

# Stability and Dissolution of Heavy Minerals in the Neogene-Pleistocene Sandstones from Western Foothills, Taiwan

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**Abstract.** Stability and dissolution of heavy minerals were studied in the sandstones from the Western Foothills, Taiwan, where the continuous sequence from Miocene to Late Pleistocene develops. On the basis of modal proportions of heavy minerals in sandstones, it is concluded that many heavy minerals have been dissolved even in Late Pliocene sandstones. Stable minerals throughout the sequence are zircon, tourmaline, rutile, monazite, apatite and spinel. Ilmenite was dissolved in the Early Miocene sandstone. Dissolutions of aluminous silicates such as chloritoid, staurolite and  $Al_2SiO_5$  polymorphs are observed in the Middle Miocene sandstones. Epidote, titanite and grossular-andradite garnet are more unstable minerals. Chemical analyses of detrital almandine garnets revealed that garnet with grossular component more than 10% was selectively dissolved in the Early Miocene sandstone. It is important for provenance study at first to recognize the selective dissolution of heavy minerals in the sandstones.

**Key words:** Taiwan, Western Foothills, sandstone, heavy mineral, dissolution.

## Introduction

One of the approaches to provenance studies of sandstones is based on determination of relative abundance of the detrital mineral. Although such a method provides a strong basis for the interpretation of provenance, we must take into serious consideration of any possibility of the post-depositional dissolution of detrital minerals in sandstones. Besides, chemical compositions of heavy minerals in the sandstones have given more detailed information for their provenance (Morton, 1991; Yokoyama *et al.*, 1990). Recently, age analyses of detrital monazite and zircon have been used to decipher their provenances, since both minerals are usually treated as stable minerals and they will not be dissolved after deposition. As discussed in many papers (e.g., Pettijohn, 1941; Morton, 1984; McBride, 1985), dissolu-

tions of detrital minerals in sandstone are more or less existed and only relic detrital minerals are preserved in the sandstones. Discrimination of detrital minerals either stable or having been dissolved must be at first considered for provenance interpretation. Among heavy minerals, zircon and tourmaline are treated as ultrastable minerals. Apatite, rutile, spinel, garnet, monazite, and xenotime are treated as relatively stable detrital minerals (Morton, 1984; Pettijohn, 1941). In this paper, we analyzed modal proportion of all the minerals and chemical composition of detrital garnet in the sandstones of Early Miocene to Pleistocene stratigraphic sequence of the Western Foothills from the central Taiwan in order to obtain the stability order of detrital minerals.

## Geological outline of Taiwan

Taiwan can be broadly divided into three

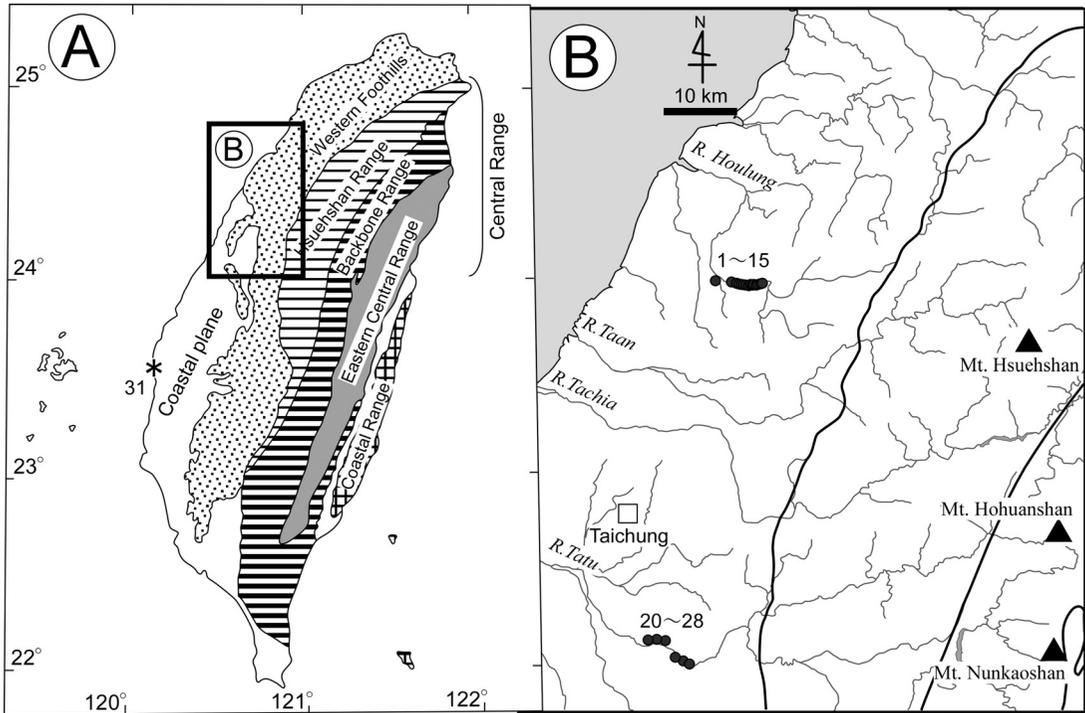


Fig. 1. Maps of Taiwan. A, Geological provinces of Taiwan (\* locality for fresh sand sample); B, sampling localities (No. 1–15 from the Houlung River and No. 20–28 from the Tatu River) for the Tertiary-Quaternary sandstones.

major geologic provinces which form long narrow belts and are, from west to east, Western Foothills including Coastal plain, Central Range, and Coastal Range (Ho, 1988; Fig. 1). Western Foothills is composed of late Oligocene to late Pleistocene clastic rocks which are deposited mainly in shallow marine environment and occasionally intercalated with coal seams, limestones, and tuff lenses.

The Central Range forming the backbone ridge system of Taiwan is subdivided on the basis of lithology, age, and metamorphic grade into, from west to east, Hsuehshan Range, Backbone Range, and Eastern Central Range. The Hsuehshan Range is a weakly metamorphosed belt of mainly shallow water Eocene to middle Miocene clastic rocks that have been juxtaposed against the Western Foothills by a major eastward dipping fault. The Backbone Range is composed mainly of weakly metamorphosed clastic rocks, and is underlain by the Eastern Central Range pre-Tertiary

metamorphic complex. The Coastal Range of eastern Taiwan is composed of rocks deposited in a Neogene volcano-sedimentary basin associated with a volcanic arc on the western leading edge of the Philippine Sea plate.

Chou (1980) inferred that the sources of the Tertiary sediments lay northwest and west of Taiwan with sediments being transported southward and eastward down a gentle paleoslope. In contrast, the Late Pleistocene sediments of the Toukoushan Formation were derived from the Central Range in Taiwan (Chou, 1977). The change in provenance area has been recognized in the Pliocene sequence of the Chinshui Shale (Chou, 1974). Yokoyama *et al.* (in preparation) analyzed monazite age in the sandstones from the Western Foothills and Hsuehshan Range. They concluded that the detrital minerals, except for the Pliocene to Late Pleistocene sandstone derived from the Central Range of the Taiwan, were derived from various terranes in the Asian continent ranging

from the Korean Peninsula to Guangdong Province in China.

## Materials and Methods

### Sampling localities

More than 20 sandstones were collected from the Western Foothills in the central part of Taiwan (Fig. 1B). They were collected along the two routes studied by Chen *et al.* (1992). The depositional ages of these sandstones range from Early Miocene to Late Pleistocene (Table 1). No metamorphic texture has been observed in each sample. Fresh sand sample was collected from the western coastal beach of Taiwan (Fig. 1A). As the sample contains various detrital minerals without any dissolution, the mineral species was compared with those in the sandstones collected from the Early Miocene to Late Pleistocene sequence.

### Analytical procedures

Procedures for the separation of heavy minerals and their subsequent identification are the same as those described by Yokoyama *et al.* (1990). Carbonate, micaceous minerals, and authigenic pyrite were not subjected to examination, and magnetic fractions were removed prior to the separation of the heavy minerals. All the minerals were identified through the X-ray profile of the Energy Dispersive Spectrometer. Hence, minerals with the same chemical compositions were described as polymorphs, i.e. TiO<sub>2</sub> polymorphs for rutile, anatase, and brookite, and Al<sub>2</sub>SiO<sub>5</sub> polymorphs for sillimanite, kyanite, and andalusite. Modal proportions of representative heavy and light minerals are listed in Table 2. Light minerals were also suffered from dissolution, but they are not discussed here because of less source-diagnostic in the provenance study.

## Results

### Heavy mineral species

About twenty heavy mineral species were observed in the Western Foothills sandstones. In most of the samples, more than 200 grains were

Table 1. Neogene-Pleistocene stratigraphic sequence in central Taiwan (after Ho, 1988).

Sampling route (Ma) Stratigraphy	Tatu River route	Houlung River route
Pleistocene	Toukoshan F. ●●●	
Early (1.6)	Cholan F. ●●●	Cholan F. ●
Pliocene (5.2)		Chinshui Shale
		Kueichulin F. ●●
		Shangfuchi S.S. ●●●
		Tungkeng F. ●
Late		Kuanyinshan F. ●
		Talu F. ●
		Peilliao F. ●●
Middle		Chuhuangkeng F. ●●●
Early		
(23)		

identified and the abundance of each mineral is listed in Table 2. In some samples where pyrite is abundant, only restricted grains are observed in the heavy fraction. One sample, Tai-13, is rich in fossil and includes abundant apatite. The apatite was probably condensed as a part of the fossils. While it is described that the minerals listed here are mostly detrital, TiO<sub>2</sub> polymorphs and florencite are difficult to recognize whether they are detrital or secondary phases after deposition. They are listed tentatively in the separated column.

There is a variety of heavy mineral species in the younger rocks or sediments, whereas a restricted number of species were observed from the older rocks due to the common dissolution of some detrital minerals (e.g., Pettijohn, 1941; Morton, 1984, 1991). In both the Tatu River and Houlung River routes, the heavy mineral species decrease apparently with the increasing of depositional age (Fig. 2). It is expected that the Present sand sample from the western coastal beach should contain all the mineral species found in

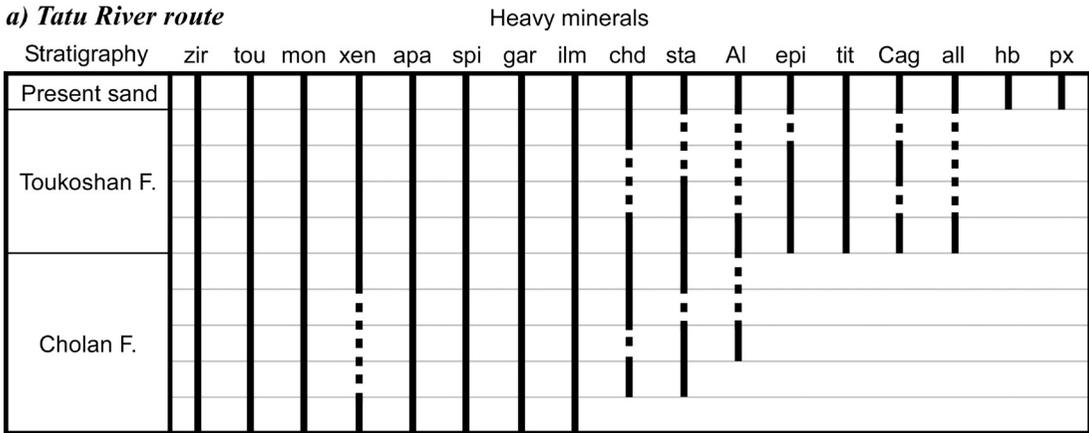
Table 2. Heavy and light minerals in the sandstones collected from the two routes in central Taiwan. Each numerical value shows a number of grain identified under EDS profile. Some minerals in the beach sand were not observed in the identification, but they were assumed to be present and are noted as P in the list. Minerals in D/A column cannot be discriminated into detrital or authigenic. Mineral abbreviations: zir, zircon; tou, tourmaline; mon, monazite; xen, xenotime; apa, apatite; spi, spinel; gar, garnet; ilm, ilmenite; chd, chloritoid; sta, staurolite; Al, Al<sub>2</sub>O<sub>3</sub> polymorphs; epi, epidote; tit, titanite; Cag, grossular or andradite; all, allanite; hb, hornblende; opx, orthopyroxene; cpx, clinopyroxene; Ti, TiO<sub>2</sub> polymorphs; flo, florencite; Qz, quartz; Ab, albite; Pl, Ca-bearing plagioclase; Kf, potash feldspar.

Tatu River route		Heavy minerals													D/A		Light minerals							
Stratigraphy	Sample	Heavy minerals													Ti	flo	Qz	Ab	Pl	Kf				
		zir	tou	mon	xen	apa	spi	gar	ilm	chd	sta	Al	epi	tit							Cag	all	hb	opx
Beach sand	Tai-31	60	2	32	P	9	14	7	50	P	P	P	4	3	P	P	4	13	55	1	82	12	1	5
Toukoshan F.	Tai-28	76	6	5	1	2	8	9	22	1					1				81	2	95	2		4
Toukoshan F.	Tai-27	74	10	10	2	4	7	14	22				2	2	1				57	2	93	1		6
Toukoshan F.	Tai-26	11	11	9	1	8	4	9	13	3			2	3					81	2	96	2		2
Toukoshan F.	Tai-25	53	14	1	1	12	9	40	28	1	4	1	12	5	1	1			104	3	92	2		6
Cholan F.	Tai-24	34	4	2	1	6	3	15	21	2	1								44	1	90	5		7
Cholan F.	Tai-23	80	4	3		24	5	15	18	1									55	2	80	11		15
Cholan F.	Tai-22	71	9	1	1	11	1	25	26	1	1								67	2	85	7		7
Cholan F.	Tai-21	82	6	8		6	3	25	19	2	1								70	1	84	4		12
Cholan F.	Tai-20	110	5	7	1	2	2	21	12										69		89	2		9

Houlung River route		Heavy minerals													D/A		Light minerals							
Stratigraphy	Sample	Heavy minerals													Ti	flo	Qz	Ab	Pl	Kf				
		zir	tou	mon	xen	apa	spi	gar	ilm	chd	sta	Al	epi	tit							Cag	all	hb	opx
Cholan F.	Tai-15	36	18	2				2	8	65	3	5	3	18	2	1	6		56		95			5
Kuechulin F.	Tai-14	33	16	2				5	3	43	7		13	2	3	1			94	1	90	2		9
Kuechulin F.	Tai-13	3				80		4	8				2	1					3		84	6	2	8
Shangfuchi SS	Tai-12	82	7	3	1	6	1	9	48										40		79	7		14
Shangfuchi SS	Tai-11	16	3	2		5	2	24	43	1	3								44		87	2	1	12
Shangfuchi SS	Tai-10	58	5	5	3			3	110		3								31		98			2
Tungkeng F.	Tai-9	15	14	1				3	38	80	2	4	1						47	3	90	3		8
Kuanyinshan F.	Tai-8	29	8	3		17	2	21	9	1									50	1	77	11	1	11
Tatu F.	Tai-7	20	2	2		22		5	1	1									64		67	12	9	10
Peiliao F.	Tai-5	22	17			18	1	17	1										40		78	8		13
Chuhuangkeng F.	Tai-3	6	2			3	3	4	1										20		82	9	1	8
Chuhuangkeng F.	Tai-1	97	13	8	2	13	1	21											67	1	85	13		2
Chuhuangkeng F.	Tai-2	76	7	10	1	24	3	7											89	2	77	16	2	5
Chuhuangkeng F.	Tai-4	64	12	11		30	1	32											81	6	74	12	2	14

**a) Tatu River route**



**b) Houlung River route**

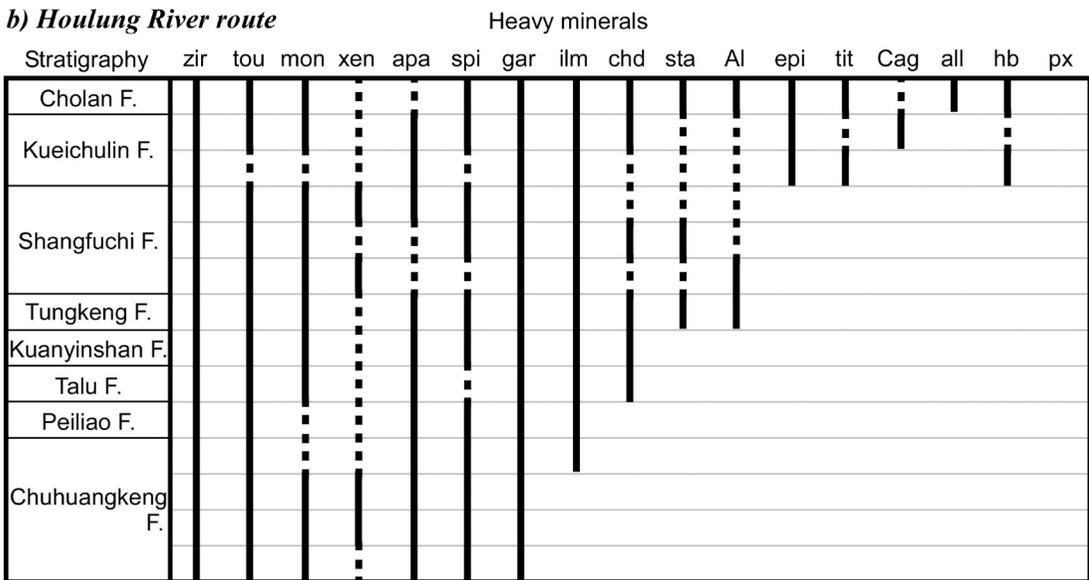


Fig. 2. Heavy mineral persistent charts in the Neogene-Pleistocene sandstones in the Tatu and Houlung rivers routes. Dotted line indicates that the mineral was not confirmed in the restricted mineral identification, but was assumed to be present in the samples. Mineral abbreviations are the same as those in Table 2.

the sandstones of older ages, but some minerals are too rare to be found under a restricted number of mineral identification. In more detailed analyses, some minerals that were absent in the Table 2 may be confirmed as rare minerals in both the Present sand and Neogene-Pleistocene sandstones.

Pyroxenes, present in the beach sand sample, are not observed in any sandstone of older ages. Hornblende is found only in the upper part of the Houlung River, but it is not observed

in the Tatu River samples. Epidote, titanite, grossular, andradite, and allanite are observed only in the young sequence of both the rivers. Aluminous minerals such as chloritoid, staurolite, and Al<sub>2</sub>O<sub>3</sub> polymorphs are found from middle to top of the sequence including Talu F. up to Cholan F. of the Houlung River route. Ilmenite is one of the major minerals in the heavy fractions. It is rare or absent in the Early Miocene sandstones.

TiO<sub>2</sub> polymorphs are abundant. Among them,

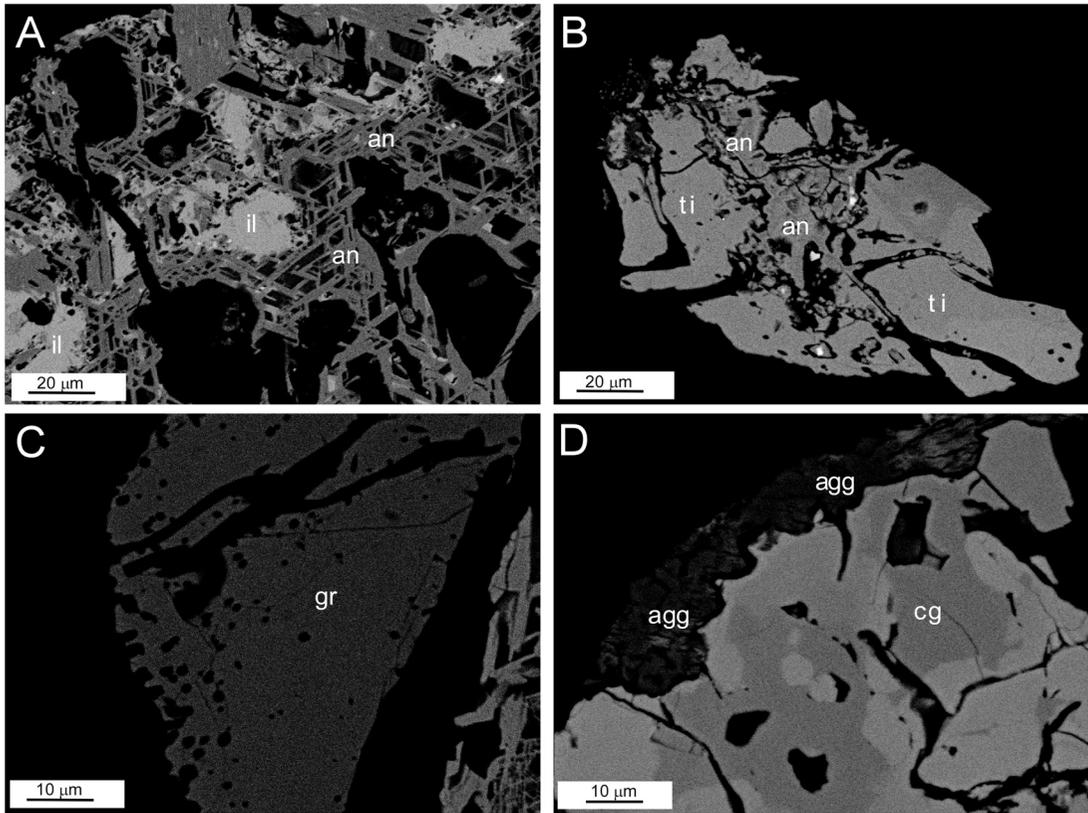


Fig. 3. Back-scattered images showing dissolution and decomposition of detrital minerals. A, Ilmenite (il) is decomposed into anatase(an)-bearing aggregate (sample Ta-8); B, titanite (ti) replaced by anatase (sample Ta-25); C, grossular (gr) with many wormholes (sample Ta-14); D, andradite (cg) with irregular rim (sample Ta-14), possibly dissolved into goethite-bearing aggregate (agg).

anatase is common, occurring as an aggregate with quartz occasionally replacing ilmenite. Rutile is optically identified in the heavy fractions from most of the sandstones. It occurs as a well-crystallized and rounded grain under electron microscope. Rutile is treated as an ultrastable mineral in many papers (e.g. Pettijohn, 1941; Morton, 1984). Florencite in the uppermost part of the sequence is clearly detrital, but, in its lower part, it is probably secondary mineral formed by the decomposition of allanite.

Dissolution or decomposition texture is observed in many samples from the lower sequence. Ilmenite is decomposed into  $\text{TiO}_2$  polymorphs, goethite, and quartz (Fig. 3A). Titanite is also replaced by anatase (Fig. 3B). Rounded grossular grain has numerous wormholes (Fig. 3C). Andra-

dite has irregular rim due to the replacement by goethite-bearing aggregate (Fig.3 D).  $\text{Al}_2\text{O}_3$ -rich minerals occur as skeletal grain surrounded by clay minerals. Amphibole in the sandstones has a saw-edged rim. These textures show that the minerals, absent in the lower sequence, were dissolved or decomposed after deposition.

#### Garnet composition

Garnet is mostly treated as a stable detrital mineral (e.g., Pettijohn, 1941) and its chemical compositions were used for provenance study (Morton, 1991; Yokoyama *et al.*, 1990). On the other hand, Smale and Morton (1987) inferred dissolution of Ca-rich garnet in the drilled core from the Taranaki site and Yokoyama (1998) insisted the selective dissolution of garnet with

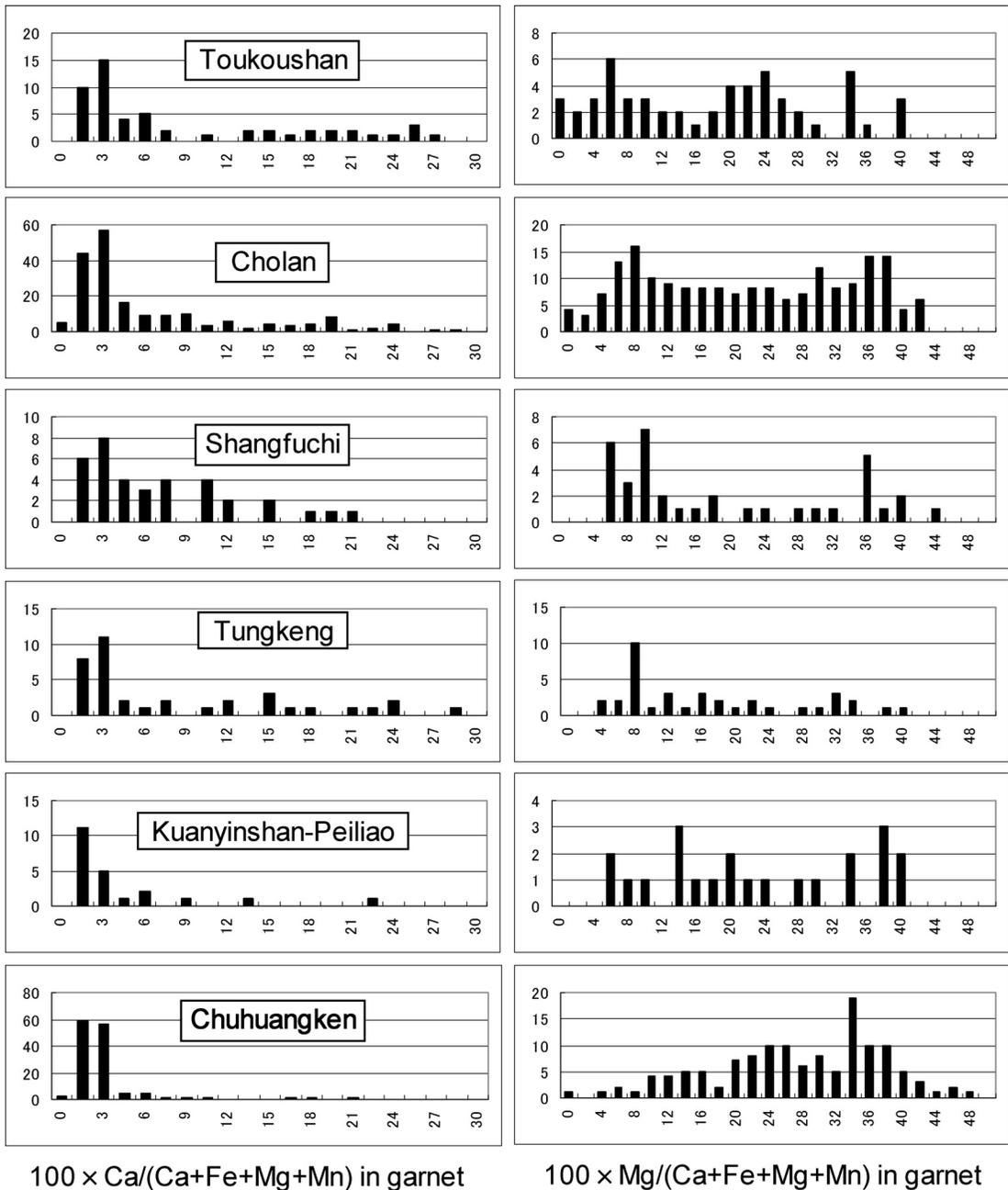


Fig. 4. Compositional variations of detrital garnets from the Late Pleistocene Toukoshan Formation to Early Miocene Chuhuangkeng Formation. Vertical axis is a number of analyses.

grossular component more than 10% in the pre-Jurassic sandstones. Chemical composition of garnet except for grossular and andradite was analyzed by an electron microprobe analyzer (EPMA, JEOL JX-1). The term of Ca-rich or Ca-

poor garnet is tentatively used here for garnets with grossular component more or less than 10%, respectively. As shown in Fig. 4, detrital garnets show a wide compositional variation. Pyrope component exceeds 40% in all the forma-

tions. As far as the range of pyrope component is concerned, no significant change is observed throughout the sequence. Grossular component is also variable, up to 40% in the sandstones from the Toukoushan to Tungken formations. It is, however, mostly less than 5% in the sandstones from the Kuanyinshan to Chuhuangkeng formations.

### Discussion

Although the modal proportions and chemical compositions of heavy minerals in the sandstones have produced significant information on the provenance for sandstone, it is important to check the stability and dissolution of the heavy minerals. The post-depositional change of mineral association modifies the original detrital mineralogy, so that discussions based on the present constituent minerals are sometimes questionable. After Pettijohn (1941) studied stabilities of heavy minerals, only a few workers were discussing about their stability (Morton, 1984; McBride, 1985; Smale & Morton, 1987). Stability order of minerals depends on many factors: pore-fluid temperature, pore-fluid movement, pH, burial history, and length of time for dissolution. In many cases, zircon, tourmaline, monazite, and spinel were treated as stable minerals which have not been suffered any dissolution after the deposition.

In the Early Miocene to Late Pleistocene sequence from the Western Foothills, the stable minerals mentioned above are persistent throughout the sequence (Fig. 2). Other persistent minerals are xenotime, apatite, garnet, and rutile (one of  $\text{TiO}_2$  polymorphs). Absence of some of them in the sequence is not the result of the total dissolution, but is due to the restricted number of mineral identification. Other minerals are absent in the lowest part of the sequence. Ilmenite is one of the major minerals in the Tatu River route. It is common in the upper sequence of the Houlung River and is rare or absent in its lower sequence. The decomposition of ilmenite into anatase-bearing assemblage shows that it is relatively unstable

than the persistent minerals mentioned above. Aluminous minerals such as chloritoid, staurolite, and  $\text{Al}_2\text{SiO}_5$  polymorphs are present in the upper part of the Houlung River sequence and absent in the lower sequence. It indicates that the minerals are more easily dissolved than ilmenite.

Epidote, titanite, allanite, grossular, and andradite are common minerals in the low-grade metamorphic rocks. They are observed in the upper part of the Tatu River sequence and in the upper sequence of the Houlung River. Although it is hard to point out the order of stability among them, it is concluded that they are less stable than the aluminous silicates mentioned above. The order of persistence is generally consistent with those obtained by Pettijohn (1941) and Morton (1984).

Hornblende is found only in the upper sequence of the Houlung River. As mentioned by many workers, hornblende is dissolved more easily than epidote and allanite as well as pyroxenes. Pyroxenes are observed only in the Present sand. Both hornblende and pyroxenes are major constituents in the igneous rocks and are common minerals in the high-grade metamorphic rocks as well as garnet. As Mg-rich garnets are common in all sandstones (Fig. 4), it is expected that amphibole and pyroxenes were present in many sandstones at the depositional stage. Their absence in almost all the samples indicates that they are totally dissolved before the dissolution of other heavy minerals.

The stability order of heavy minerals in the sandstones does not depend simply on geological time. In the Tatu River route, epidote, titanite, allanite, and Ca-garnet were not observed in the Pliocene Cholan Formation, but in Houlung River route they are present even in the Yutengping and Kuantaoshan formations underlying the Cholan Formation. Thick conglomerate layer, upper part of the Toukoushan Formation, develops in the Tatu River route. The conglomerate was formed as a fluvial fan deposit at the mouth of a river that had run from the central part of Taiwan. In addition to the geological time, one probable factor for different dissolution system in the nar-

row area may be local overburden pressure. As an alternative explanation, the restricted mineral assemblage in the lower sequence of the Tatu River route is due to a consequence of re-working of older sediments. The explanation is partly reasonable because of the evidence that the sediments in the Cholan Formation were derived from the older sediments in the central part of Taiwan (Chou, 1974; Chen *et al.*, 1992). However, the mineral assemblage in the Cholan Formation in the Houlung River route is not so simple as those in the Tatu River route, but is complex as well as those in the Toukoshan Formation. In any case, it is important to be aware that heavy minerals are easily dissolved and can not be used simply for the provenance study of sandstones, even in the Pliocene or younger sediment. Furthermore, the results are not strictly applicable to particular formations or depositional basins.

As a result of the dissolution of heavy minerals, a restricted mineral assemblage is observed in the heavy fractions of the Early Miocene sandstones. More restricted mineral assemblage such as zircon-tourmaline-monazite-apatite-garnet was reported in most of the Mesozoic sandstones in the Japanese Islands (e.g., Yokoyama & Saito, 2001). In such old sandstones, garnet is variable in pyrope component, but is usually low in grossular component as found in the Early Tertiary sandstones from the Western Foothills of Taiwan (Fig. 4). Presence of Mg-rich garnet suggests that it was derived from the high-grade metamorphic terrane where both Ca-rich and Ca-poor garnets occur in mafic and felsic metamorphic rocks, respectively. It is hard to conclude that only Ca-poor garnet was derived from the terrane at the depositional stage. In drilled cores, Smale and Morton (1987) reported that the Ca-rich garnets decrease with the depth and are scarcely observed in the deeper part of the core. They also suggested that the trend is probably due to an intrastatal solution effect, i.e. selective dissolution. Hence, it is reasonable to conclude that Ca-rich garnet in the Early Miocene sandstones from the Western Foothills of Taiwan had been dissolved after deposition. Although chemi-

cal composition of garnet has been used for provenance study, phenomena of such a selective dissolution of garnet will cause misleading for drainage basin if simply applied the chemical composition to the provenance study.

### Summary

Almost continuous sequence from Late Miocene to Early Pleistocene develops in the Western Foothills, central part of Taiwan. Stability and dissolution of heavy minerals were studied in the sandstones from two routes: Tatu River and Houlung River. Various species of heavy minerals are observed in the sandstones from the upper sequence, whereas mineral species are restricted in the lower sequence, due to the dissolution of some minerals after the deposition. Persistent minerals throughout the sequence are zircon, monazite, xenotime, spinel, apatite, tourmaline, and Ca-poor garnet. Ilmenite, chloritoid, epidote, and pyroxenes are absent in the lowest part. Among the dissolved minerals observed, the order of dissolution is (1) pyroxenes, (2) epidote, allanite, titanite, and Ca-garnet, (3) aluminous silicates (chloritoid, staurolite, and  $Al_2SiO_5$  polymorphs), and (4) ilmenite. The dissolution rate is different between two routes from the Tatu and Houlung rivers, indicating the rate is not simply related with geological time. It is important for provenance study at first to recognize the selective dissolution of heavy minerals in the sandstones.

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## 台湾の Western Foothills 地域に分布する第三紀–第四紀砂岩中の鉍物の安定性と溶解に関する研究

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台湾の Western Foothills 地域には、中新世前期から第四紀前期の地層がほぼ連続して分布する。この地層の砂岩の重鉍物の出現頻度を調べた結果、多くの鉍物は時代が新しいにも関わらず分解や溶解していることが明らかになった。全体を通して安定な鉍物は、ジルコン、電気石、ルチル、モナズ石、燐灰石、スピネルであった。チタン鉄鉍は、中新世前期の砂岩では殆どが分解している。アルミナに富むクロリトイドや十字石は中新世中期の砂岩でも溶解している。緑廉石、チタナイトや灰磐ザクロ石はさらに不安定である。ザクロ石は、すべての砂岩で観察されるが、カルシウムに富むものから選択的に溶解していることが判明した。砂岩中の重鉍物を使って供給源を調べる研究では、それぞれの地域での鉍物の選択的な分解や溶解を調べる必要がある。