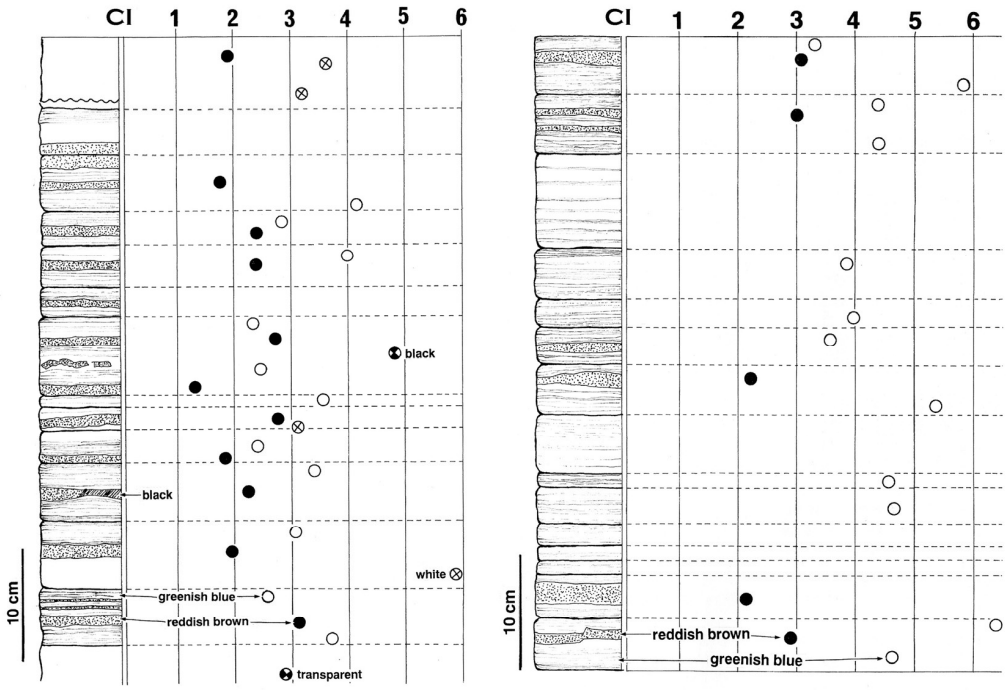

Fig. 2. Crystallinity indices of quartz in Triassic bedded cherts at the northwest of Mt. Fujikura (left) and at the Fujikura-dani Track (right) in the Nanjo Massif, Fukui Prefecture, Central Japan. The crystallinity indices of red chert (black circle) is clearly lower than those of green chert (open circle). The distance between both sites are 10 km. Stippled layers and layers shown with horizontal thin lines are red cherts and green cherts, respectively. Reproduced from Umeda (1999).

Table 5. Ranges and mean of **CI** and the standard deviation (SD) of red, black and gray, green, and white and transparent cherts in the Nanjo Massif. The whole average is 3.56. **F**=1.36 ～ 1.4 .

Color of chert	N	Range of CI	Mean of CI	SD
red	28	1.29～4.84	2.89	0.96
black and gray	8	1.81～4.80	3.38	0.98
green	53	2.12～6.09	3.67	0.86
white and transparent	11	2.88～7.71	4.68	1.97

Presumably, because repetition of grinding increases amorphous components in the powder, peak heights of **a** and **b** decrease (Smith, 1997). However, the ratios are relatively unchanged.

3.2 Measurement of Chert

Seven chert samples collected from the Mesozoic accretionary terrane in Central Japan called the Mino-Tamba Terrane (Hattori, 1982; Mizutani and Hattori, 1983) were XRD-analyzed. Chert 1 is a Triassic red chert from Unuma Area, Gifu Prefecture. Cherts 2 to 7 are Triassic cherts from the Nanjo Massif, Fukui Prefecture: Cherts 2 and 3 are white cherts, Cherts 4 and 5 are red cherts, Chert 6 is a green chert, and Chert 7 is a black chert. Chert 2 is present close to a dolomite layer. Cherts 3 to 7 were collected at a small area far from the dolomite. 1 to 2 mm thick plates were cut out of chert chips and the cut surfaces were XRD-analyzed (Step B1). Thin sections without glass covers polished by #800 polishing powder and 1/4 μm diamond paste were XRD-analyzed (Steps B2 and B3). Part of chert samples were crushed and ground, then XRD-analyzed (Step B4). Further grinding and analysis (Steps B5, B6, and B7) were done. It took at least five days from Step B1 measurement to Step B6 measurement.

Table 4 indicates that peak heights, **a** and **b**, vary clearly measurement by measurement. However, ratios, **a**/**b**, are not identical but relatively stable as observed in the cases of rock crystals. The variation of peak heights may be a result from difference in packing firmness

of mounted powders and difference in irradiation spot (domain) in cut surfaces and polished thin sections. Preferred orientation of quartz powder, if present, seems to have given another influence to the peak heights. Formation of amorphous components due to repetition of grinding could be another reason causing the peak variations. However, stability of **a**/**b** means that the ratio hardly depends on sample states (powder, cut surface, or polished surface), grain size of the powders prepared through grinding, and time after grinding. (XRD charts for samples of polished thin sections showed existence of small amount of amorphous materials. Chert sheets on the thin sections are very thin and presumably part of X-ray could penetrate to the glass base.)

In order to see whether **a**/**b** changes in a wet condition, a drop of water was dripped on the powder mounted on the holder of the final measurement of each sample. The powder was dried in air, and then XRD measurement was done. The measurement indicated that ratio, **a**/**b**, of every samples is not different from **a**/**b** of Step B6 and B7 of them. In the powder formed by hand grinding, damages of crystal structure, even if present, are not annealed even in wet condition.

The series of XRD analyses indicates that the most influential factor on CI is what kind of standard materials are selected. To avoid this disadvantage, ratio, **a**/**b**, is the best to describe quartz, not CI defined by Murata and Norman (1976). The error level may be 15% as a whole, though. Because two of three variables, CI, **F**, and **a**/**b**, are independent, one is dependant on the rest. Thus, **CI** should be described together with **F**. As CI's have been used traditionally, in the following description, they are presented with **F**, if possible.

4 Quartz Crystallinity in Chert of Central Japan

4.1. Relation between Crystallinity Index and Color of Chert

There are abundant chert layers in the Mesozoic Nanjo Massif, Central Japan (Hattori and Yoshimura, 1979). The depositional (radiolarian) ages of the cherts are mainly Triassic (Hattori and Yoshimura, 1982). They are white, black, gray, green, and red in color (Hattori, 1993: Hattori et al., 1993). In this Massif, many samples of chert were collected and CI's were determined. A clear relation between CI and color of chert is observed (Fig. 2). As shown in Fig. 2 and Table 5, although the distributions of CI are overlapped considerably, the mean CI of red chert is the lowest, the next is of black and gray chert, then of green chert, and the highest mean CI is of white and transparent chert. The average CI of chert in Table 5 is 3.56.

4.2. Relation between Crystallinity Index and Distance from Volcanic Dykes

Locally in the Nanjo Massif, many igneous dykes (rhyolite and andesite) intruded into the Mesozoic sedimentary rocks. Many igneous dykes are 50 cm to 2 m wide in outcrop. Within 2 m around the dykes, we collected many chert samples, some are partially argillaceous. Their CI's and the distances from the dykes are graphically illustrated in Fig. 3. CI is highest at places close to the igneous contact and gradually lowered with the distance. The correlation coefficient between CI of cherts at the distance of less than 60cm and distance from dykes is 0.55. This observation implies that CI increases toward igneous dykes and suggests dependence of CI on the thermal history of host cherts.

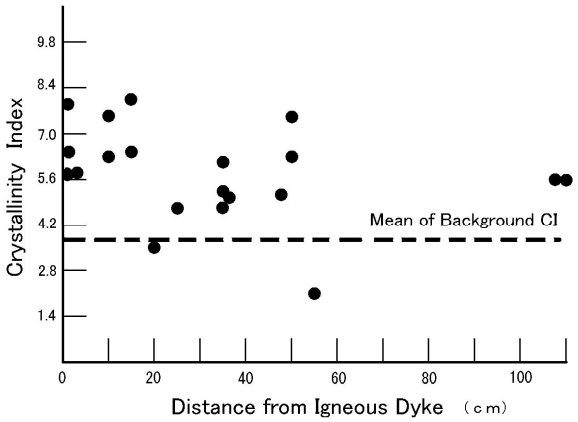

Fig. 3. Relationship of quartz crystallinity (CI) of chert around igneous dykes. Clear relationship between CI and distance from the dyke is visible. Mean CI (=3.56) of other cherts of the Nanjo Massif listed in Table 5 can be used as background CI. **F**=1.36～1.4.

4.3. Crystallinity Index in the Tamba area

The Tamba Area belongs to the same terrane to the Nanjo Massif called the Mino-Tamba Terrane. The Tamba Area and the Nanjo Massif were formed simultaneously by plate convergence at the late Mesozoic (Hattori, 1982: Mizutani and Hattori, 1983). There are many cherts in the Tamba Area and we determined CI of the cherts in a portion of the Tamba Area. In our results (#39 in Table 1, simply #39 hereafter), the mean CI of 16 black and gray chert samples is 8.18 (**F**=1.46). The CI's are clearly high compared to those of chert in the Nanjo

Massif (#35 and #36) (Fig. 4).

4.4. Crystallinity Index and Depositional Environment of Chert

There are some Miocene cherts at a few places 30 to 40 km north of the Nanjo Massif (Hattori et al., 1996: Umeda, 2003). One of them was described by Umeda (2003). The chert, called the Yachi chert, was deposited in a Miocene ephemeral lagoon occurring at the Japan-side margin of the Sea of Japan. According to Umeda (2003), quartz was deposited directly from solutions in the lagoon. Mean CI of 11 chert samples from Yachi is 6.13 (**F**=1.25) (#40). This is clearly higher than that of the cherts of the Nanjo Massif. The cherts of the Nanjo Massif is considered to have been deposited on a Mesozoic deep-sea floor as radiolarian ooze. On the other hand, the Yachi chert is considered to have accumulated as quartz layers in a Miocene lacustrine to near-shore environment. It may be likely to say that quartz in cherts deposited in lacustrine environments has higher crystallinity than in the deep-sea radiolarian cherts. The difference in origin may result in the difference in quartz crystallinity.

In the Nanjo Massif, epigenetic chert nodules in a sandstone succession are developed (Hattori, 1999a). The chert is composed of quartz and minor amount of kaolin minerals. Some quartz crystals are megaquartz, some microquartz, some are contacted to kaolin minerals, some not contacted to kaolin minerals. CI's of the cherts determined separately spread between 5.01 and 7.93 (**F**= 1.33) (#34). The crystallinity of the epigenetic chert is also clearly higher than the bedded cherts in the Massif, implying difference in environment of quartz precipitation.

4.5. Crystallinity Index and Provenance Analysis

Many conglomerate beds containing chert cobbles and pebbles are distributed in the areas west and north of the Nanjo Massif. The depositional ages are Cretaceous, Oligocene, Miocene, and Pleistocene. CI's were determined for the chert cobbles in these conglomerates (Umeda, 2007). CI's of chert cobbles collected from the early

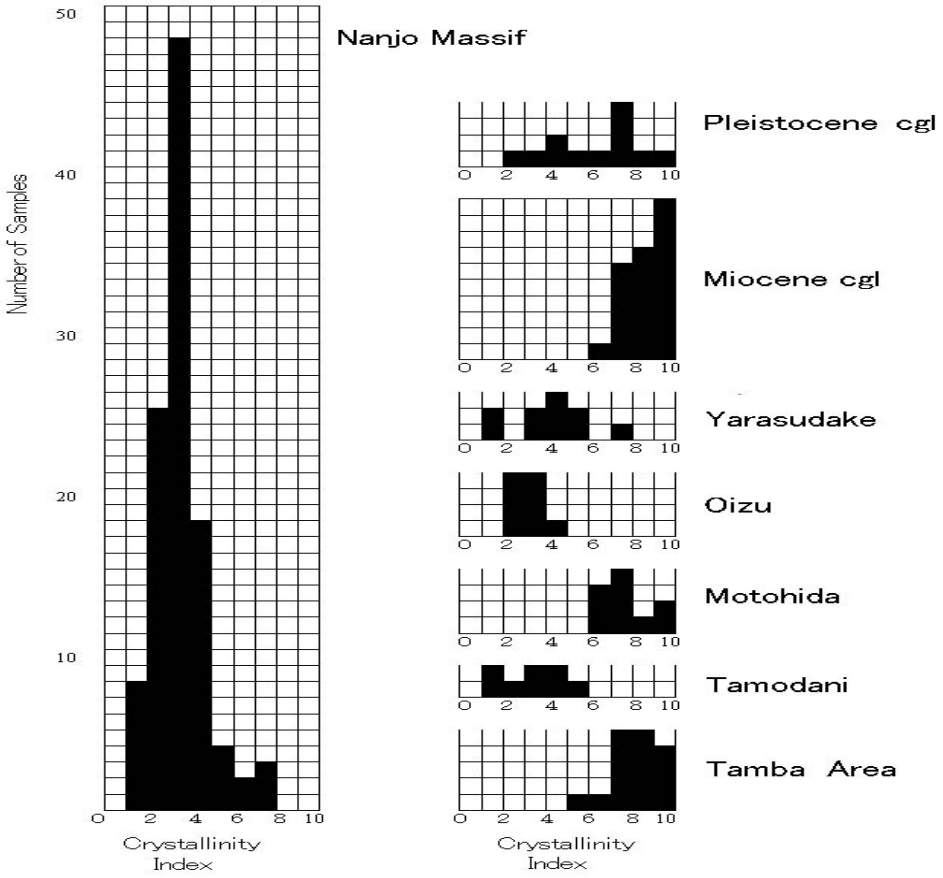

Fig. 4. Crystallinty indices determined for quartz in bedded cherts of the Nanjo Massif and the Tamba Area, and in chert clasts of Pleistocene terrace conglomerate in the southern part of the Niu Area, Miocene conglomerate in the Niu Area, Oligocene Yarasudake conglomerate, late Cretaceous Oizu conglomerate, late Cretaceous Motohida conglomerate, early Cretaceous Tamodani conglomerate. The sample number of the cherts in the Nanjo Massif of this figure is 108. 8 samples were added to the samples shown in Table 5. CI's larger than 10 were included to CI of 10. For brief description of these bedded cherts and conglomerates, see Umeda (2007)

Cretaceous Tamodani conglomerate, western Fukui (Saida, 1987) range between 1.68 and 5.71, and the mean is 3.60 (N=8, F=1.46). CI's of chert cobbles of the Late Cretaceous conglomerate (Oizu Conglomerate: informal name) just north of the Nanjo Massif are distributed between 2.05 and 4.94, and the mean is 3.10 (N=9, **F**=1.35). Chert cobbles of another Late Cretaceous conglomerate (Motohida conglomerate, Kido, 2001) at the west of the Nanjo Massif which were metamorphosed in the Early Miocene have CI's between 6.06 and 9.73, and the mean the is 7.65 (N=10, **F**=1.35). CI's of the Oligocene conglomerate (Yarasudake conglomerate, Umeda, 1997) overlying the Nanjo Massif unconformably spread in a range between 1.59 and 7.05, and the mean is 4.10 (N=10, **F**=1.48). CI's of chert cobbles of the Miocene conglomerate which were deposited near the Niu chert (#33) are distributed between 6.42 and 10.33, and the mean is 8.58 (N=24, **F**=1.42). The distribution ranges of CI's of chert cobbles in the Yarasudake and Oizu conglomerates are similar to that of chert in the Nanjo Massif (N=100, F=1.36～1.4, mean CI=3.56, Table 5). On the other hand, that of the Miocene conglomerate resembles to that of the Tamba Area (#39, N=16, F=1.46, CI=5.9～9.7, the mean is 8.18, Table 1). This evidence may imply that chert cobbles of the former two conglomerates (Oizu and Yarasudake) were derived from the Nanjo Massif and the chert cobbles of the Miocene conglomerate came from a different area in which cherts with high CI have been developed.

The Pleistocene conglomerate is present as terrace conglomerate along current rivers in the Niu Area. The conglomerate contains abundant chert cobbles, and CI's of them are distributed in a wide range from 2.65 to 10.36, and the mean is 6.18 (N=12, **F**=1.64). This wide range of CI may imply more than one provenance of the chert cobbles. The cobbles of low CI are derived probably from the Nanjo Massif, just south of the terrace. On the other hand, the chert cobbles of high CI are derived from a missing geological formation.

5 Discussion

1. CI and **F** are determined by XRD measurements of rock crystals. However, it varies crystal by crystal. For example, Moxon et al. (2007) showed that the CI's of a colorless Brazilian rock crystal, rose quartz, and amethyst are 10, 10.1, and 9.6, respectively. The CI's of the rock crystals described in this paper change between 10.0 and 8.5 (Table 1). This difference corresponds to the difference in the scaling factor by 1.18 (=10.0/8.5). Accordingly to rock crystals used as a standard, quartz with CI higher than 10 could exist.

Table 1 lists CI and **F** from various cherts and siliceous rocks in different areas. As recognized in this table, **F** distributes between 1.25 and 1.99. This difference results in about 60 % error and is too large to compare CI's reported in different studies. CI is not stable even for identical crystals measurement by measurement. Ui and Mizukami (1994) made it clear that CI's obtained from 23 measurements of **F** for an identical crystal varied between 10.0 and 9.1. These results indicate that **F** defined by Murata and Norman (1976) includes at least this range of error. Presentation of quartz crystallinity with ratio, **a**/**b**, has an advantage in avoidance of this difficulty. However, traditional CI is widely used and many data have been reported from many different studies in the form of CI. To reconcile this dilemma, it is recommended to present traditional CI together with **F**.

According to Murata and Norman (1976) and Moxon et al. (2007), different XRD in different laboratories may give different CI's. One reason why Murata and Norman (1976) introduced the scaling factor was to invalidate difference of XRD models used in different studies. It seems that every XRD gives a not so different reflection pattern of quartz and that almost all geologists never determine CI with different XRD's. Thus, the influence produced by the difference can not be accurately answered. If the difference gives significant difference in resultant CI, the CI includes two types of error, one is from difference of XRD models, the other from difference of standard crystals used.

Mikami et al. (2002) determined CI's of many cherts distributed in the Tamba Area (#38). The cherts were collected in different outcrops from ones of our study (#39), and their instrument is different from our model. The results are very similar each other, and we think that the error from the difference in XRD model is negligible. It is undoubted, however, that accurate comparison among CI should be done for data offered from same instrument models.

2. Dependence of CI on color of host chert layers is shown in Fig. 2 and Table 5. According to Murata and Norman (1976), iron-rich cherts such as hematitic chert tend to give too high CI. CI's of the red cherts in Fig. 2

are invariably lower than those of the adjacent green cherts, indicating that almost no effect is given from iron contained in the red cherts. The individual chert beds of red/green color analyzed in this paper are characterized with central red stripes and top and bottom green stripes. It is considered that circulating water along bedding planes reacted to marginal portions of red chert beds and formed the green stripes. This reaction is considered to have enhanced crystallinity of quartz and resulted in difference in CI between red cherts and green cherts. According to Thurston (1972), preservation of radiolarian fossils is better in red chert than in green chert in Radiolaria-bearing bedded chert of western Italy. He noticed that grain size of quartz is larger in the green chert than in the red chert, and considered that water-quartz reaction in chert destroyed fossil radiolarians and increased the grain size of quartz in the chert. Hein et al. (1981) insisted that quartz crystallinity has a relation to duration of connection between water and chert : longer reaction, higher crystallinity. The same phenomenon is recognized in the bedded chert in the Nanjo Massif (Umeda, 1999), and change in color of chert from red to green is regarded to have enhanced the crystallinity.

CI's in older silicified woods are higher than those in younger ones (Stein, 1982) (#25). Moxon et al. (2006) analyzed crystallinity of chalcedony in agates of different ages (#41), and concluded that crystallinity of chalcedony increases rapidly to Cretaceous agates. These observations indicate that older quartz has higher crystallinity than young quartz, and long-time connection between quartz and water produces quartz of higher crystallinity. It seems that CI is dependant not only on color of chert but also on duration after the deposition of chert.

3. The Tamba Area of central Japan is the western extension of the Mino Terrane including the Nanjo Massif. The origin and geology are very similar to the Nanjo Massif. The only difference is the metamorphic grade. The Tamba Area was intruded by some late Mesozoic granitic plutons and is considered that the granite underlies the terrrane. The metamorphic grade of the greenstones in this area is characterized with pumpellyite facies to greenschist facies (Hashimoto and Saito, 1970). On the contrary, the Nanjo Massif has no visible thermal source. The metamorphic grade of the greenstones in the Nanjo Massif is from zeolite facies to prehnite-pumpellyite facies (Hattori, 1978). CI's of bedded chert in the Tamba Area range from 3.6 to 9.3 (**F**=1.31) (#38) (Mikami et al., 2002), and from 5.9 to 9.7 (**F**=1.46) (#39). Those in the Nanjo Massif (#35 and 36) range from 1.3 to 6.4 (**F**=1.4) (Umeda, 1999). One of the conclusions led by Mikami et al. (2002) in the Tamba Area is that CI seems to depend on the thermal history which the host cherts underwent, such as diagenesis and metamorphism. Their result is very similar to our result (#39), and high CI's of the cherts of the Tamba Area imply that the Tamba area underwent thermal events of higher temperature than those of the Nanjo Massif.

Metamorphic effect, in other words, thermal effect is also important to change CI of quartz crystallinity. As shown in Fig. 3, thermal effect on CI is apparent within some tens centimeters around volcanic dykes intruded into bedded cherts in the Nanjo Massif. Ui and Mizukami (1994) analyzed systematically the change in CI in Permian bedded cherts of the Mino Terrane of Central Japan which underwent contact metamorphism by a granitic intrusion (#32). The metamorphic grade at the contact is characterized with the green hornblende zone which is higher than the cordierite-orthoclase isograd and the metamorphic temperature was estimated to have been about 650°C (Suzuki, 1975). The metamorphic grade declines from the granitic contact to the distance, and CI's decrease from 10 to about 5.0 at the distance of 6.5 km from the granite contact. Because the background level of CI's in this area is about 4.0 (Ui and Mizukami, 1994), CI's increase clearly toward the granite contact.

Lee et al. (1994) studied the change in CI in Eocene and pre-Tertiary metapelite in a greenschist facies area of regionally metamorphosed district in Taiwan (#42). CI and grain-size of quartz increase along with progressive metamorphic increase in common. These observation suggests that CI can be used as important information with respect to analysis of progressive metamorphism.

As found in the above discussion, determination of CI offers a rapid method to trace qualitatively paleothermal history of host rocks (Moxon et al. 2006), and Moxon et al. (2007) demonstrated that CI can be used to trace post-depositional thermal processes within the host rock.

4. Origin of chert is variable: some are deposited as siliceous oozes in the deep-sea and transformed later to bedded cherts diagenetically (Mizutani, 1970: Williams et al., 1985), some deposited directly as quartz cherts on land, and some epigenetic in origin. In Table 1, CI's in various chert layers are represented. The bedded cherts of

the Nanjo Massif are Triassic cherts deposited as deep-sea siliceous ooze. Hein et al. (1983) measured CI's in Miocene chalk near the Costa Rica Ridge and reported that they range between <1.0 and 2.1 (#26). CI's in the Triassic red and green cherts of the Nanjo Massif are 1.3 to 6.4 (**F**=1.4) (#35, 36). The Miocene Niu chert (Hattori et al., 1996) and Yachi chert (Umeda, 2003) were deposited in small lagoonal to lacustrine environments. The host rocks are altered to zeolitic facies (Hattori, 1999b). CI's from these cherts are 2.44 to 4.35 and 5.1 to 7.0 (**F**=1.4), respectively (#33 and 40). The silica is considered to have been deposited as chalcedony and quartz (Hattori et al., 1996: Umeda, 2003). The epigenetic chert was deposited in a late Mesozoic coarse-grained sandstone sequence of the Nanjo Massif. The silica is considered to have been deposited as quartz (Hattori, 1999a). CI's in the epigenetic cherts are 5.0 to 7.9 (**F**=1.33) (#34).

5. Provenance analysis using chert cobbles has been often done. For example, Seiders et al. (1979) and Seiders (1983, 1988) considered that because some chert cobbles in the Great Valley Sequence contains radiolarian fossils resembling to those found in the cherts of the Franciscan Complex, the chert cobbles presumably were supplied to the former from the latter. Dutta (1998) analyzed oxygen isotope characteristics of chert cobbles in the Permian to Triassic sandstones of the Sydney Basin and made it clear that they were not derived from the New England Orogen.

As noticed in these examples, because chert is very hard and durable through weathering and transportation in sedimentary processes, chert is a good material in provenance analysis. Key characteristics to have been paid attention to chert cobbles are generally fossils and chemistry including isotope analysis, and should include CI analysis too. CI can be regarded to be fossils of thermal experiences of chert. By using effectively CI's in chert cobbles, reliability of provenance analysis about sedimentary basins can be enhanced, as described in this paper.

6. We can not explain why CI's of chert are not uniform. As shown in Table 1, they distribute from <1 to 10. The variation seems to be significant, even if operation errors are involved in the variation. Some factors are considered to be responsible for the variation: accumulation process of quartz, impurity in quartz, crystal morphology and size of quartz and so on. Because, in the XRD method, sample quartz crystals are crushed and ground, the crystal morphology and size seem to give no contribution to CI determined for ground quartz. If quartz crystals have internal defects and lattice distortions, they may cause to decrease CI's. Determination of CI involves many unexplained factors such as variation of **F**, contribution of difference in firmness and grain-size of packed powder on the sample plates (holder), effects caused from difference of measurement conditions and difference of instruments used. These unknown factors may cause errors of 1 to 2 in CI. By treating CI's statistically, we can use them in the geological application, if we take this range of error in account.

6 Conclusion

In use of CI to compare and correlate the origin and diagenesis of cherts, there have been some uncertainties. One is that **F** necessary to calculate CI defined by Murata and Norman (1976) depends on rock crystals used as standard materials. The second is the effect of grain size of sample powder on the XRD resolution. Our results show that the first uncertainty can be relieved by presenting not only CI but also **F**. The second uncertainty cannot be deleted but our result showed that ratios of peak heights, **a** and **b**, in XRD charts are relatively stable and fluctuate within 10% variation, if we cut, crush, and grind samples by handwork. Although the effect of packing firmness of sample powder still remains uncertain, CI together with F seems reliable if we accept 10 to 15% error.

It is likely to say that description of crystallinity of quartz with CI has advantage in easy communication because the maximum CI is 10. When CI is still used for description of quartz, it had better to be described together with **F**.

Crystallinity of quartz in cherts seems to depend on origin, and post-depositional diagenesis and thermal history which the cherts underwent. Based on this viewpoint, CI was determined for many types of chert: some are bedded cherts, and some are chert cobbles in conglomerate of central Japan. CI increases from red color chert to green and to white, black chert, toward thermal sources intruded into host chert, along with increase in meta-morphic grade, with reaction time of host cherts and water, and probably with age. CI of chert of deep-sea origin differs from that of chert deposited on land. Thus,

it was confirmed that CI has a relation to origin, diagenetic and post depositional thermal history of sedimentary quartz, and this result seems to make it clear that CI and **F** can be used as a simple and effective index describing characteristics of sedimentary quartz.

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中部日本のチャート中の石英の結晶度：その信頼性および地質学的応用

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（要旨）　X線解析により決められる石英の結晶度は世界各地から報告されている．報告された結晶度は１から10に拡がる．この違いは結晶度を決める際の試料の調合状態によるのか，あるいは真に石英に結晶度の差があることによるのか不明であった．我々は，試料をいろいろな状態にし，水晶やチャート中の石英の結晶度を測定し，結論的には，結晶度はその測定方法や試料の状態に依存はしないが，標準試料としてどの水晶を用いたかに依存することを明らかにした．このような不都合を取り除いても石英の結晶度には大きなばらつきがあり，それは石英の起源や石英が被った熱史により異なることに依存し，一様ではないことを意味する．

我々のデータと文献に現れたデータは以下のことを示す．

１）新生代の深海軟泥中やそのチャート中の石英結晶度は陸上の中生代層状チャートの結晶度より小さい．

２）熱を被った層状チャートは高い石英結晶度をもつ．

３）湖沼性あるいは河川域性のチャートの石英結晶度は深海起源のチャート中の石英の結晶度より高い．

４）年代の古いチャートの石英結晶度は同じ起源の若いチャートより高くなっている．

この解析は石英結晶度は堆積性珪質岩の起源，その続成過程，そして変成度を解き明かす有用な指標となることを示す．

キーワード：結晶度，石英，チャート，続成過程，供給地，熱史，X線解析

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