

Factors Controlling the Distribution of Zeolites in the Andesitic Rocks of the Central Coastal Range of Eastern Taiwan

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ABSTRACT

This study is a survey of zeolites in the andesitic rocks of the Central Coastal Range (CCR) of eastern Taiwan. Methods of the study include field observations and laboratory techniques such as X-ray diffraction, petrography, and scanning electron microscopy.

It is found that zeolites are abundant only in the andesitic rocks along two streamlets, i.e., Tuwei-Hsi (TH) and Yungshui-Hsi (YH), and are rare to absent in others. Zeolites generally occur in hyaloclastites but rarely in lava flows in the TH section. On the other hand, breccias are usually richer in zeolites than lava flows, peperites, and tuffs in the YH section. Natrolite is the unique zeolite in TH hyaloclastites. It usually associates with apophyllite and calcite. Phillipsite is the only zeolite in YH breccias. It mostly occurs alone and occasionally associates with calcite.

The duration of exposure to seawater is considered as the main factor influencing the amount of zeolites in submarine erupted andesitic rocks of the CCR. The longer the andesitic rocks are exposed to seawater, the larger the amount of zeolites would be in them. On the other hand, the species of zeolite and related authigenic minerals in the andesitic rocks of the CCR are presumably controlled by the kind of their phenocrysts which are the essential minerals suffered from alteration.

(Key words : Natrolite, Phillipsite, Andesitic rocks)

1. INTRODUCTION

Natural zeolites are considered to form essentially in alkaline nonmarine, freshwater and marine environments (Hay, 1996, 1978; Mumpton, 1978; Surdam and Sheppard, 1978; Sheppard, 1989).

This study is a survey of zeolites in the andesitic rocks of the Central Coastal Range of

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eastern Taiwan. These andesitic rocks are regarded as products of submarine eruptions (Song and Lo, 1987), and so provide an example to illustrate the genesis of zeolites in andesitic rocks in a marine environment.

The survey indicates that zeolites are abundant only in the andesitic rocks along two streamlets and are rare to absent in others. Meanwhile, the species of zeolites are quite different in the andesitic rocks along the two streamlets, and so detailed studies are only performed on those andesitic rocks which are rich in zeolites. They include field observations and laboratory techniques such as X-ray diffraction (XRD), petrography and scanning electron microscopy (SEM).

The aim of this study is to ascertain the essential factors controlling the distribution of zeolites in andesitic rocks in a marine environment.

2. ZEOLITE-BEARING ANDESITIC ROCKS

A survey of zeolites and related authigenic minerals in the volcanic rocks from Hsiukulan-Hsi to Yungshui-Hsi of the Central Coastal Range (CCR) of eastern Taiwan was carried out. Igneous rocks of both the Chimei Igneous Complex and the Tuluanshan Formation, especially the latter, are widely distributed in this region.

The igneous rocks exposed in the Chimei area are older than those in the others of the CCR (Hsu, 1956; Ho, 1969; Wang and Yang, 1974; Teng et al., 1988). The older rocks, designated the Chimei Igneous Complex, are comprised of diabase and andesitic lavas and dikes (Hsu, 1956; Ho, 1969; Song, 1990). The others are denoted as the Tuluanshan Formation (TF), and are composed mainly of andesitic pyroclastic rocks (Hsu, 1956; Ho, 1969; Lo et al., 1993).

TF varies widely in thickness at different localities, and may be over 1000 meters thick, but on average, it is about 500 meters (Hsu, 1956; Ho, 1969; Song, 1990). It has been observed that zeolites are prevalent only in the andesitic rocks of TF along two streamlets in the surveyed area. One is Tuwei-Hsi (TH) and the other is Yungshui-Hsi (YH), as shown in Fig. 1. These andesitic rocks are of the middle to late Miocene, as based on radiometric dates (Richard et al., 1986; Chen, 1988; Song, 1990). They have been recognized as products of submarine eruptions (Song and Lo, 1987; Song, 1990).

In the TH section, andesitic rocks with a total thickness of about 500 meters were studied (Fig. 2). They occur as hyaloclastites, pillow breccias and lava flows. Hyaloclastites and lava flows of 20 to 60 meters in thickness occur in alternation in the bottom 200 meters, while hyaloclastites capped with pillow breccia (about 40 meters thick) appear in the top 300 meters of the section. On the other hand, andesitic rocks with a total thickness of about 700 meters were investigated in the YH section (Fig. 3). Volcanic breccias are the main constituents, and lava flows and tuffs occur subordinately in the bottom 450 meters of the section. At the top, Paliwan shale of the Pliocene (Chang, 1967; Chen, 1988) appears (Fig. 3).

The following petrographic descriptions are confined to the hyaloclastites of TH and the breccias of YH. The other volcanic rocks, including lava flows, tuffs and peperites, are excluded because zeolites are rare to absent.

In the hyaloclastites of TH, phenocrysts are subordinate to the predominantly

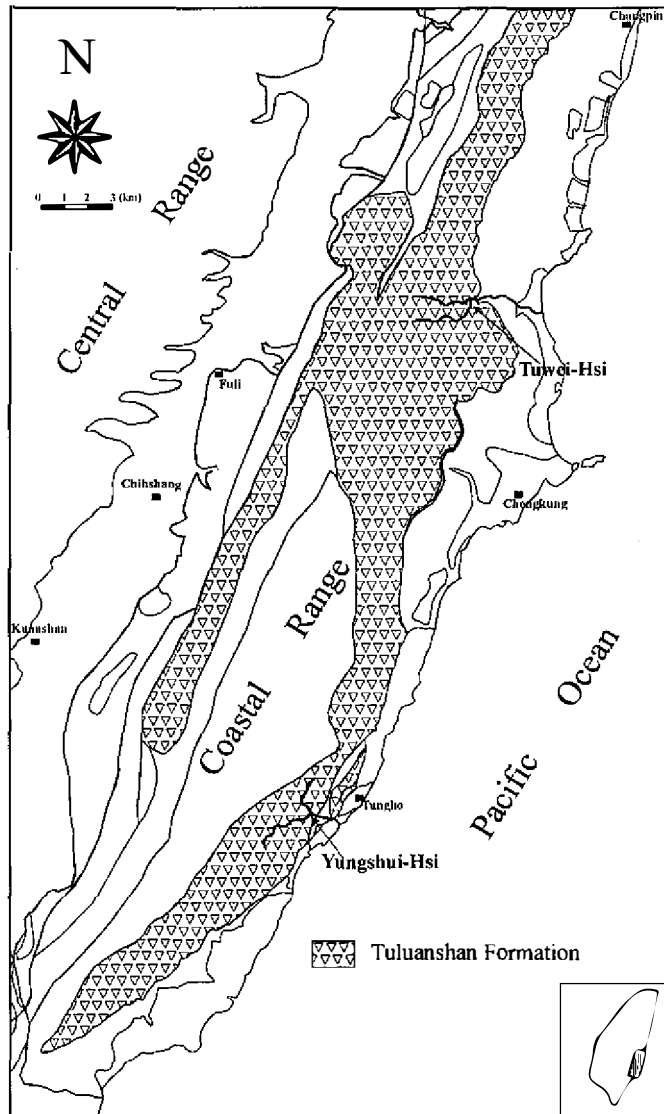


Fig. 1. Map showing the location of Tuwei-Hsi and Yungshui-Hsi, and the distribution of Tulanshan Formation in the CCR.

hypocrystalline groundmass (Pl. IA). The phenocrysts compose 25 to 30 % vol. of the rocks and are comprised of augite, hornblende and bronzite in decreasing proportion. The crystal sizes of the phenocrysts range from 100 to 300 μm and the predominant sizes are 150~200 μm . The groundmass is composed of glass material and microlites of plagioclase and augite.

Phenocrysts are also subordinate to the predominantly holocrystalline to hypocrystalline groundmass in the breccias of YH (Pl. IIA). The phenocrysts compose 30 to 50 % vol. of the rocks. They are comprised of augite, plagioclase and bronzite with augite predominant (about

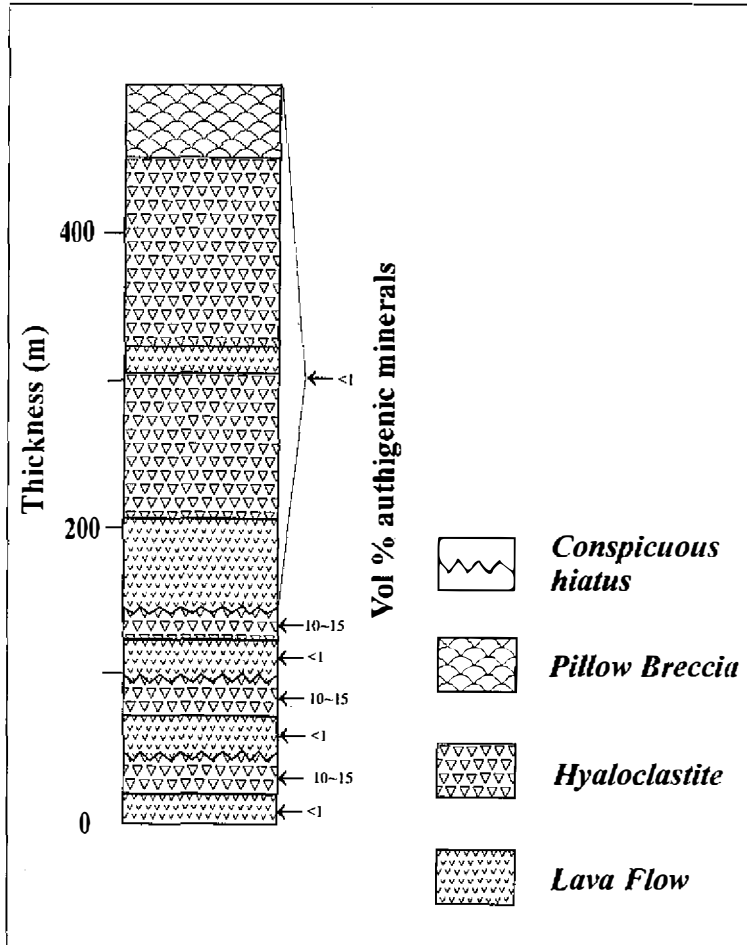


Fig. 2. Lithology of the TH section. Volume % of authigenic minerals in each rock unit being shown.

60~70 % vol. among the phenocrysts). The crystal sizes of the phenocrysts range from 100 to 500 μm and the predominant sizes are 150~250 μm . The groundmass is composed of microlites of plagioclase and augite in phenocryst-rich breccias (40~50 % vol. of phenocrysts), and of microlites of plagioclase and augite, and glass material in phenocryst-poor ones (30~40 % vol. of phenocrysts).

Both TH hyaloclastites and YH breccias show different degrees of alteration. Alteration mainly occurs in phenocrysts, but rarely in groundmass. Special features observed in the altered phenocrysts are micropits and microcracks (Pls. IB, IIB). The phenocrysts also show various susceptibility to alteration. The observed decreasing order of susceptibility is augite \rightarrow hornblende \rightarrow bronzite in TH hyaloclastites and augite \rightarrow bronzite \rightarrow plagioclase in YH breccias. As a whole, the degree of alteration varies from 5 to 20 % vol. and usually decreases from the top to the bottom of a volcanic bed for both TH hyaloclastites and YH breccias.

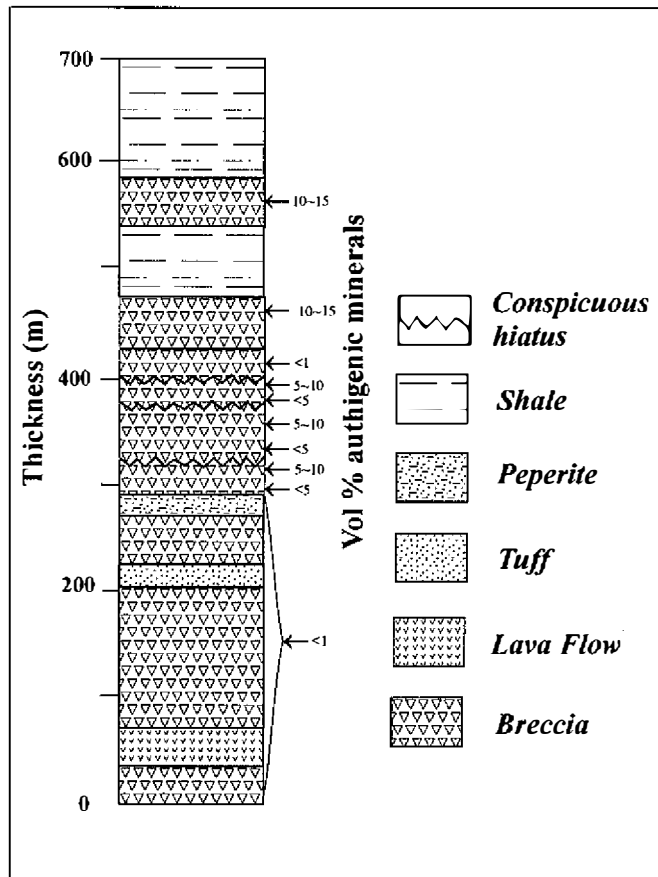


Fig. 3. Lithology of the YH section. Volume % of authigenic minerals in each rock unit being indicated.

3. ZEOLITES AND RELATED AUTHIGENIC MINERALS

Zeolites generally occur in hyaloclastites but rarely in lava flows in the TH section. Meanwhile, thin-bedded hyaloclastites (15~30 m thick) at the bottom are more abundant in zeolites than the thick-bedded ones (100~130m thick) at the top of the section (Fig. 2). Usually, zeolites are not evenly distributed but concentrated in the upper parts (10~15m) in the hyaloclastite beds.

On the other hand, breccias are generally more abundant in zeolites than lava flows, peperites and tuffs in the YH section (Fig. 3). It is also found that breccias at the top are more abundant in zeolites than those at the bottom of the section. Two breccia beds which contact directly with Paliwan shale at the top of the section are not only more abundant in zeolites, but are also evenly distributed with zeolites. As in the TH section, zeolites are also concentrated in the upper parts (10~15m) of the breccia beds, except those in contact with shale.

Thirty and fifty samples of authigenic minerals from the TH and YH sections, respec-

tively, were collected for X-ray diffraction and microscopy studies. Natrolite is found as the unique zeolite in TH hyaloclastites. It rarely occurs alone but usually associates with other authigenic minerals. Mineral associations such as apophyllite-calcite, apophyllite-calcite-natrolite, and apophyllite-natrolite are common. These minerals generally grow in crustification with apophyllite in the outer part (or near the wall), then calcite, and natrolite in the inner part of a vesicle (Pl. IIIA). Natrolite crystals exhibiting good crystal faces in the center of a vesicle of a TH hyaloclastite are shown in Pl. IVA. On the other hand, phillipsite is the only zeolite found in YH breccias (Pl. IIIB). It mostly occurs alone (Pl. IVB) and occasionally associates with calcite.

There are two modes of occurrence of zeolites in the studied volcanics. One is vesicle-filled and the other is fissure-filled. Natrolite, apophyllite and calcite occur mainly as the fissure-filled type, and occasionally as the vesicle-filled one. They may constitute up to 10~15 % vol. of TH hyaloclastites. Meanwhile, phillipsite occurs essentially as the vesicle-filled type and may also make up as much as 10~15 % vol. of YH breccias. The percentage by volume of zeolites was estimated from the fieldwork and petrographic study.

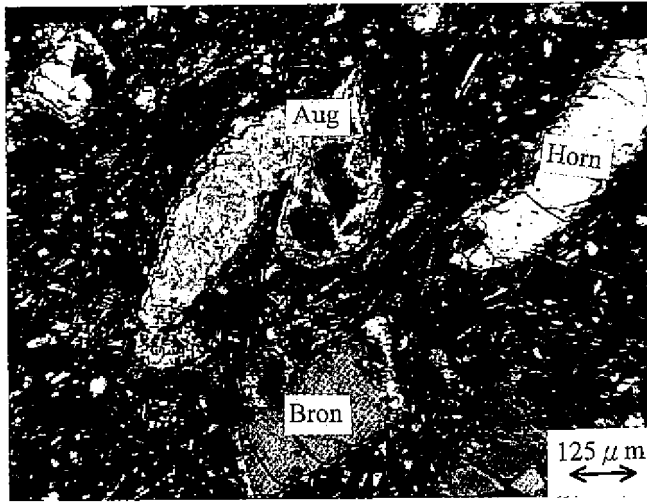
4. DISCUSSION

Based on the above field and laboratory data, the main factors which control the amount and species of zeolites in the rocks studied are discussed below.

The duration of exposure to seawater is believed to be the main factor controlling the amount of zeolites in submarine erupted andesitic rocks of the CCR of eastern Taiwan. The results of the study indicate that the amount of zeolites is correlatable with the degree of alteration of their host rocks. It has long been recognized that the essential factors affecting submarine alteration include seawater / rock ratio, rock permeability, salinity and temperature (Hay, 1966; Honnorez, 1978; Iijma, 1978; Watanabe et al., 1986). Experimental studies show that the reaction rate between a solid and a liquid is proportional to the liquid / solid ratio of the experiments (Lo, unpublished data). So, it is reasonable to assume that submarine alteration will be stronger in a high seawater / rock ratio condition than in a low one. Consequently, rock alteration is higher in a more permeable rock than in a less permeable one. This is merely due to the high liquid / rock ratio of the former. Experimental studies invariably suggest that the rate of rock-liquid interaction is proportional to the salinity of the liquid and that it is accelerated by increasing temperature as well (Lo et al., 1994, 1995, 1998).

It is rational to consider that effects of salinity and temperature on alteration of the volcanic rocks studied are generally similar because they have formed in a similar environment beneath the sea (Song, 1990). Accordingly, if salinity and temperature are excluded, rock permeability and seawater/rock ratio are considered as the two most important factors affecting the rock alteration.

As mentioned previously, the volcanic rocks in the area studied occur as breccias, tuffs and lava flows. It is generally recognized that rock permeability decreases in the order breccias, tuffs and lava flows. This is also confirmed by the field observations and petrographic examinations of the distribution of zeolites and rock alteration in this study. For instance,



A



B

Plate I

- A. Micrograph of a TH hyaloclastite showing predominantly hypocrySTALLINE groundmass with subordinate phenocrysts. Phenocrystic augite (Aug), hornblende (Horn) and bronzite (Bron), and microlitic plagioclase and mafics, and glass material of the groundmass being indicated.
- B. Micrograph of a TH hyaloclastite displaying micropits and microcracks in a partially altered augite crystal.



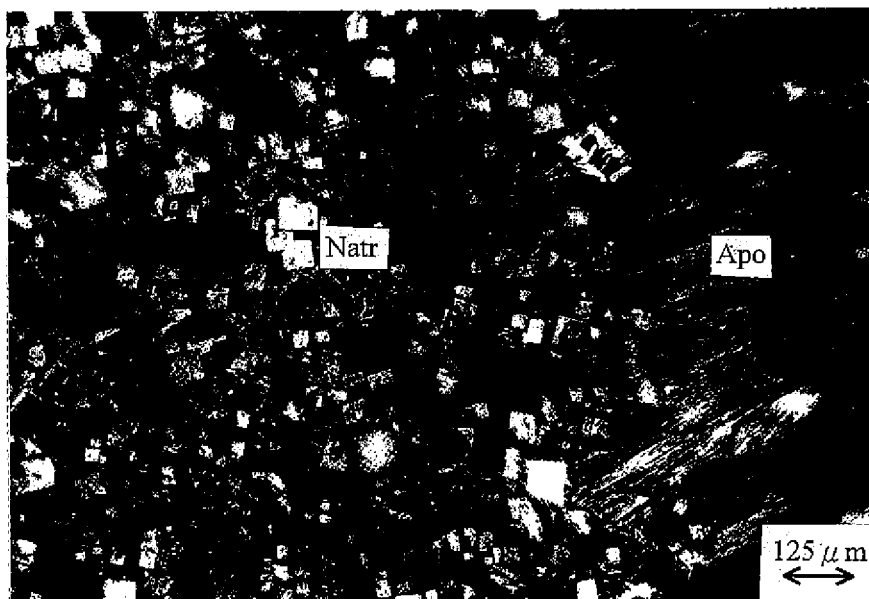
A



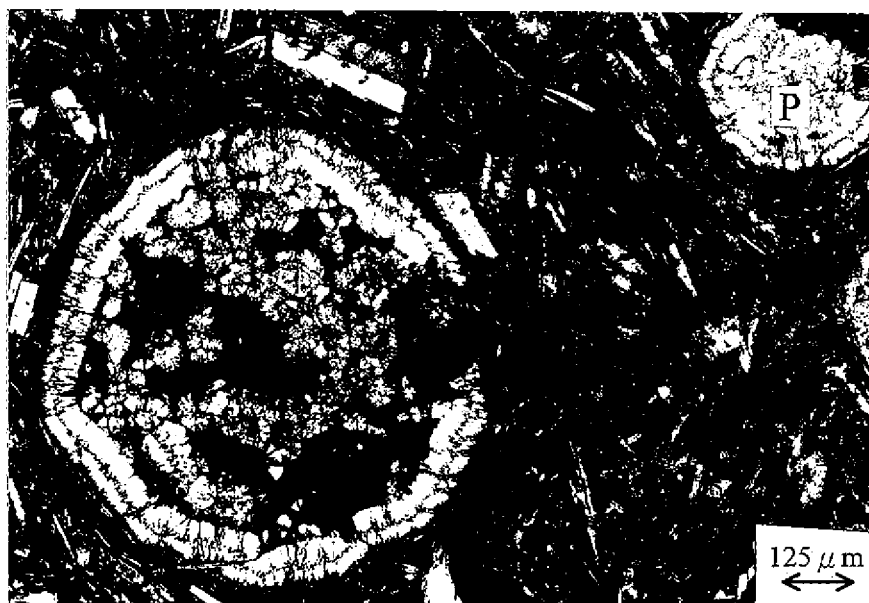
B

Plate II

- A. Micrograph of a YH breccia manifesting predominantly hypocrySTALLINE groundmass with subordinate phenocrysts. Phenocrystic augite (Aug), plagioclase (Plag) and bronzite (Bron), and microlitic plagioclase and mafics, and glass material of the groundmass being shown.
- B. Micrograph of a YH breccia exhibiting micropits, and microcracks (along and cross cleavages) in a partially altered plagioclase crystal(Plag):



A



B

Plate III

- A. Micrograph evincing the growth of apophyllite (Apo) at the wall (right) and natrolite (Natr) at the center (left) in a vesicle of a TH hyaloclastite.
- B. Micrograph displaying vesicle-filled phillipsite (P) in a moderately altered YH breccia.



A



B

Plate IV

- A. Micrograph showing vesicle-filled natrolite exhibiting good crystal faces in a TH hyaloclastite.
- B. Micrograph manifesting vesicle-filled phillipsite indicating good crystal faces in a YH breccia.

comparing two adjacent rock units in both TH and TH volcanic sequences, the amount of zeolites (Figs. 2, 3) and the degree of rock alteration have been observed to decrease in the same order as the recognized rock permeability just stated.

The rock permeability of the studied volcanics is believed to be fairly low even for the most permeable breccias. Observations indicate that the degree of alteration decreases downwards obviously, as do the amount of zeolites in a breccia bed (Fig. 3). In the breccia about 50 meters thick in the YH section, for example, only the rock in the top 2 meters has obviously altered. Clearly, the breccia would alter evenly if its permeability were fairly high.

Rock permeability may certainly influence the formation of zeolites since they appear mostly in more permeable breccias and rarely in less permeable tuffs and lava flows. Nevertheless, it is also noted that the amount of zeolites may vary from none to 15 % vol. in breccias. Consequently, though rock permeability affects the amount of zeolites to some extent, it should not be an essential factor in the present case. The duration of exposure of the andesitic rocks to seawater is here assumed to be the most important factor in controlling the amount of zeolites in the rocks studied. The reasons are discussed below.

As a high seawater/rock ratio can be produced at the surface of a rock when it is directly exposed to seawater, so can be a high rate of rock alteration. This is due to a positive correlation between the seawater/rock ratio and rate of rock alteration (Lo, unpublished data). As previously stated, alteration creates the rocks with expansion microcracks and dissolution micropits which facilitate downward passage of seawater to accelerate the inward alteration of the rocks. The above processes will occur continuously so long as the rock body is exposed to seawater. Consequently, just as the degree of alteration is proportional to the duration of exposure of the andesitic rocks to seawater, so is the amount of zeolites. This argument is strongly supported by field observations of the rocks studied. It is noted that a conspicuous alteration and a considerable amount of zeolites are generally found only in the top 1 to 5 meters of the breccia layers which may range from 50 to 60 meters in thickness. A special case shown in the YH section is that all the breccia layers (about 50~60 meters thick) overlaid with shale (the top two breccia layers in Fig. 3) are strongly altered and evenly distributed with zeolites. It is conceivable that the breccias overlaid with shale may experience longer exposure to seawater than those covered with volcanic rocks.

It is generally accepted that materials for the formation of zeolites and related authigenic minerals are released mostly from the immediate host rocks owing to alteration (Hay, 1966, 1978; Mumpton, 1978; Iijma, 1980; Lo, 1987, 1993; Sheppard, 1989). As previously mentioned, micropits and residues are the main features observed in altered minerals. These observations indicate that dissolution is the most important process for alteration of the rocks studied. The dissolution may be both congruent, as suggested by micropits, and incongruent, as indicated by altered residues which, as proved by XRD and SEM techniques, are mainly composed of montmorillonite. Though both congruent and incongruent dissolutions are observed, the former seems predominant in the rocks studied. The reasons for this are not clearly known, however. Chemical analyses of primary igneous minerals and authigenic minerals are useful in understanding their mutual relationship. Chemical data for the relevant minerals in this study can be taken from literature (Table 1), since the major components are very similar, in spite of the chemical variations of these minerals in nature (Deer et al., 1963; Hay, 1966;

Table 1. Chemical compositions of relevant minerals in this study.

	Aug. ¹	Bron. ¹	Horn. ¹	Plag. ¹	Apo. ²	Natr. ³	P. ⁴
SiO ₂	51.27	53.19	44.18	51.98	52.24	47.60	52.52
Al ₂ O ₃	2.78	2.53	11.63	29.43	n.d.	26.40	17.60
ΣFeO	6.65	16.46	9.63	0.45	n.d.	0.23	0.07
MgO	16.05	26.26	15.99	n.d.	0.11	n.d.	0.09
CaO	21.68	0.66	11.55	13.58	25.11	0.10	5.85
Na ₂ O	0.34	0.04	2.02	3.39	0.27	15.41	0.64
K ₂ O	nil	nil	0.61	0.11	5.05	0.01	4.39
H ₂ O	n.d.	n.d.	n.d.	n.d.	16.41	9.85	14.43
F	n.d.	n.d.	n.d.	n.d.	1.51	n.d.	n.d.
	98.77	99.14	95.61	98.94	100.70	99.60	95.59

1. Data from Yang (1992)

2. Data from Gossner and Kraus (1928)

3. Data from Alberti *et al.*, (1982)

4. Data from Galli and Loschi-Ghittoni (1972)

Abbreviations : Aug., augite; Bron., bronzite; Horn., hornblende; Plag., plagioclase;

Apo., apophyllite; Natr., natrolite; P., phillipsite.

n.d., not determined.

Gottadi and Galli, 1985). As mentioned above, phenocrysts of augite, hornblende and bronzite are the main minerals altered in TH hyaloclastites and those of augite, bronzite and plagioclase are the essential minerals altered in YH breccias. From Table 1, it is conceivable that the pore solutions will gradually enrich Si, Ca (from aug., bron. and horn.), Na and K (from horn. and seawater) in altered TH hyaloclastites, and Si, Ca, Al (from aug., bron. and plag.), Na and K (from plag. and seawater) in altered YH breccias, as alteration proceeds. Consequently, it is not surprising that apophyllite ($KFCa_4Si_8O_{20} \cdot 8H_2O$) is the main authigenic mineral formed in the altered TH hyaloclastites and phillipsite [$K(Ca_{0.5}, Na)_2Al_3Si_5O_{16} \cdot 6H_2O$] is the essential authigenic mineral in the altered YH breccias. The paragenesis of apophyllite, calcite (early) and natrolite (late) in TH hyaloclastites can be easily understood from pore solution chemistry. Since the pore solution is gradually rich in Si, Ca, Na and K during alteration, it is easy to see that apophyllite is ready to form, as is calcite (CO_3^{2-} from seawater). If a certain amount of apophyllite (and calcite) forms from the pore solution, a gradual decrease of Ca and K but an increase of Na and Al (from altered horn.) should eventually lead to the formation of natrolite ($Na_2Al_2Si_3O_{10} \cdot 2H_2O$).

In summary, the duration of exposure to seawater is considered the main factor influencing the amount of zeolites in submarine erupted andesitic rocks of the CCR of eastern Taiwan.

In other words, the longer the andesitic rocks are exposed to seawater, the larger the amount of zeolites would be in them. Consequently, the andesitic rocks which are rich in zeolites may represent the products of a submarine eruption followed by a long quiescent period without volcanic activity. On the other hand, the species of zeolites and related authigenic minerals in the andesitic rocks of the CCR of eastern Taiwan are presumably related to the kind of phenocrysts present, these being the essential minerals suffered from alteration. Thus, apophyllite and calcite appear as early, and natrolite as late, authigenic minerals in TH hyaloclastites, with augite, hornblende and bronzite as phenocrysts. However, phillipsite, occasionally associated with calcite, occurs in YH breccias with augite, bronzite and plagioclase as phenocrysts.

5. CONCLUSIONS

In the volcanic rocks from Hsiukulan-Hsi to Yungshui-Hsi of the CCR of eastern Taiwan, zeolites are abundant only in the andesitic rocks along two streamlets, i.e., Tuwei-Hsi (TH) and Yungshui-Hsi (YH), and are rare to absent in others. Natrolite is the unique zeolite in TH hyaloclastites and usually associates with apophyllite and calcite. On the other hand, phillipsite is the only zeolite in YH breccias and occurs mostly alone. The duration of exposure to seawater and the kind of phenocrysts are considered as the main factors controlling the amount and the species of zeolite and related authigenic minerals, respectively, in submarine erupted andesitic rocks of the CCR of eastern Taiwan.

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