

## MINERALOGY OF DICKITE AND NACRITE FROM NORTHERN TAIWAN

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**Abstract**—Nacrite and dickite are found in two localities in northern Taiwan. One, containing nacrite and dickite, is associated with a gold-energite deposit as a vug-filling clay in the Chinkushih (CKS) mine district near the northern coast. The other is the occurrence of dickite in the interstices of a Miocene coarse-grained quartzose sandstone in the Nanshihchiaio (NSC) area, near Taipei city. X-ray diffraction (XRD), differential thermal analysis (DTA), scanning electron microscope (SEM), and petrographic examinations were used to characterize the mineralogical features. Nacrite most often takes the unusual form of rhombic platelets, and dickite is commonly elongated in habit with the shapes possibly related to their origin. Based on geological evidence, we believe that both nacrite and dickite are of hydrothermal origin. In the CKS area, the formation of nacrite and dickite is related to the hypogene gold-energite mineralization. In contrast, the transformation of dickite in the NSC area is due to the influence of the raised temperatures of sandstone formation, resulting from volcanic activity during the Kungkuang stage after the deposition of the sandstone.

**Key Words**—Alteration, Dickite, Hydrothermal, Nacrite, Taiwan.

### INTRODUCTION

The kaolin group minerals, nacrite and dickite, are polytypes and are rare phases. Ross and Kerr (1930) considered them to be of hydrothermal origin. In recent years, many diagenetic occurrences of dickite have been reported (Segonzac, 1970; Keller, 1976; Frey, 1987; Cellé *et al.*, 1993; Shen *et al.*, 1994; Ruiz Cruz, 1996; Ruiz Cruz and Andreo, 1996). Therefore, it seems that temperature is not the only factor controlling the formation of dickite. Nacrite is believed to be primarily hydrothermal or related to an elevated temperature environment (Hanson *et al.*, 1981; Lippmann, 1982), although authigenetic nacrite was documented by Böhmann (1988). The scant information about occurrences of nacrite is not only related to the rareness of this species, but is also due to the difficulty in identifying polytypes by conventional instrumental methods, especially where nacrite occurs in subordinate amounts or with other kaolin minerals (Prost *et al.*, 1989).

There are only two known localities in northern Taiwan that contain dickite and nacrite. One of them is in the Chinkushih (CKS) mining district near the north coast (Figure 1). Nacrite and dickite at CKS are associated with gold-copper mineralization. In the Nanshihchiaio (NSC) area, a low hilly suburb of Taipei city, dickite is found in the matrix of Miocene quartzose sandstones. The sandstone and kaolin have been mined for ceramic uses.

The objective of this study is to characterize the mineralogy of these occurrences and to investigate the genesis of dickite vs. nacrite. The geochemical conditions for the formation of nacrite and dickite in the CKS and NSC mine areas are deduced from their geological occurrences.

### GEOLOGY AND OCCURRENCE

#### *Geology of CKS mine*

The hypogene CKS gold-copper mining district is located on a coastal hilly region covering an area of ~10 km<sup>2</sup>. This district is underlain by Miocene rocks. The formations are composed of alternating sequences of marine and coal-bearing clastic sediments. They were intruded and partly buried by several dacite bodies of Pleistocene age (Yen and Chen, 1953). The mine sites are located in northern Taiwan at latitude 25°08'N and longitude 121°55'E (Figure 1).

The CKS mine is subdivided geographically into two series, the Penshan intrusive dacite and the sedimentary Changjen series (Figure 1b). The gold-copper deposit of the CKS mine is mainly of fissure-filling and replacement origin and occurs in both dacite and the surrounding sedimentary rocks. The mineralization of the gold-copper deposit exhibits a zonal arrangement. Gold is richer in the upper part (above adit no. 4, 417 m in elevation), and copper ore is largely concentrated in the lower and central parts (Huang, 1955; Juan *et al.*, 1958). Also, hydrothermal wall-rock alteration and zoning are discernible in the dacite body, with an advanced argillization assemblage (Meyer and Hemley, 1967) showing silicified and alunitized rocks in the inner zone and an intermediate outer argillic zone and a broad propylitic zone in the periphery. The outer argillized rock consists of clay minerals roughly in the sequence from illite outwards to kaolinite, halloysite, smectite and subordinate interstratified illite-smectite, and chlorite in the outer weakly altered zone (Chen, 1969, 1972; Wang, 1973). In the Penshan series of the CKS mine, dickite and nacrite are distributed mainly between adits No. 4 and No. 8 from 171 to

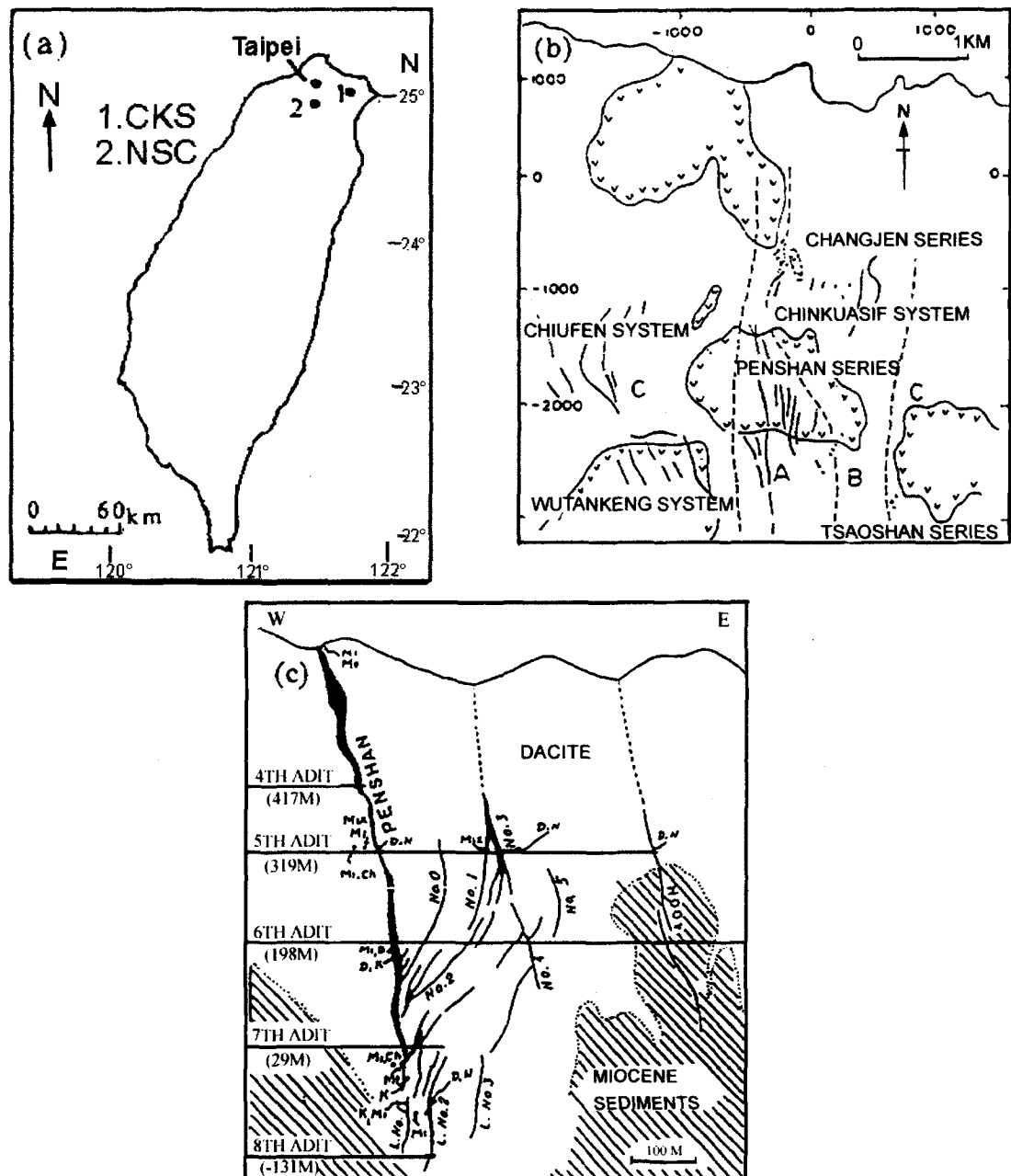


Figure 1. Location and geology of the CKS mine and NSC area. (a) Location map of Taiwan showing the sites of (1) CKS and (2) NSC. (b) Surface geological map of the CKS area showing the location of the dacite bodies and ore system. (c) Geological cross-section of the cone-shaped Penshan dacite and the bearings of ore bodies along a W-E line. (d) Map of 5th adit, showing sampling sites and clay compositions (Chen, 1969).

417 m in elevation, or from 200 to 700 m below the summit of the outcrop of the Penshan dacite (Figure 1c). The locations of dickite-nacrite clays collected from adit No. 5 of the CKS mine are shown in Figure 1d.

Formation of the clay minerals in the outer zones may involve complicated processes. The alteration was

affected by an earlier (pre-metallization) epithermal alteration and later superposed by meteoric processes (Wang, 1973).

The ore minerals in the CKS district are composed mainly of enargite, luzonite, silver-bearing gold and pyrite, and a few other accessory metallic minerals. The common gangue minerals comprise quartz, barite,

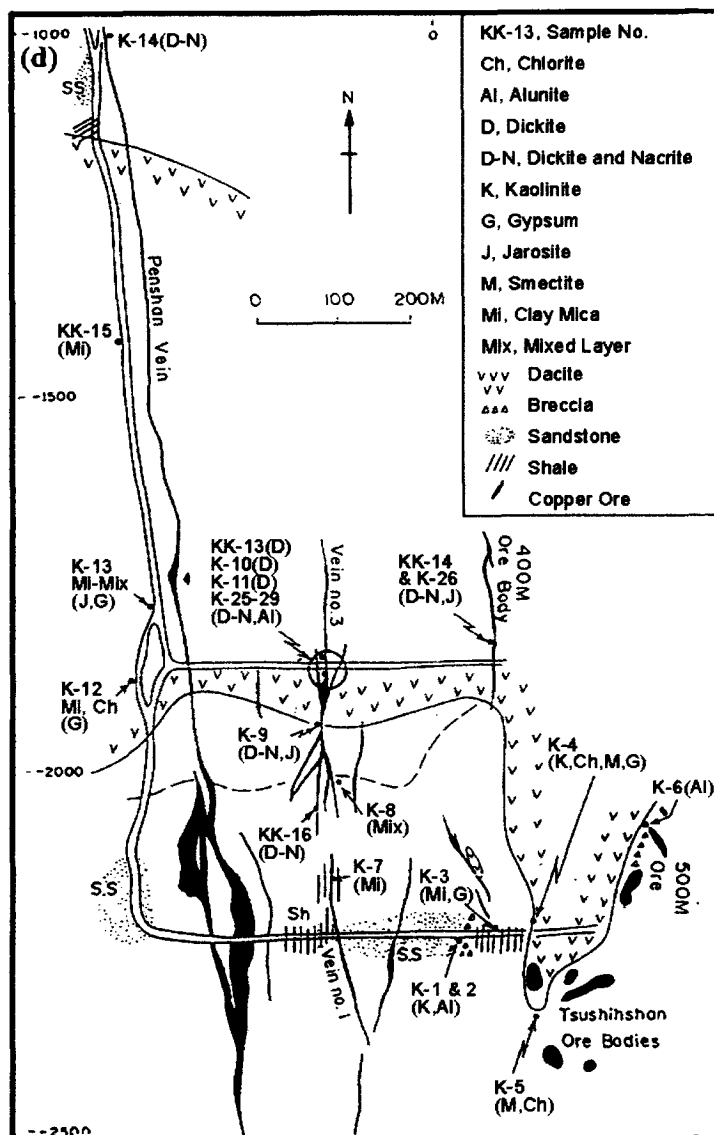


Figure 1. Continued.

alunite, natroalunite, sericite, clay minerals, and with local accessory sulfur, diaspore, limonite, wurtzite and chrysocolla.

#### Occurrence of dickite-nacrite in the metallized zone

There are four modes of dickite and nacrite occurrence in the CKS district: (1) in vugs in veins of enargite or in contact with the ore body; (2) along fractures or joints in adjacent wall rocks; (3) in the interstices of a breccia zone near the ore body; and (4) in the argillized wall rock (largely in dacite) adjoining the ores. The thickness of the clay seams and argillized rock varies from <1 cm to ~2 m wide. There are no significant differences between the occurrence of dick-

ite or nacrite, except that nacrite is commonly present in the argillized dacite adjoining the ore bodies.

Clays of the four modes of occurrence are largely fine-textured, plastic or friable, and white to bluish-gray in color. The clays are acidic (pH ~2–4) due to contamination by fine-grained iron sulfides and water-soluble sulfates. For each occurrence, dickite is commonly the major or only clay component. Nacrite is a predominant clay mineral only in a few samples.

#### Geology of NSC area

The Nanshihchio (NSC) area is a region of low rolling hills and valley flats on the border of Taipei basin (Figure 1). This area consists of sedimentary rocks of

the Mushan formation, which is overlain by basaltic tuff of the Kungkuan formation, both of early Miocene age (Chen, 1969). The Mushan formation is a succession of alternating white sandstones and gray shales, and is coal bearing. The sandstone beds are mostly thick bedded, with medium to very coarse grains. This sandstone unit can be traced ~1.5 km along the strike of the northwestern limb of an anticline. The NSC area is located in northern Taiwan at latitude 24°50'N and longitude 121°35'E (Figure 1).

#### *Petrography of sandstone and occurrence of dickite in the NSC area*

The sandstone comprises ~90% framework grains and 10% matrix. Dickite occurs in the clayey matrix of the pore-space of the quartz grains. This clay matrix is composed of dickite aggregates sometimes associated with lath-shaped illite. In addition to abundant quartz and minor feldspar, muscovite, pyrite, marcasite, and occasionally, traces of barite are the accessory constituents. Locally, stringers or thin intercalations (~10 cm thick) of earthy iron sulfide (mainly marcasite) impregnate the very coarse-grained zone of the sandstone. This dark iron sulfide-bearing portion is usually mottled with abundant white specks of dickite, filling pores or cavities ~1–2 mm in diameter. In addition to iron sulfide, stringers of indurated bitumen are also occasionally found in some parts of the sandstone.

### MATERIALS AND METHODS

#### *Sampling and sample preparation*

A total of 100 clay samples from various rock types from the surface and underground openings in the CKS mine were investigated. Twenty samples consisted mainly of dickite and dickite-nacrite mixtures. The other samples were mainly of kaolinite, illite, chlorite, interstratified illite-smectite, and non-clay minerals, such as microcrystalline quartz, pyrite (octahedral habit), and secondary goethite, gypsum and jarosite.

The samples were dispersed by ultrasonic vibration in deionized water, and the <8 µm particle size-fraction was removed by repeated settling in water. The impurities were largely eliminated from most of the samples by this procedure. The samples were freeze-dried.

Powder X-ray diffraction (XRD) studies were performed using a Philips diffractometer PW1771 with a graphite monochromator. The operating conditions were CuKα radiation at 35 kV and 15 mA. Random powders of the <8 µm size-fractions were scanned from 3–60°2θ at 1°2θ min<sup>-1</sup>. The goniometer was calibrated against quartz, and the peaks yielded sufficient intensity to differentiate dickite and nacrite.

Differential thermal analyses (DTA) were performed on a DuPont 9900 thermal analyzer and the

system was heated at ~10°C min<sup>-1</sup>. Scanning electron microscopy (SEM) was used to examine the textural features of the samples and mineral morphology. Freshly broken chips from samples or <8 µm powdered clay were fixed onto aluminum stubs with double-sided adhesive tape, coated with gold and viewed using a JSM-35 scanning electron microscope.

Clays and argillized rocks were also examined using a polarizing petrographic microscope to determine textural and optical properties. Samples examined included epoxy-impregnated thin-sections and powdered samples on a glass slide, which were dispersed in water or an immersion liquid.

### RESULTS

The XRD patterns of different kaolin minerals in the CKS and NSC areas are shown in Figure 2. To distinguish nacrite from dickite and kaolinite, X-ray patterns with reflections between 20–24 and 34–40°2θ provide the most diagnostic features (Chen, 1977). Nacrite differs from the others in the presence of a doublet at 20.0–20.4°2θ (4.43–4.35 Å) (Figure 2, trace a). Dickite displays a triplet in this 2θ range (compare Figure 2 trace a with trace b), and a prominent peak at 3.79 Å. In the higher angle range, kaolinite usually displays two groups of triplets in the range 34.8–36.0°2θ (2.58–2.50 Å) and 37.8–39.4°2θ (2.38–2.29 Å) (Figure 2, trace d). Dickite has a triplet at 35–36°2θ (the middle one often not distinct) and a single peak at 38.8°2θ (2.32 Å). Nacrite, in addition to a diffused triplet between 34.8–35.7°2θ, features two prominent peaks (or a doublet) at 37.0–37.4°2θ (2.44–2.40 Å) (*cf.* Figure 2 trace a and *vs.* trace b). See also Hanson *et al.* (1981) and Bailey (1963).

The DTA curves of nacrite and dickite differ remarkably from those of kaolinite in the temperature range of 500–700°C where dehydroxylation occurs. The endothermic reaction of the CKS nacrite (Figure 3, trace a) forms a broad and asymmetric valley, from ~500°C to a maximum at ~680°C. In addition, a 'terrace' (sub-endotherm) appears midway at ~580°C. The DTA curves of dickite from the CKS mine (Figure 3, trace c) and from the NSC area (Figure 3, trace d) are similar in shape but the maximum endothermic peak temperature is near 700°C. The differential thermogram of kaolinite (Figure 3, trace e) from the CKS mine shows a single symmetric endothermic peak at 610°C and an exothermic peak at 970°C, which matches the DTA curves of kaolinite of Mackenzie (1957). A small exothermic peak at 420°C is present commonly in both nacrite and dickite-nacrite mixtures, from the CKS mine (Figure 3, trace b). This endothermic peak is probably caused by a small amount of pyrite (Mackenzie, 1957, p. 366). Micro-pyrite with octahedral habit was found concentrated from some clay samples by settling.

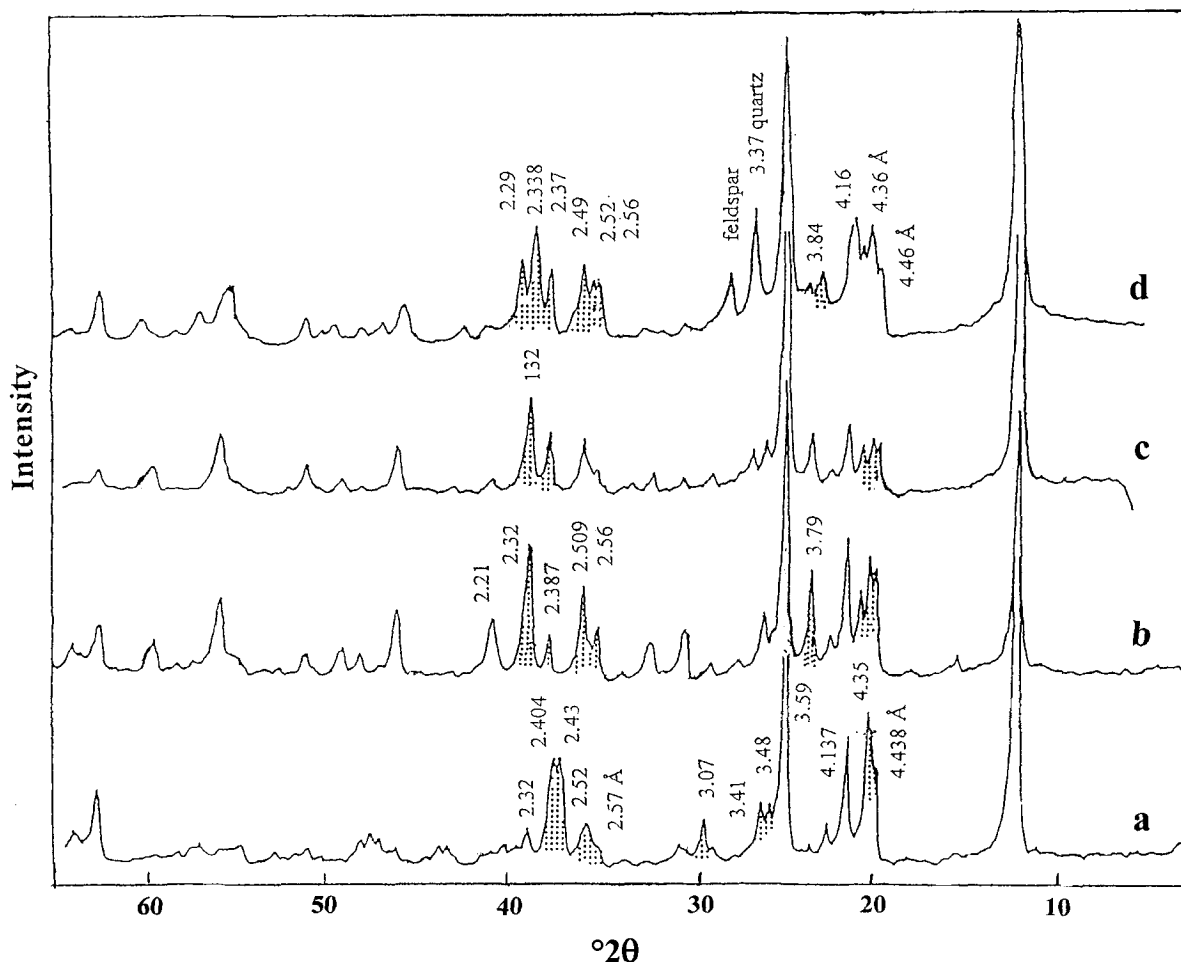


Figure 2. X-ray diffraction patterns of (a) nacrite and (b) dickite from the Penshan series of the CKS mine, (c) dickite in the matrix of quartzose sandstone of NSC mine, and (d) vesicle-filling kaolinite in the weathered basaltic tuffs of the NSC mine.

The DTA curves of nacrite and dickite (Mackenzie, 1957; Brindley and Porter, 1978; Nemeč, 1981; Hanson *et al.*, 1981) show that the temperature of the endothermic and exothermic peaks may vary considerably ( $\sim 50^\circ\text{C}$ ). The shape of the dehydroxylation endothermic peak, however, can be classified into two forms. Some samples exhibit a nearly symmetrical and relatively sharp peak (Mackenzie, 1957; Brindley and Porter, 1978), but most, including those in the present study, exhibit a broad asymmetric peak, often with a sub-endothermic peak at near midway between  $\sim 450^\circ\text{C}$  and the maximum. Brindley and Porter (1978) reviewed the DTA curves of Jamaican dickite and concluded that structurally well-ordered dickite generally gave a sharp endothermic peak near  $700^\circ\text{C}$  and generated a  $14 \text{ \AA}$  *d*-spacing phase when dehydroxylation occurred (Brindley and Wan, 1978). Disordered dickite gives mainly a broad DTA endothermic peak. According to this interpretation, the dickite in our study may be a moderately disordered structure. Because our

DTA curves of both nacrite and dickite have similar broad and asymmetric morphology, the conclusions of Brindley and Porter (1978) concerning dickite probably also apply to nacrite.

Other possible additional effects, such as particle size, thickness of platelet and stacking, as well as adsorbed ions on the clays (Mackenzie, 1957), may modify the peak temperature and the shape of the differential thermograms. The CKS clays commonly contain trace amounts of Sr as detected by X-ray fluorescence analysis and the effect of this trace element requires further study. Schmidt and Heckrodt (1959) in DTA experiments of dickite found that the  $6.3\text{--}20 \mu\text{m}$  size-fractions showed double endothermic peaks, but for the  $<2 \mu\text{m}$  fractions, the higher temperature peak was missing. Kawano and Tomita (1991) showed a similar result and obtained a single, broad and nearly symmetric endothermic peak for dickite at  $564^\circ\text{C}$ . Therefore, particle size influences whether there should be a single or double endothermic peak for dickite.

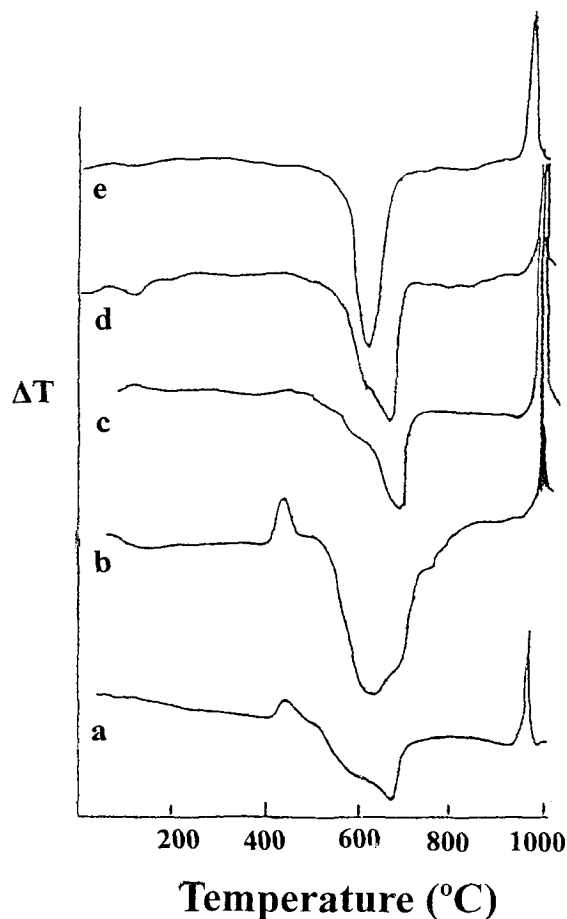


Figure 3. DTA curves of (a) CKS nacrite, (b) sample containing both dickite and nacrite, from the Penshan series, (c) dickite from the Penshan series, (d) dickite in the matrix of quartzose sandstone of the NSC mine, and (e) vesicle-filling kaolinite in the weathered basaltic tuffs of the NSC mine.

The morphology and related petrographic properties of the clays were examined through both light and electron microscopy. In thin-sections of the CKS samples, the clay seams show numerous accordion-like to book-like laminar aggregates of kaolin platelets. These rouleaux of platelets are generally randomly oriented. Partial silicification is commonly observed in argillized rocks. In some argillized rocks (Figure 4), islands of clastic quartz mosaics are surrounded by microcrystalline quartz. Accordion-like kaolin aggregates such as in Figure 4 are a common mode of dickite and nacrite in altered dacite near the enargite vein. In the CKS area, dickite or nacrite is usually associated with alunite but kaolinite is not. In argillized dacite (Figure 5), the phenocrysts, both plagioclase and pyrobole are argillized and replaced by dickite-nacrite (Figure 5a). Moreover the aggregate sizes are larger than for kaolinite books in general.

In all modes of occurrence, dickite is most often the major or the only clay component. Nacrite becomes

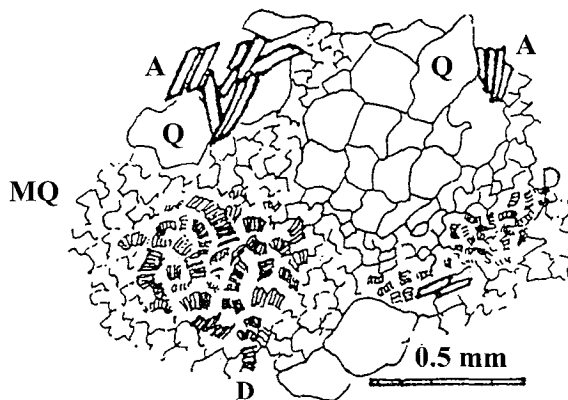


Figure 4. Sketch of microscopic view in thin-section of partly argillized dacite. Q—mosaic quartz, a xenolithic fragment of fine-grained sandstone; MQ—microcrystalline quartz (silicification); A—alunite; and D—rouleaux of dickite.

predominant only in a few samples. Dickite in both CKS and NSC (Figure 6) samples occurs as hexagonal platelets; many are elongated approximately parallel to the pinacoid faces (Figure 5 b). In samples that consist chiefly of nacrite as identified by XRD (Figure 2, trace a), rhombic-shaped platelets become predominant. No apparent difference in particle size between dickite and nacrite was noted. Some large platelets (Figure 7) reach the size  $50 \times 30 \times 0.05 \mu\text{m}$  (Figure 7b), but most are  $<20 \mu\text{m}$ . Vermicular books of dickite (Figures 5 and 7) are commonly found among the samples as shown in Figures 5c and 7a. The larger particles are often composite or cohesive stacks (Chen *et al.*, 1997), or twinned crystals (Mansfield and Bailey, 1972). The rhomboid nacrite platelets exhibit smooth basal crystal surfaces with straight edges, although embayed or irregular edges due to corrosion by solution are common (Figure 5b).

The external shape may be related to the different growth rates of crystal faces or to the fast growth of pinacoid planes and the relatively slow growth of prism faces. Thus, the (100) faces are shortened or truncated by (110) faces (Klein and Hurlbut, 1993; Borchardt-Ott, 1995). Platelets with embayed or corroded edges are common to nacrite and dickite (Figure 5b).

Dickite in sandstone occurs as pore-filling material and is visible by optical microscopy. Dickite occurs as aggregates and forms a mosaic of fan-shaped or accordion-like piles in the interstices of quartz grains (Figure 6a). The dickite mass may occupy the entire pore-space or only the central portions, in which case dickite is commonly surrounded by illite. Dickite crystals are either of hexagonal form or are platelets elongated parallel to the *a* axis (Figure 6b). This elongated habit is common in hydrothermal dickite (Schmidt and Heckrodt, 1959).

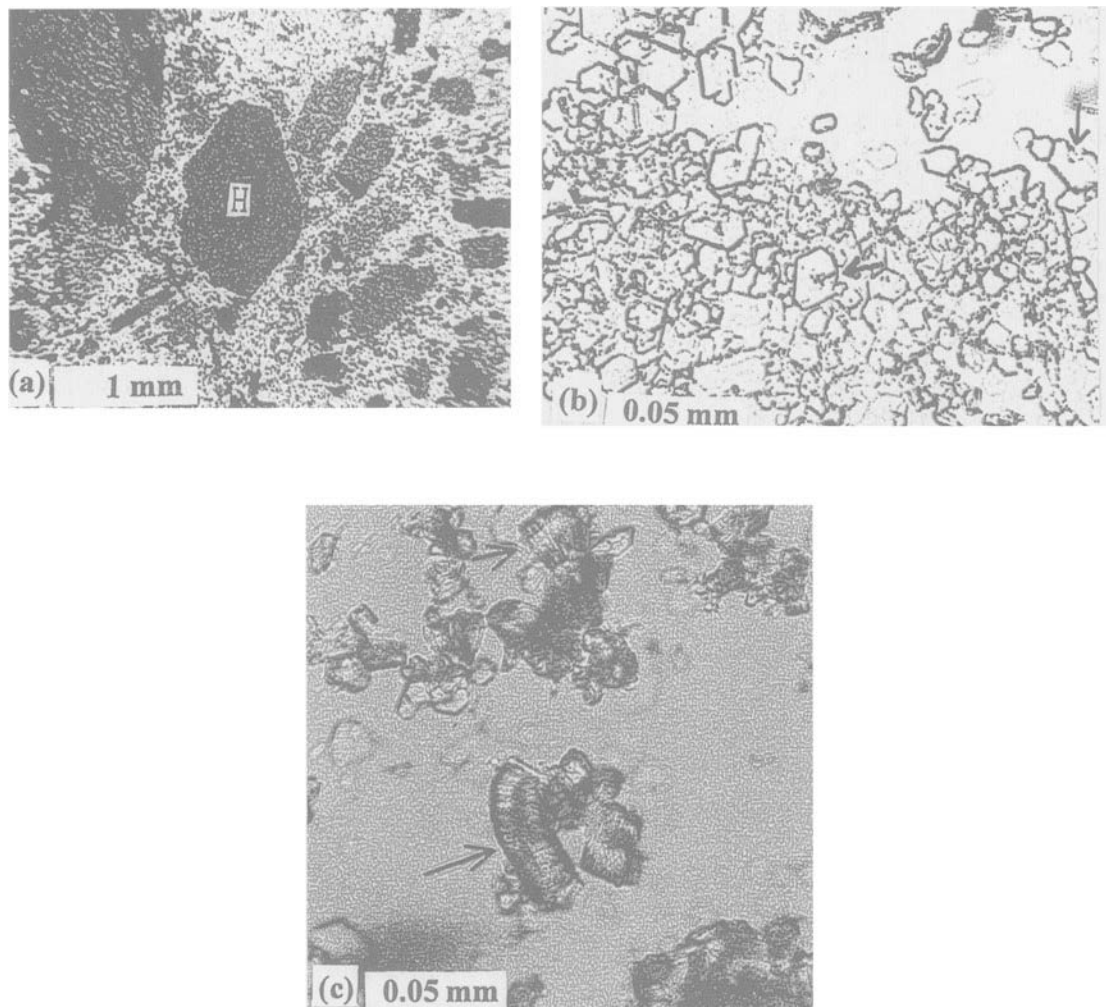


Figure 5. Photomicrographs of (a) selective argillization of the CKS dacite. All phenocrysts (both plagioclase and hornblende) were replaced by dickite ( $\times 200$ ) (H = hornblende). (b) Hexagonal platelets of dickite dispersed in water. (c) Vermicular dickite stack (see arrows) ( $\times 200$ ).

## DISCUSSION

### *Dickite and nacrite in the CKS mines*

Dickite and nacrite, especially the latter polytype, are generally considered hydrothermal minerals (Ross and Kerr, 1930; Hanson *et al.*, 1981; Lippmann, 1982). The presence of both minerals in the CKS mines supports this interpretation. Dickite and nacrite show the following features: (1) the clay occurs mainly in the center of the metallized zone and occurs in vugs of enargite ores; (2) dickite and nacrite occur at depth with the metalliferous veins and are known to occur from 200 to 700 m below the outcrops of the metallized dacite. The metallization is generally believed to be related to post-igneous activity of the Penshan dacite magma and occurred after the emplacement of dac-

ite intrusions during the Pleistocene (Huang, 1955). The K-Ar age of the Penshan dacite is close to 1 Ma (Juang and Chen, 1989). Tan and Wei (1997) reviewed the metallogenetic environment of the CKS mine and suggested that the time of metallization ranged from 0.92–1.08 Ma by K-Ar and Ar-Ar dating. Argillization and alunization occurred at 0.88–0.96 Ma. Different formation temperatures for enargite-luzonite deposition have been estimated by different investigators, but are probably at 200–370°C. The loci for copper ore deposition is estimated to be  $\sim 1000$  m underground. Consequently, the CKS district is a young and shallow metallogenetic province.

The dickite-nacrite is generally cavity-filling between enargite crystals, and to a lesser extent, it occurs in argillized wall rock adjoining the metalliferous

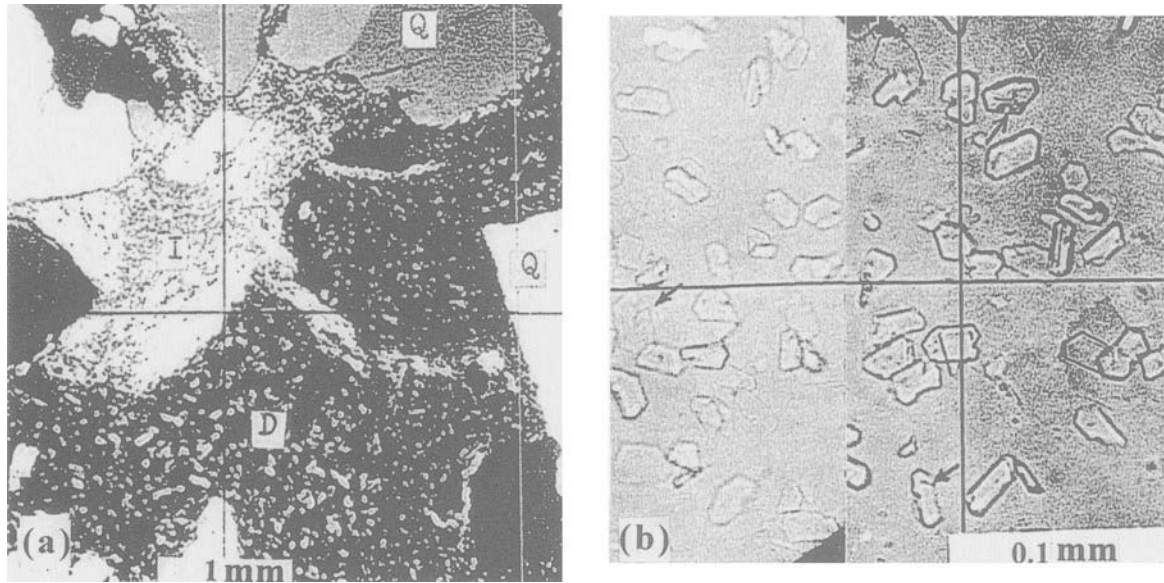


Figure 6. Photomicrographs of (a) aggregates of dickite rouleaux (D), replacing the original illitic clay matrix in the interstices of a quartzose sandstone from the NSC area, and (b) elongated dickite crystals parallel to the pinacoid edge (arrow indicates corroded edge).

veins, as a result of metasomatism. The mode of occurrence of the clay implies that its formation is intimately related to sulfide metallization (Keller, 1988), and that the clay is deposited after crystallization of enargite. Pyrophyllite was identified in some clay samples. The acidic environment favors the formation of pyrophyllite (Keller, 1988). Under low pressure (1–2 kbar), pyrophyllite is stable at temperatures

of 250–350°C (Stringham, 1952; Jacob and Kerr, 1965; Haas and Holdaway, 1973). According to the metallogenetic temperature of this mine district above, the hydrothermal solution leading to the deposition of the clay probably occurred at temperatures ranging from 200 to 300°C. Nacrite may be generated locally where the temperature is greater than the average temperature of dickite formation. Alunite, pyrite and

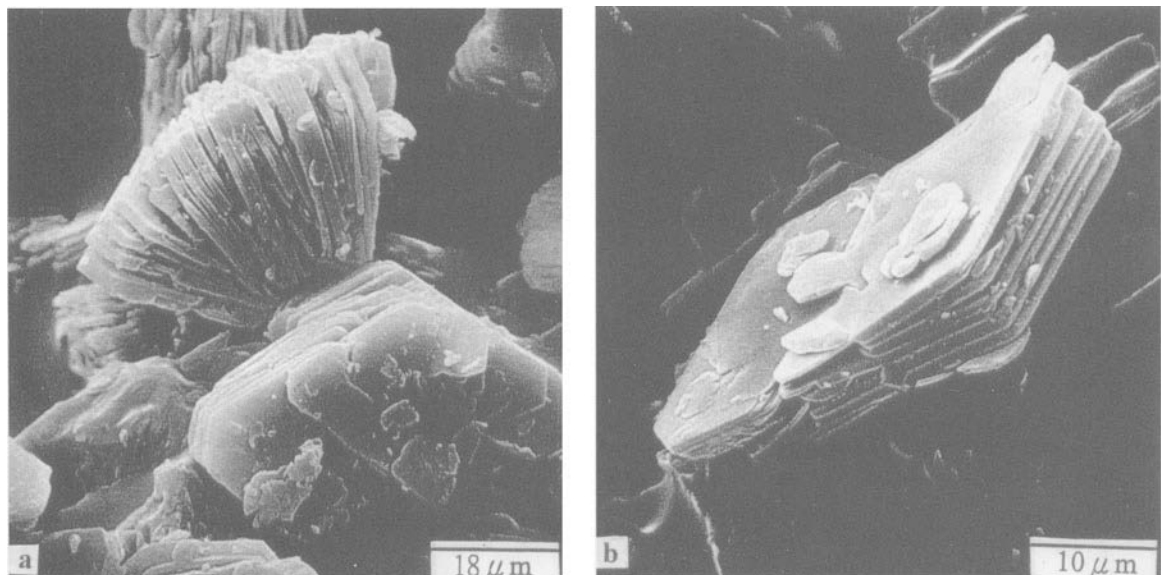


Figure 7. Scanning electron micrographs of (a) nacrite and (b) dickite from the CKS mine. (a) A rouleau composed of platelets with longer (110) edges and shorter (100) edges, and (b) a packet of rhomboid nacrite.



quartz are commonly associated with dickite and nacrite occurrences. All these minerals prefer low to moderate pH environments.

#### *Dickite from the NSC area*

The clay component in the sandstones is assumed to be of sedimentary origin. However, the dickite in the sandstone at the NSC mine is believed to be of hydrothermal origin. This is inferred from the general clay mineral composition of the so-called white sandstone in northern Taiwan. The clay matrix of the Miocene white quartzose sandstone in northern Taiwan is composed of lath-shaped 1M illite and kaolinite with moderate crystallinity (Chen, 1959). In the NSC area, the clay-mineral matrix of the sandstone consists of dickite and minor lath-shaped illite. The textural character of the dickite mosaics in the sandstone matrix indicates that they are apparently not of detrital origin. The pore-filling dickite must be formed by solutions after the deposition of the sediments, either involving growth during diagenesis or as hydrothermal replacement.

From thin-section evidence, the fine-textured illitic clay matrix was displaced or was consumed by the newly-formed dickite (Figure 6a). Larger intergranular spaces favor the growth of dickite, whereas the narrow spaces between grains are favorable for illitic matrix. The effects of elevated temperature on sandstone and shale formations and the origin of the hydrothermal solutions are related to the Kungkuang volcanic activity in this area.

#### *Relationship of genesis to crystal morphology*

Nacrite platelets of rhomboid shape have not been reported previously in the literature. Additional data closely related to the hypogene enargite metallization need further study. We believe that the genesis of the elongate dickite crystals in the CKS and NSC areas has a hydrothermal origin as postulated by Schmidt and Heckrodt (1959).

### CONCLUSIONS

Dickite and nacrite occurring in the CKS mine are mainly formed by vug fracture filling during enargite metallization in the mine. Geological and mineralogical evidence indicates that these clays were formed at a depth of ~1000 m, at temperatures between 200 and 300°C, and deposited by acidic solutions. Several alunite samples were dated by K-Ar and Ar-Ar methods as ~0.95 Ma in age. Alunite and the deposition of dickite and nacrite are thought to be contemporaneous since they are commonly associated with the zone of advanced argillic alteration.

The lower abundance of nacrite relative to dickite in the NSC area was ascribed to the temperature difference; the greater temperature required for nacrite formation occurred only in some places. The transfor-

mation of illitic clay to dickite in the matrix of the late Oligocene quartzose sandstones in the NSC area is believed to be related to elevated temperatures derived from deposition of the Kungkuang tuffs in this area during the early Miocene.

### ACKNOWLEDGMENTS

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