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ARTICLE

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Depth of emplacement, fluid provenance and metallogeny in granitic terranes: a comparison of western Thailand with other tin belts

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Abstract The most important tin mineralization in Thailand is associated with the Late Cretaceous to Middle Tertiary western Thai granite belt. A variety of deposit types are present, in particular pegmatite, vein and greisen styles of mineralization. A feature common to most of the deposits is that they are associated with granites that were emplaced into the Khang Krachan Group, which consists of poorly sorted, carbonaceous, pelitic metasediments. Most of the deposits contain low to moderately saline aqueous fluid inclusions and aqueous-carbonic inclusions with variable CH_4/CO_2 ratios. Low salinity aqueous inclusions represent trapped magmatic fluid in at least one case, the Nong Sua pegmatite, based on their occurrence as primary inclusions in magmatic garnet. Aqueous-carbonic inclusions are commonly secondary and neither the CO_2 nor NaCl contents of these inclusions decrease in progressively younger inclusions, implying that they are not magmatic in origin. Reduced carbon is depleted in the metasediments adjacent to granites and the δD values greisen muscovites are variable, but are as low as −134 per mil, indicative of fluid interaction with organic (graphitic) material. This suggests that the aqueous-carbonic fluid inclusions represent fluids that were produced, at least in part, during contact metamorphism-metasomatism. By comparing the western Thai belt with other Sn-W provinces it is evident that there is a strong correlation between fluid composition and pressure in general. Low to moderately saline aqueous inclusions and aqueous-carbonic inclusions are characteristic of mineralization associated with relatively deep plutonic belts. Mineralized pegmatites are also typically of deeper plutonic belts, and pegmatite-hosted deposits may contain cassiterite that is magmatic (crystallized from granitic melt) or is orthomagmatic-hydrothermal (crystallized from aqueous or aqueous-carbonic fluids) in origin. The magmatic aqueous fluids (those that were exsolved from granitic melts) are interpreted to have had low salinities. As a consequence of the low salinities, tin is partitioned in favour of the melt on vapour saturation. Thus with a high enough degree of fractionation, the crystallization of a magmatic cassiterite (or different Sn phase such as wodginite) is inevitable. Because tin is not partitioned in favour of the vapour phase upon water saturation of the granitic melts, it is proposed that relatively deep vein and greisen systems tend to form by remobilization processes. In addition, many deeper greisen systems are hosted, in part, by carbonaceous pelitic metasediments and the reduced nature of the metasediments may play a key role in remobilizing tin. Sub-volcanic systems by contrast are characterized by high temperature-high salinity fluids. Owing to the high chlorinity, tin is strongly partitioned in favour of the vapour and cassiterite mineralization can form by of orthomagmatic-hydrothermal processes. Similar relationships between the depth of emplacement and fluid composition also appear to apply to other types of granite-hosted deposits, such as different types of molybdenum deposits.

Introduction

Evolved granites are interpreted to be the source of mineralization for most tin deposits. These granites typically contain one or more of muscovite or Li-micas, garnet, fluorite, topaz and tourmaline and their genesis is commonly explained by fractional crystallization followed by high temperature metasomatism, e.g., Charoy (1986). Three different mechanisms have been proposed for cassiterite crystallization:

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1. Magmatic. Direct crystallization of cassiterite from granitic melt, e.g. Linnen et al. (1992);
2. Orthomagmatic-hydrothermal. Partitioning of tin into the vapour phase exsolving from the melt, followed by cassiterite deposition, e.g. Eadington (1983),
3. Hydrothermal redistribution. Leaching of tin from granite and/or country rocks and re-deposition as cassiterite, e.g. Lehmann and Harmanto (1990).

In order for cassiterite to occur as a magmatic mineral tin must be incompatible throughout the crystallization history of the melt and at vapour saturation, tin must either be partitioned in favour of the silicate liquid, or the vapour fraction must be sufficiently small such that the concentration of tin in the melt will continue to increase with crystal fractionation. For orthomagmatic-hydrothermal cassiterite, tin must be partitioned into aqueous fluids that are exsolved from the melt and in redistribution, fluids leach tin from minerals such as biotite during granite alteration and later deposit cassiterite.

The western Thai granite and metallogenic province offers the opportunity to investigate the relative importance of the three mechanisms in the formation of different types of Sn-W ore deposits. It is one of the worlds largest tin belts, containing the most important deposits in Thailand (Suensilpong et al. 1983) and Burma (Zaw 1990). An unusual diversity of mineralization styles are present, including pegmatite, disseminated, vein, greisen, skarn, sulphide-replacement and breccia Sn-W deposits. This belt is particularly well known for an abundance of pegmatite-hosted deposits, which are also an important source of Ta and Nb. The aim of this study is to examine the genesis of the different Sn deposits in this belt in terms of fluid composition, temperature and pressure, in order to explain the diversity of deposit types in terms of the three basic mechanisms, discussed already. The results for western Thailand are then compared to other important belts in order to develop a more general model for tin metallogeny.

Characteristics of the western Thai Sn-W province

Tectonic setting

Southeast Asia comprises several tectonic terranes including the Shan-Thai (or Sibumasu), Indochina, East Malaysia (or Sundaland), and South China blocks (Bunopas and Vella 1983; Lee and Lawyer 1995; Metcalfe 1996). The details of the tectonic history are not completely resolved, but there is consensus that these terranes had amalgamated and collided with Eurasia by the Late Jurassic (Yang et al. 1995). The West Burma block subsequently collided with Southeast Asia in the Late Cretaceous (Metcalfe 1996). Tapponnier et al. (1982) proposed that movement along the major strike-slip faults and rifting in the Gulf of Thailand developed during the Tertiary in response to the Indian collision. There has not been a significant change in the latitude of Indochina since approximately the Middle Jurassic, but a clockwise rotation of ~24° has occurred since that time (Achache et al. 1983). In detail, Southeast Asia did not act as a single block and differential rotation occurred (McCabe et al. 1988; Richter and Fuller 1996), and although the extrusion model of Tapponnier et al. (1982) satisfactorily explains many of the tectonic features of Southeast Asia, movement between the amalgamated terranes and within terranes likely occurred in the Late Mesozoic to Early Tertiary, prior to the onset of the collision of India with Eurasia.

Four major granite belts are recognized in Southeast Asia (Hutchison 1983; Cobbing et al. 1986; Charusiri et al. 1993). The Main Range province consists of "S-type" Triassic granites in Malaysia and southern Thailand. These are biotite and two-mica granites with initial $^{87}Sr/^{86}Sr$ ratios of 0.711 to 0.719. The northern Thai province is characterized by Late Triassic to Early Jurassic biotite and two-mica "S-type" granites (Dunning et al. 1995), but the relationship between the northern Thailand and Main Range granites is not well established. The eastern province consists of "I-type" Permo-Triassic granites in Malaysia and eastern Thailand. These granites contain hornblende and biotite and have lower initial $^{87}Sr/^{86}Sr$ ratios of 0.708 to 0.712. The western Thai province consists of Late Cretaceous to Middle Tertiary "S-type" (biotite, two-mica and leucocratic) granites with minor "I-type" (hornblende-biotite) intrusions in Thailand and Burma. Initial $^{87}Sr/^{86}Sr$ ratios of the western Thai province range from 0.714 to 0.745. The western Thai belt in Thailand is bounded to the north by the Three Pagodas fault and to the southeast by the Khlong Marui fault (Fig. 1). There are other Cretaceous plutons outside of this block but they are not considered to be part of the western Thai province (Cobbing et al. 1992).

Mitchell (1977) first proposed that the western Thai granite belt was emplaced in a back-arc setting, related to the subduction of oceanic crust beneath Eurasia and to the collision of the West Burma block with Southeast Asia. Subsequent models for the tectonic setting of the granites have represented variations on this theme, e.g. Beckinsale et al. (1979), Hutchison (1983), Suensilpong et al. (1983), Mitchell (1985) and Zaw (1990). However, interpretation of the geochronological studies of the biotite and two-mica granites from western Thailand are complicated by resetting. Rb-Sr isochron ages range from 78 to 124 Ma, Rb-Sr mineral ages from 55 to 72 Ma and K-Ar ages from 54 to 74 Ma (Beckinsale et al. 1979; Nakapadungrat 1983; Putthapiban 1984). $^{40}Ar/^{39}Ar$ age dates from Charusiri (1989) and Charusiri et al. (1993) indicate Late Cretaceous to Middle Tertiary ages 80-50 Ma, but even the $^{40}Ar/^{39}Ar$ ages are not straightforward, e.g. at Pilok the mineralization (plateau age) is 2 to 5 Ma older than the granite (total fusion age; 72 Ma). The study of Charusiri (1989) also indicates that the Three Pagodas, Ranong and Khlong Marui faults were active during and after granite emplacement. Movement along these faults during granite emplacement complicates the interpretation of the timing of the exhumation of the different blocks. Mitchell (1985) suggested that the unroofing that exposed the western Thai granites was related to isostatic rebound in the back-arc and Charusiri et al. (1993) also propose that uplift occurred at 65 to 55 Ma. By contrast, Bunopas and Vella (1983) propose that rapid uplift occurred in the Quaternary in an extensional tectonic regime. The timing of uplift or exhumation is important to the discussion below on the depth of emplacement of the granites and mineralization. In considering all these data it is likely that granites were emplaced in a back-arc setting which was tectonically active, dominated by strike-slip faulting. It is probable that some exhumation was coeval with granite emplacement, although the major phase of unroofing occurred during the Tertiary to Quaternary. The different granites within the western Thai belt are separated by strike-slip faults and thus may have been emplaced at slightly different crustal levels.

Granites and ore deposits of the western Thai belt

Several authors have proposed that the western Thai granite belt consists dominantly of S-type granites, with a minor amount of I-type granites. The "S-type" granites essentially consist of biotite, two-mica and muscovite ± tourmaline ± garnet leucocratic granite. These granites typically are ilmenite-bearing, may contain minor amounts of titanite, and have low magnetic susceptibilities (ilmenite series), but exceptionally some "S-type" granites have higher magnetic susceptibilities (magnetite series). The "I-type"

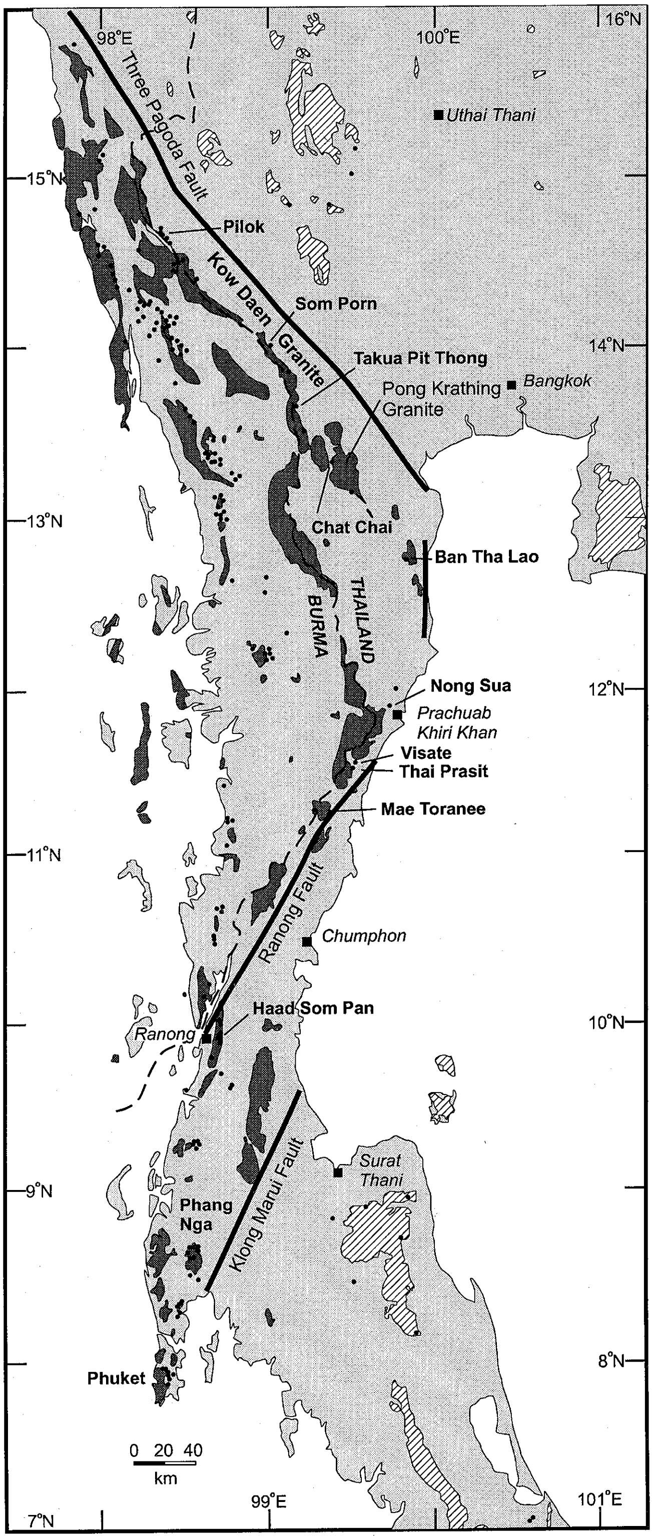


Fig. 1 Location of some of the major primary Sn-W deposits, granites and strike-slip faults of the western Thai province, modified after Schwartz et al. (1995). The abbreviations are *TPFZ*, Three Pagodas fault zone; *RFZ*, Ranong fault zone; *KMFZ*, Klong Marui fault zone. Smaller tin deposits are unlabelled and are represented as *black dots*. Major cities and towns are represented by *black squares*. The western Thai granites are *shaded dark grey* and *diagonal lines* represent other granites

granites typically contain hornblende, biotite and titanite. They may also contain magnetite, as well as ilmenite and have higher magnetic susceptibilities (magnetite series). However, some "S-type" and ilmenite-series biotite granites also contain hornblende. The source(s) of the western Thai granites are not well constrained, therefore, the terms "I-" and "S-type" and "magnetite-" and "ilmenite-series" will be discontinued for the remainder of this work. Mitchell (1985) proposed that the granites of the western Thai belt are anatectic, related to the collision and underthrusting of a continental fragment during the Late Cretaceous, whereas Pollard et al. (1995) maintain that biotite and two-mica granites of Phuket have a meta-igneous source and are direct products of the hornblende granites via fractional crystallization.

The chemistry of the granites has been summarized elsewhere (e.g., Cobbing et al. 1992; Schwartz et al. 1995 and references within) and will only briefly be considered here. The biotite and two-mica granites plot in the syn-collisional field in the Rb versus (Nb + Y) diagram of Pearce et al. (1984) and their compositions are usually near the 1 kbar cotectic in terms of normative Q-Ab-Or. However, the use of normative compositions is not a reliable means of estimating pressure since it assumes granite crystallization along an isobaric water-saturated cotectic, and the effects of B, Li, F, Al, Ca, Fe etc., are ignored (see Johannes and Holtz 1996 for a review of the parameters that affect granite minimum compositions). One constraint on the pressure of crystallization of granitic melts is the intersection of the alkali feldspar critical curve with the haplogranite solidus. This occurs at ~2500 bar (Martin and Bonin 1976), although the addition of F can shift this intersection, e.g. hypersolvus St. Austell granites can crystallize at ~1500 bar (Weidner and Martin (1987). Crystallization at 1 kbar will result in the formation of hypersolvus granites, whereas all the granites in Thailand are subsolvus. A deeper emplacement thus is more likely, discussed later, which is supported by a lack of coeval volcanism. A few deposits in western Thailand are associated with limestone. These include, sulphide replacement and skarn Sn-W (Takua Pit Thong, Som Porn) as well as cassiterite associated with calc-silicate alteration (Som Porn). In these cases the composition of the host rock played an important role in controlling the type of mineralization. However, the western Thai granites and associated mineral deposits were more commonly emplaced into the Kaeng Krachan Group or its stratigraphic equivalent. This is a thick sequence of carbonaceous, poorly sorted pelite, sandstone and pebbly sandstone. Cassiterite at most deposits is associated with granite-metasediment contacts, and at some locations mineralization is hosted by the metasediments, as well as granite or pegmatite.

In this study the granites and mineral deposits of the western Thai belt are divided into northern and southern regions, based on their location with respect to the Ranong fault (Fig. 1). Descriptions of most of the deposits from the northern region are found in Linnen (1992), from the southern region in Charusiri (1989) and many of the deposits in both regions are discussed in Schwartz et al. (1995). A list of references for each mineral deposit is given in Table 1. The northern region can be further divided into four areas. From north to south these are the Kow Daen, Pong Krathing, Prachuab Khiri Khan and Ranong areas. In the Kow Daen area the largest deposits are at Pilok. Mineralization in this area dominantly consists of cassiterite-wolframite hosted by quartz veins and disseminated in greisen alteration. A minor amount of scheelite is present as late, fracture-filled alteration of wolframite. Significant Cu-Zn mineralization is also present at Pilok but these metals are not recovered. Miarolitic pegmatites are relatively rare and are typically barren but may contain minor molybdenite or wolframite ± cassiterite. Quartz vein hosted wolframite-cassiterite deposits are also associated with the extension of this granite belt in Burma (Zaw 1990). At Som Porn, further south in the Kow Daen area, cassiterite mineralization is hosted dominantly by quartz veins, but a variety of other mineralization styles are present, in part due to the intrusion of granite into limestone as well as carbonaceous pelite. Cassiterite is present disseminated in granite, with sulphide replacement of carbonate, associated with calc-silicate alteration, and in veins that cut limestone with no apparent alteration. In addition, cassiterite that is probably magmatic in origin is

Table 1 Some of the Sn-W deposits in western Thailand

Deposit-area	Mineralization	Comments	Reference
Northern Region			
Pilok – Kow Daen	Stockwork-vein	Boron enriched	Linnen and Williams-Jones (1995)
Som Porn – Kow Daen	Stockwork, pegmatite disseminated	Minor lepidolite	Linnen (1992)
Takua Pit Thong – Pong Krathing	Sulphide replacement, disseminated, skarn, breccia	Fluorine enriched	Linnen (1992)
Chat Chai – Pong Krathing	Stockwork	Fluorine enriched	Linnen (1992)
Ban Tha Lao (Hub Kapong) – Prachuab Khiri Khan	Vein	Boron enriched	Linnen (1992)
Nong Sua – Prachuab Khiri Khan	Pegmatite	Boron enriched	Linnen et al. (1992), Linnen and Williams-Jones (1993, 1994)
Thai Prasit (Tap Sakae) – Prachuab Khiri Khan	"Pegmatite"	Quartz-muscovite ± feldspar dykes	Linnen (1992)
Visate (Tap Sakae) – Prachuab Khiri Khan	Pegmatite	Boron enriched	Charusiri (1989)
Mae Toranee (Bang Saphan) – Prachuab Khiri Khan	Pegmatite	K-feldspar rich	Charusiri (1989)
Haad Som Pan – Ranong	Disseminated, stockwork	Boron enriched	Charusiri (1989)
Khao Chai – Ranong	Vein, stockwork	boron enriched	Charusiri (1989)
Southern region			
Nok Hook – Phang Nga	Disseminated	Boron enriched	Charusiri (1989), Pollard et al. (1991)
Juti – Phang Nga	Pegmatite	Boron enriched	Charusiri (1989)
Ngan Tawee – Phang Nga	Pegmatite	Boron enriched	Charusiri (1989)
Bang-I-Tum – Phang Nga	Pegmatite	Boron enriched, minor lithium	Garson et al. (1975) Charusiri (1989)
Reung Kiet – Pnang Nga	Pegmatite	Lithium enriched	Garson et al. (1975) Charusiri (1989)
Ban Nguan – Phuket	Pegmatite	Lithium enriched	Garson et al. (1975) Charusiri (1989)
Tor Soong – Phuket	Pegmatite	Boron enriched	Charusiri (1989)
Chao Fa – Phuket	Pegmatite	Boron enriched	Charusiri (1989)
Khao Kow – Phuket	Disseminated, stockwork	Boron enriched	Charusiri (1989)

disseminated in pegmatites at Som Porn. The pegmatites are thin (<1 m) dykes that contain coarse-grained K-feldspar and albite in a matrix of quartz, white mica (of unknown Li content), garnet, tourmaline and cassiterite. The cassiterite crystals also contain inclusions of Ta-Nb oxides. Som Porn is also one of the few locations in the northern region where lepidolite is present. However, it is noteworthy that Som Porn is a W-poor deposit.

The granites in the Pong Krathing area are fluorine-rich, reflected by abundant fluorite at the Takua Pit Thong and Chat Chai tin mines, F-rich biotite and amphibole at the former deposit and granite-associated fluorite deposits elsewhere in this area. Tin mineralization at Chat Chai occurs dominantly in quartz veins associated with greisen altered granite. The greisen alteration consists of quartz, muscovite, fluorite and rarely topaz. Sulphide minerals, pyrite in particular, are also associated with greisen, similar to Pilok. At Takua Pit Thong tin mineralization is hosted by sulphide replacement deposits in calc-silicate or marble roof pendants in leucocratic granite. Less abundant styles of mineralization include disseminated Sn-Ta-Nb mineralization in feldspathic altered granite, scheelite hosted by garnet-pyroxene skarn and breccia-hosted cassiterite mineralization.

The Prachuab Khiri Khan area hosts several pegmatite-hosted tin deposits, e.g. at Nong Sua, Tap Sakae and Bang Saphan (Fig. 1). Most of these pegmatites are boron-rich but typically contain low concentrations of Li, F, Ta and Nb. At Nong Sua the earliest mineralization is cassiterite disseminated in aplite and pegmatite, that is interpreted to have crystallized from melt. This was followed by cassiterite-wolframite hosted by quartz-tourmaline veins. Mineralization at the Thai Prasit deposit is peculiar for a pegmatite-hosted deposit. Cassiterite is disseminated in thin dykes that crosscut pelitic metasediments, similar to elsewhere, but the 'dykes' are composed almost entirely of coarse-grained quartz and muscovite, with only minor amounts of feldspars. They are in sharp contact with the wallrock and there is no apparent alteration of the metasediments at the 'dyke' contacts. Cassiterite crystals from these dykes contain abundant inclusions of Nb-Ta oxides, in contrast with other pegmatites in the area. Vein-hosted mineralization is present at Ban Tha Lao, near Hub Kapong. At this location cassiterite is hosted in quartz veins that cut tourmaline-muscovite granite within 20 m of the contact with pelitic metasediments (Khang Krachan Group). In the Ranong area disseminated, greisen and vein-hosted cassiterite-wolframite deposits at Haad Som Pan are associated with tourmaline-rich leucocratic granite. Quartz vein hosted mineralization at Kow Chai is tungsten-rich (as wolframite), with only minor cassiterite.

In the southern region (south of the Ranong fault) numerous deposits are located in the Phang Nga and Phuket areas. These include vein and disseminated styles of mineralization, but of particular importance is the abundance of pegmatites (Table 1). These pegmatites are typically Li- or B-rich and contain Ta-Nb minerals (most commonly columbite) which are recovered as a by-product of tin mining. In the Phang Nga area tin mineralized tourmaline-muscovite pegmatites have been mined at the Juti, Ngan Tawee, Bang-E-Tum deposits. Reung Kiet is a cassiterite-bearing lepidolite pegmatite, and the Nok Hook mine contains disseminated cassiterite mineralization as well as mineralized tourmaline-muscovite pegmatites. At Phuket, the Tor Soong, Tantikovit and Chao Fa pegmatites contain tourmaline and muscovite, whereas Ban Nguan, Pad Roi and Sinpatana are lepidolite pegmatites, and the Sahakit mine (Khao Kao) is a disseminated and quartz vein-hosted cassiterite deposit.

The major element chemistry of the granites in both regions is similar but there are few trace element analyses of lithophile and high field strength elements (Li, F, B, Sn, Ta, Nb etc.). Granites from the northern region are lithium- and fluorine-poor (excluding Pong Krathing which is likely F-rich, but there are no analyses). The granites at Pilok contain a maximum of 150 ppm Li and 3200 ppm F (Lehmann et al. 1994, and Lehmann, personal communication cited in Linnen and Williams-Jones 1995). Aplite and pegmatite at Nong Sua similarly contain a maximum of $\sim$30 ppm Li and 480 ppm F (Linnen et al. 1992). By contrast, the granites in the southern region contain up to 1000 ppm Li and 1.4 wt.% F (Pollard et al. 1995). The relative abundances of lepidolite, fluorite, tourmaline, columbite and wolframite also vary with location. The tin deposits in the southern region commonly contain minerals of Ta-Nb (dominantly columbite) and Li (lepidolite) but tungsten mineralization is less common. This is not to say that W mineralization is completely absent in the southern region. Wolframoixiolite for example is reported for pegmatites from Phuket (Suwimonprecha et al. 1995), but wolframite (huebnerite) is only reported as a late-stage mineral. The amount of tungsten produced from southern (Phuket and Takua Pa) region is < 1% of that produced from the northern (Kanchanburi) region (Charoensri 1982). Minerals of Ta, Nb and Li, are less common in the northern region, but wolframite is an important ore mineral. Further north in the Chaing Mae area, the Sn-W deposits (e.g. Samoeng, Mae Chedi, Mae Liang and Doi Mok) similarly are mined for tungsten, but do not contain significant Li, Nb or Ta (Charoensri 1982). However, the latter deposits lie outside the western Thai granite belt and will not be considered further. The association of Nb and Ta with Li can be explained in part by the higher solubility of columbite-tantalite in granitic melts with increasing amounts of Li (Linnen 1997), and in part from extreme fractionation which concentrates both Li and rare metals. It is not clear why there is a general antipathetic relation between these elements and W.

Fluid inclusion and stable isotope constraints on tin mineralization

Fluid inclusions from tin deposits in western Thailand are well documented for the Nong Sua aplite-pegmatite (Linnen and Williams-Jones 1994) and the Pilok vein-greisen deposits (Linnen and Williams-Jones 1995) in Thailand, and in the Burmese portion of this belt, vein type deposits at Mawchi (Zaw and Thet 1983) and at the Pennaichaung and Yetkanzintaung prospects in Tavoy Township (Zaw 1984). Additional data from reconnaissance investigations are provided by Jackson and Helgeson (1985), Charusiri (1989) and Linnen (1992). A limited number of oxygen isotope determinations on barren western Thai granites were reported by Kerrich and Beckinsale (1988), and oxygen and hydrogen isotopic compositions were determined for Nong Sua (Linnen and Williams-Jones 1994) and Pilok (Lehmann et al. 1994; Linnen and Williams-Jones 1995).

Table 2 summarizes the fluid inclusion and isotopic data from the Nong Sua and Pilok deposits. Fluids of very similar composition are present at both deposits, despite the contrast between pegmatite and greisen mineralization styles. The magmatic fluid at Nong Sua is preserved in primary low salinity aqueous fluid inclusions in magmatic garnet, and at Pilok the magmatic fluid is also interpreted to have had a low to moderately salinity. The primary magmatic aqueous inclusions at Nong Sua lack CO_2 (CO_2 concentrations are below Raman detection limits and vapour bubble pressures

Table 2 Comparison of fluid inclusion and stable isotope data

Inclusion/isotope	Nong Sua	Pilok
Aqueous (L)	Absent	Late stage, < 1 wt.% NaCl equivalent
Aqueous (L-V)	Early orthomagmatic $T_h \sim 300$ °C, $\sim$3 wt.% NaCl equivalent, later external $T_h \sim 400$ °C, $\sim$20 wt.% NaCl equivalent	Older $T_h \sim 300$ °C, 10 wt.% NaCl equivalent, younger $T_h \sim 200$ °C, 2 wt.% NaCl equivalent
Aqueous (L-V-H)	Late, 33 to 36 wt.% NaCl equivalent, significant Ca, Fe, Mn, K	Late, 32 to 35 wt.% NaCl equivalent, significant bivalent cations probable
Aqueous-carbonic (L-L-V)	Indeterminate origin, X_{CH_4} 0.0 to 0.15, T_h 300 ° to 400 °C, 0 to 20 wt.% NaCl equivalent	Indeterminate origin, X_{CH_4} 0.0 to 0.2, T_h 280 ° to 320 °C, < 6 wt.% NaCl equivalent
Aqueous-carbonic (V-L-L)	Indeterminate origin, X_{CH_4} 0.0 to 0.25, T_h 300 ° to 400 °C, 0 to 15 wt.% NaCl equivalent	Indeterminate origin, X_{CH_4} 0.0 to 0.3, T_h 280 ° to 400 °C, < 6 wt.% NaCl equivalent
δ^{18}O qtz	12.3 to 12.7 per mil magmatic, 11.5 to 12.9 per mil hydrothermal	10.8 and 10.9 per mil pegmatite, 9.1 and 9.4 per mil wolf-peg, 8.9 to 12.4 per mil greisen
δ^{18}O mu	9.8 to 10.2 per mil magmatic, 8.0 to 9.4 per mil hydrothermal	8.2 and 8.9 per mil pegmatite, 7.1 and 9.3 per mil wolf-peg, 6.1 to 9.0 per mil greisen
δ^{18}O cas	4.9 to 6.4 per mil disseminated and vein	3.4 to 3.7 per mil greisen and vein
δ D mu	−91 to −106 per mil magmatic, −85 to −88 per mil hydrothermal	−100 and −114 per mil magmatic, −108 to −134 per mil greisen

Abbreviations are qtz, quartz; mu, muscovite; cas, cassiterite; wolf-peg, wolframite-bearing pegmatite

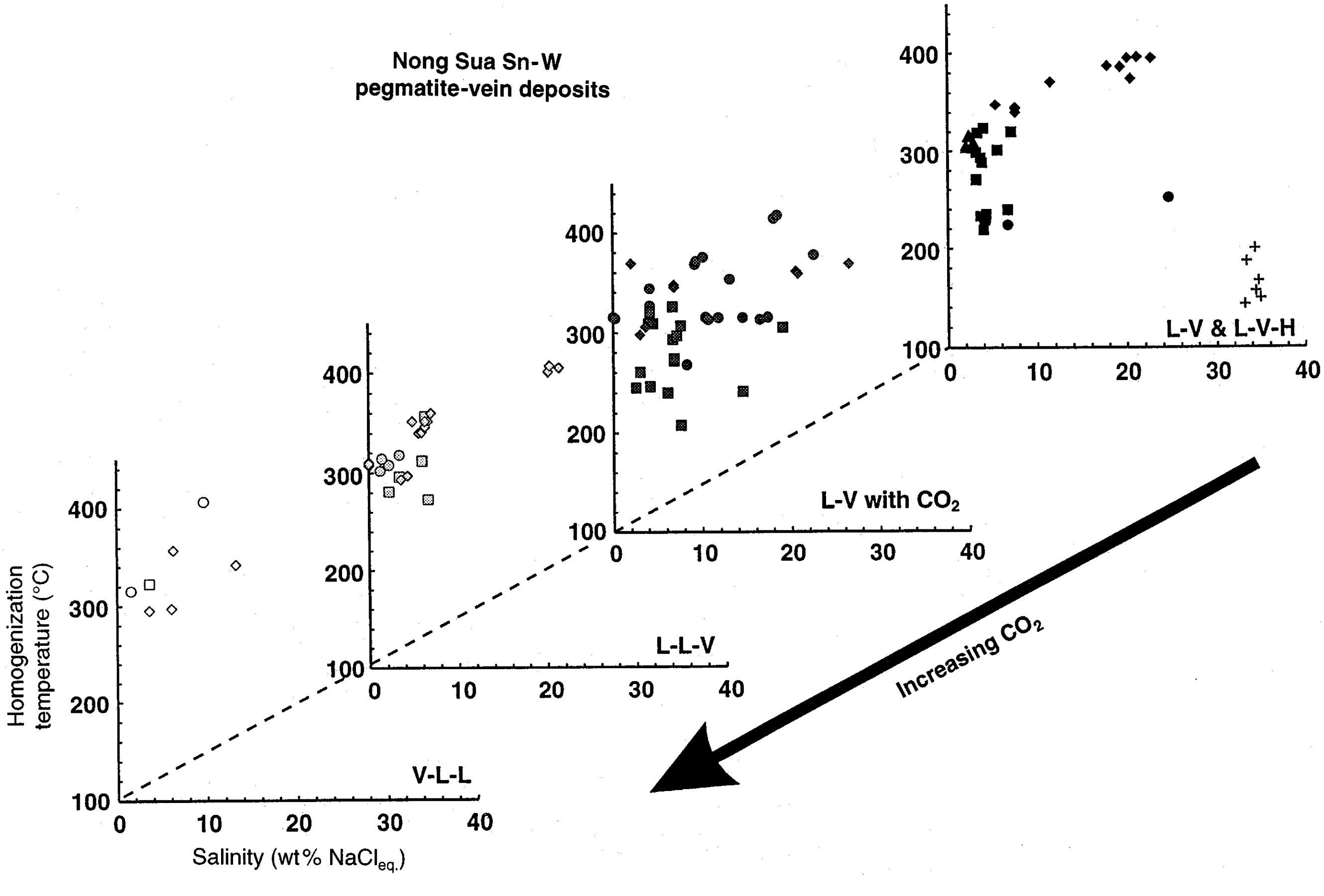
<1 bar, established by crushing tests). CO_2 is preferentially partitioned into a vapour phase coexisting with a granitic melt, discussed later, which implies that the younger, H_2O-CO_2 fluid inclusions are not magmatic in origin.

One of the most striking similarities between the fluid inclusions at Nong Sua and Pilok is the composition and homogenization temperature of the aqueous-carbonic inclusions (Figs. 2, 3). Aqueous-carbonic fluid inclusions commonly occur as secondary inclusions in quartz. At both locations a metamorphic origin is proposed for the aqueous-carbonic fluids, involving different organic carbon contents and fo_2 conditions to account for variations in the proportions of H_2O-CO_2-CH_4. This is based on:

1. The relatively young age of the aqueous-carbonic inclusions make a magmatic origin unlikely, as discussed already.
2. The δD values of greisen muscovite at Pilok are as low as −134 per mil (Table 2) and the calculated fluid compositions are as low as −118 per mil (Fig. 4). Given that Thailand's latitude has not changed significantly since the Late Cretaceous, low δD values very likely reflect fluid interaction with organic (graphitic) material.
3. The metasedimentary enclaves at Nong Sua (biotite schists) are depleted in reduced carbon, containing an average of 0.13 wt.% C, compared to the regional low-grade metamorphic equivalents (pelitic sandstones and locally, black shales) which contain up to 6.5 wt.% C. Similarly at Pilok, the reduced carbon contents in metasedimentary rocks are progressively lower with distance toward the granite contact, ranging from up to 1.2 wt.% C in regional metamorphic pelites to <0.01 wt.% C in biotite-muscovite schists and tourmaline altered metasediments at the granite contacts.

This implies volatilization of carbon or leaching of carbon by fluid-rock interaction during contact metamorphism-metasomatism. A metamorphic source for the aqueous-carbonic fluids is thus reasonable. The participation of metamorphic fluids is consistent with the association of mineralization with granite-metasedimentary contacts and, since these fluids may have controlled or influenced the fo_2 of the hydrothermal system, metamorphic fluids may have played a significant role in the transport and precipitation of cassiterite. It is important to note that the term 'metamorphic fluid'

Fig. 2 Homogenization temperature versus salinity of fluid inclusions from Nong Sua (modified after Linnen and Williams-Jones 1994). From right to left these represent: aqueous two-phase (*L-V*) inclusions (*black*); aqueous halite-bearing (*L-V-H*) inclusions (*plus signs*); aqueous two-phase (*L-V*) inclusions with CO_2 but without a visible carbonic phase (*dark grey*); aqueous-carbonic inclusions (*L-L-V*) that homogenize to the liquid phase (*light grey*) and; aqueous-carbonic inclusions (*V-L-L*) that homogenize to the vapour phase (*white*). The minerals which host the fluid inclusions are represented by *triangles* for garnet, *plus signs* and *circles* for quartz, *diamonds* for cassiterite and *squares* for tourmaline



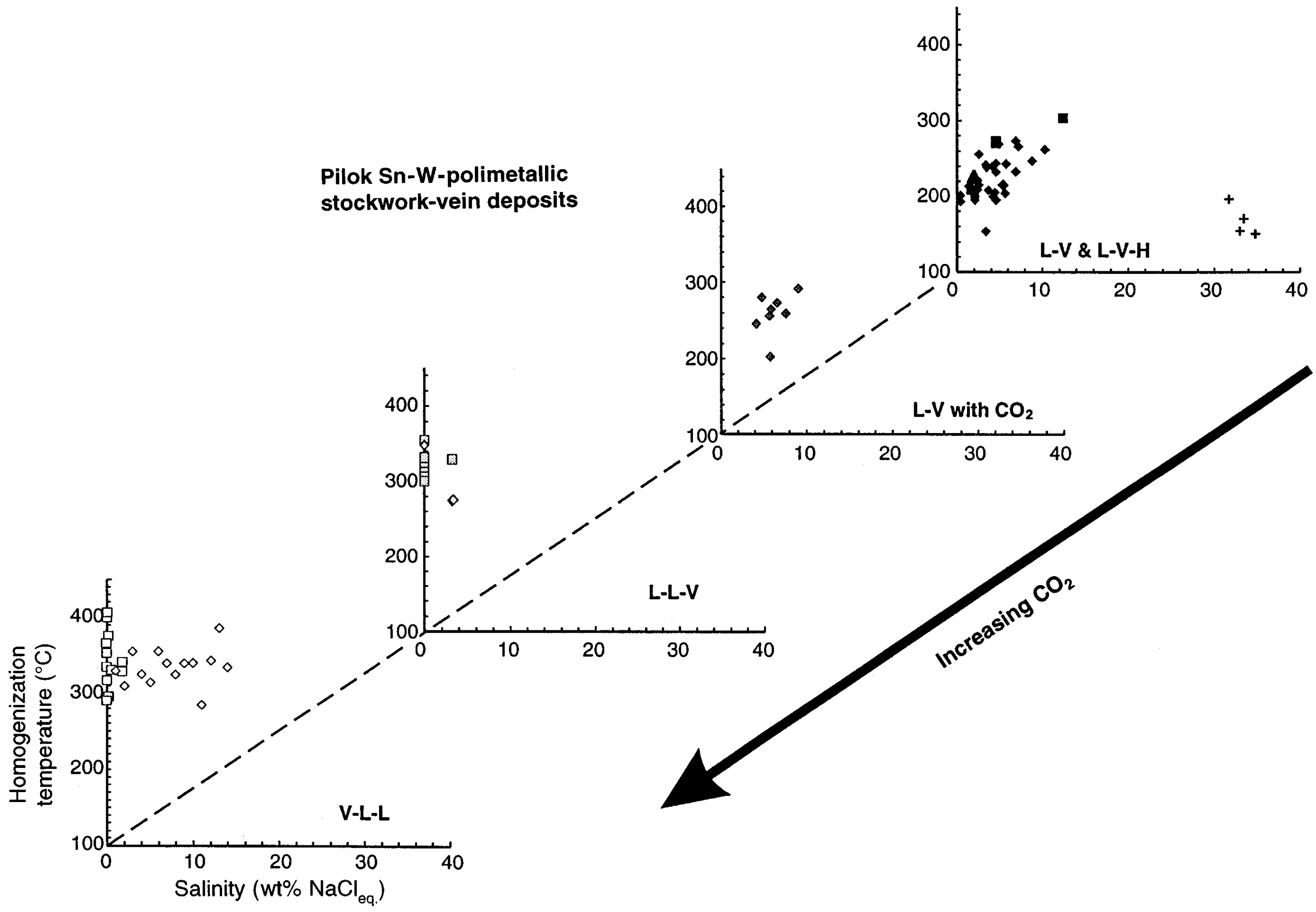


Fig. 3 Homogenization temperature versus salinity of fluid inclusions from Pilok (modified after Linnen and Williams-Jones 1995). From *right* to *left* these represent: aqueous two-phase (*L-V*) inclusions (*black*); aqueous halite-bearing (*L-V-H*) inclusions (*plus signs*); aqueous two-phase (*L-V*) inclusions with CO_2 but without a visible carbonic phase (*dark grey*); aqueous-carbonic inclusions (*L-L-V*) that homogenize to the liquid phase (*light grey*) and; aqueous-carbonic inclusions (*V-L-L*) that homogenize to the vapour phase (*white*). The minerals which host the fluid inclusions are represented by *plus signs* and *diamonds* for quartz, *squares* for cassiterite and beryl and *triangles* for fluorite

is used here to describe a fluid that has equilibrated or partially equilibrated with metamorphic rocks. This does not necessarily imply that the fluid components were produced from prograde metamorphic reactions.

The fluid evolution at Nong Sua involved three-component mixing. This is shown by Fig. 2, where similar homogenization temperature versus salinity relationships are observed for fluid inclusions with increasing amounts of CO_2: aqueous (CO_2-free), aqueous with trace amounts of CO_2 (implied from double freezing behaviour and clathrate formation), aqueous-carbonic (homogenize to liquid) and aqueous-carbonic (homogenize to vapour). The oldest fluid inclusions are primary L-V inclusions in magmatic garnet. The garnet is magmatic and the inclusions are primary. Therefore, these low salinity and CO_2-free inclusions represent trapped orthomagmatic fluids. These fluids mixed with

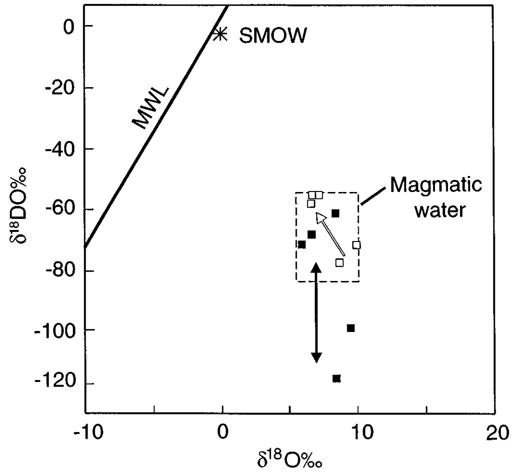


Fig. 4 Fluid compositions calculated from stable isotope analyses of pegmatite-aplite and greisen muscovite from Nong Sua and Pilok, modified after Linnen and Williams-Jones (1994, 1995). The *open white squares* represent fluids from Nong Sua and the *white arrow* the interpreted magmatic-metamorphic fluid mixing trend. The *black squares* represent fluids from Pilok and the *black arrow* the interpreted magmatic-metamorphic fluid mixing trend. In the latter case the low δD values reflect interaction with organic material in the metasediments

metamorphic aqueous-carbonic fluids and moderately saline aqueous fluids (up to 22 wt.% $NaCl_{eq.}$ with significant concentrations of Ca and other bivalent cations). The moderately saline aqueous fluids occur as pseudosecondary inclusions in cassiterite, post-dating primary low salinity inclusions. They are interpreted to be of formational or metamorphic origin because progressive degassing of a magma at the *P–T* conditions of Nong Sua should not result in an increase of salinity (see later). The high Ca concentration in these fluids also suggests that they are not magmatic in origin and the enrichment in Ca in these fluids may correspond with late Ca enrichment in tourmaline and late scheelite replacement of wolframite (Linnen and Williams-Jones 1993). In addition, the evolution from magmatic to hydrothermal muscovite at Nong Sua is marked by an increase in octahedral Fe + Mg + Mn contents, a decrease in the $\delta^{18}O$ values, and an increase (less negative) of δD values (Fig. 4). This is consistent with cooling and mixing of magmatic with isotopically lighter fluids (formational or metamorphic). Late halite-saturated fluid inclusions (33 to 36 wt.% $NaCl_{eq.}$) occur only along secondary planes in quartz. They do not lie along the mixing trends in Fig. 2 and thus likely reflect a separate, distinct, formational or metamorphic fluid.

There is also evidence of mixing between aqueous and aqueous-carbonic fluids during the early fluid evolution at Pilok (Fig. 3). Halite-bearing fluid inclusions (32 to 35 wt.% $NaCl_{eq.}$ with significant bivalent cations) are also secondary at this deposit, but are rare and high salinity fluids were not involved with magmatic-metamorphic fluid mixing (Fig. 3) similar to Nong Sua. The only major difference between the fluid evolution of Nong Sua and Pilok is the influx of late, low temperature-low salinity fluids of probable meteoric origin at the latter deposit. The lack of these late fluids, the lack of pervasive alteration and the restricted range of isotopic compositions at Nong Sua, implies a relatively closed system. The pervasive alteration and the variability of $\delta^{18}O$ and δD values at Pilok (Fig. 4) are indicative of a more open system. Lehmann and Mahawat (1989) and Linnen and Williams-Jones (1995) both interpreted that the mineralization at Pilok was dominantly as a result of remobilization of tin and tungsten from granite.

The fluid inclusion data that are available for other Sn-W deposits in the western Thai belt are summarized in Table 3. It is significant that most of these deposits contain aqueous-carbonic as well as aqueous fluid inclusions. The majority of these deposits are also hosted, at least in part, by the Kaeng Krachan Group, or in the south, the stratigraphically equivalent Phuket Group. The most likely source for the aqueous-carbonic fluid inclusions is thus metamorphic, similar to the aqueous-carbonic inclusions at Nong Sua and Pilok. Minor hypersaline fluid inclusions are present at some deposits but, like Nong Sua and Pilok, they are secondary, apparently are not related to mineralization, and are probably trapped formational or metamorphic fluids.

The only deposit studied to date that has significantly different fluid inclusions is Takua Pit Thong. This deposit is also exceptional in that it is fluorine-rich, and the granites intruded into a calc-silicate metamorphic

Table 3 Fluid inclusions in other western Thai deposits

Deposit	Aqueous inclusions	Aqueous-carbonic inclusions	Reference
Mawchit (Burma), vein-hosted	T_h 120° to 270°C, 3.8 to 8.7 wt.% NaCl equivalent	None reported	Zaw and Thet (1983)
Tavoy Township (Burma), vein-hosted	T_h 140° to 270°C, 1.8 to 5.0 wt.% NaCl equivalent	None reported	Zaw (1984)
Vistae pegmatite Tap Sakae	T_h 288° to 423°C, 2.5 to 12 wt.% NaCl equivalent	None reported	Charusiri (1989)
Tor Soong pegmatite Phuket	T_h 265° to 325°C, 0 to 3 wt.% NaCl equivalent	None reported	Charusiri (1989)
Khao Kow disseminated Phuket	T_h 278° to 294°C, 0.6 to 3 wt.% NaCl equivalent	Present but not analyzed	Charusiri (1989)
Takua Pit Thong sulphide replacement	T_h 130° to 490°C, 0 to 26 wt.% NaCl eq., primary L-V inclusions in cassiterite more restricted T_h 400° to 490°C, 22 to 26 wt.% NaCl eq., some L-V-H inclusions 30 to 38 wt.% NaCl equivalent	Liquid-rich and vapour-rich, wide range in X_{CH4} 0.0 to 1.0	Linnen and Williams-Jones (1991), Linnen (1992)
Som Porn, stockwork & disseminated	L-V and L-V-H reported, none analyzed	Vapour-rich reported, none analyzed	Linnen (1992)
Chat Chai stockwork	L-V and L reported, none analyzed	Liquid-rich and vapour-rich reported, none analyzed	Linnen (1992)
Ban Tha Lao Hub Kapong	L-V and L reported, none analyzed	Liquid-rich and vapour-rich reported, none analyzed	Linnen (1992)
Thai Prasit pegmatite Tap Sakae	L-V reported, none analyzed	Vapour-rich reported, none analyzed	Linnen (1992)

sequence, although some granite is also in contact with carbonaceous pelite (Suvunsavate 1986; Linnen 1992). Primary two-phase (L-V) aqueous fluid inclusions in cassiterite at Takua Pit Thong homogenize to liquid at 400 °C to 490 °C, are 22 to 26 wt.% $NaCl_{eq.}$, and contain high concentrations of Fe, Mn, and Ca chlorides (Linnen and Williams-Jones 1991; Linnen 1992). These inclusions may represent trapped external fluids that underwent exchange with granite (to account for the Fe and Mn) or alternatively, they may represent magmatic fluids with moderately high salinities. The aqueous-carbonic fluid inclusions at Takua Pit Thong also contrast with those present elsewhere in western Thailand by having a much greater range of compositions. Some aqueous-carbonic inclusions have $CH_4/(CH_4+CO_2)$ values of >0.7. This may be related to the fact that the hosts are calc-silicate hornfels and marble at Takua Pit Thong, whereas the other deposits are hosted by pelitic rocks. However, the organic/reduced carbon contents of the calc-silicate hornfels and their low grade regional metamorphic equivalent have not yet been determined. A feature that Takua Pit Thong has in common with some other deposits, including Pilok, is the presence of late, low temperature-low salinity fluids of probable meteoric origin.

Depth of emplacement

Cassiterite-bearing pegmatites occur both north and south of the Ranong fault, which suggests a moderately deep level of emplacement for the entire western Thai belt. The grade of contact metamorphism in western Thailand decreases with distance from the plutons, typically to chlorite-muscovite grade, e.g. at Pilok (Linnen and Williams-Jones 1995), but the grade continues to decrease with distance to sub-greenschist grade at the Kang Krachan dam (Linnen unpublished data) and Phuket (Putthapiban 1984). A contact metamorphic assemblage of biotite-andalusite-cordierite, and locally sillimanite, is reported for Phuket (Garson et al. 1975), indicating pressures of less than 3.8 kbar. It is not stated whether or not the muscovite and chlorite in these rocks also constitute part of this assemblage, but if so, the pressure of contact metamorphism is restricted to have been approximately 3 kbar (van Bosse and Williams-Jones 1988). It should be noted also that since the study of Garson et al. (1975) no other author has reported sillimanite in the contact metamorphism assemblage at Phuket, therefore the latter pressure estimate is tenuous.

The maximum pressure for the emplacement of the Nong Sua pegmatite, based on fluid inclusion isochores, is ~3.8 kbar corresponding to a depth of <12–14 km (Linnen and Williams-Jones 1994). Lithostatic pressure at Pilok is constrained by the occurrence of two alkali feldspars in the leucocratic granite (>1.5 kbar) and from andalusite in the contact metamorphic aureole (<3.8 kbar). Fluid pressures based on fluid inclusion data range from 0.5 to 2.5 kbar (Linnen and Williams-Jones 1995). The Pilok granites thus were probably emplaced at a somewhat more shallow level than at Nong Sua, i.e. at a depth of ≤ 10 km.

At Takua Pit Thong primary liquid-vapour fluid inclusions in cassiterite have final melting temperatures of −23 °C and homogenize to liquid at ~460 °C (Linnen and Williams-Jones 1991; Linnen 1992). SEM analyses of decrepitates indicate that these inclusions are complex Fe-Ca-Mn-Na-K-Cl fluids, but if modelled in the system H_2O-$CaCl_2$ salinities are 22 wt.% $CaCl_{2eq}$. a homogenization pressure of 470 bar can be estimated using the equations of Zhang and Frantz (1987). Primary vapour inclusions (CO_2-CH_4, without visible H_2O) are observed 'coexisting' with the saline aqueous inclusions in the same cassiterite grains, thus 470 bar is potentially the trapping pressure. However, the lack of water (and Cl) in the vapour inclusions argues against a boiling system (although depending on the inclusion size, ~10 mol% H_2O can be present in an aqueous-carbonic fluid inclusion without the water phase being visible). It is more likely that the carbonic inclusions were produced by volatilization of organic material in the metasediments, and that even though this constitutes two fluid phases, the aqueous and carbonic fluids were never in equilibrium and consequently the homogenization pressure is not the trapping pressure. The maximum trapping pressure can be obtained by extrapolating the 22 wt% $CaCl_2$ isochore to the granite minimum solidus, which in this case intersects at ~690 °C and 2300 bar. The cassiterite that hosts these inclusions is post-magmatic, associated with biotite alteration that overprints skarn formation and calc-silicate alteration, hence the trapping temperature must be less than the granite solidus temperature. If a trapping temperature of 600 °C is assumed, a fluid pressure of ~1500 bar is inferred. The depth of emplacement at Takua Pit Thong thus is similar to that at Pilok, but may be slightly shallower.

Pegmatite, vein and greisen deposits are present over the length of the western Thai belt, but the relative importance of the deposit types varies amongst mining camps. This suggests that the deposits were emplaced at roughly the same level, i.e. within a few vertical kilometres of each other. All the major mining camps lie close to major strike-slip faults (Fig. 1), hence post-mineralization differential uplift may have exposed different crustal levels. By contrast, it is also possible that different granites, particularly F-rich granites, were emplaced at slightly different crustal levels.

Comparison of the western Thai belt to other Sn-W provinces

Table 4 summarizes the principal characteristics of many of the major Sn-W provinces around the world. Although pressure or depth of emplacement is generally difficult to estimate, it is apparent from Table 4 that there is a strong correlation between the depth of emplacement and the composition of the observed fluid

Table 4 Characteristics of other Sn-W (W-Sn) provinces

Region	Aqueous-carbonic inclusions	Aqueous inclusions	Depth or pressure	Other features	Selected references
Bolivia	Rare or absent	Early high *T* hypersaline, later low *T*-low salinity fluids, some boiling	Associated volcanic rock *P* estimated <500 bar	B-rich	Kelly and Turneaure (1970), Turneaure (1971), Grant et al. (1980), Sugaki et al. (1988)
Erzgebire Germany	Rare or absent	Early hypersaline, later low *T*-low salinity fluids	Associated volcanic rock *P* estimated <500 to 2000 bar	F-rich and Li-rich	Thomas (1982), Tischendorf and Föster (1990)
Herberton, Australia	Rare or absent	Early high *T* hypersaline, later low *T*-low salinity	Associated volcanic rock *P* estimated <1000 bar	F-rich	Witt (1988), Charoy and Pollard (1989)
Ardlethan, Australia	Common, low to moderate salinity, commonly secondary moderate *T*	Range from high *T*-high salinity to low *T*-low salinity	Associated volcanic rock *P* estimated 500–1200 bar		Ren et al. (1995)
New England, Australia	Rare or absent	Early high *T* hypersaline, later low *T*-low salinity	Associated volcanic rock *P* estimated <700 bar	F-rich	Eadington (1983), Sun and Eadington (1987)
Cornwall England	Rare or absent	Early high *T* moderate salinity, also early hypersaline?, late low *T* -low salinity, some late Ca-Mg-rich fluids	No associated volcanic rock, depth estimated 3–6 km	Some deposits B-rich, other deposits Li-F-rich	Jackson et al. (1989), Rankin and Alderton (1983), Shepherd et al. (1985), Bottrell and Yardley (1988), Smith et al. (1996), Williamson et al. (1997)
Western Tasmania	Generally rare, abundant at some deposits, H_2O-CO_2 some fluid immiscibility	Early high T-salinity at some deposits, moderate *T*-moderate salinity at some deposits, some boiling	No associated volcanic rocks but intrusions are high-level, *P* estimated 200–500 to <2000 bar	Generally F-rich	Collins (1981), Kwak and Askins (1981), Patterson et al. (1981), Solomon (1981), Wright and Kwak (1989), Bajwah et al. (1995), Walshe et al. (1996)
French Hercynian	Early, low salinity H_2O-CO_2-CH_4-N_2, moderate *T*	Later low salinity fluids	No associated volcanic rock, fluid *P* estimated ~700 bar	B-rich	Bril (1982), Bril and Ramboz (1982), Ramboz et al. (1985), Dubessy et al. (1987)
Spanish- Portugese Hercynian	Early, low salinity H_2O-CO_2-CH_4-N_2-H_2S, moderate to high *T*	Later low salinity fluids, some late Ca-Mg-rich fluids	No associated volcanic rock, fluid *P* estimated 100–3250 bar	Some deposits are Li- or B-rich	Kelly and Rye (1979), Bussink et al. (1984), Noronha et al. (1992), Campbell et al. (1988), Mangas and Arribas (1987, 1991)
Malaysia Main Range	Early carbonic CO_2, moderate *T*	Later low salinity fluids, rare hypersaline fluids	No associated volcanic rock, fluid *P* estimated ~1 kbar	Some B-rich	Schwartz and Askury (1989)
Indonesia Main Range, Eastern Province	Some early CO_2-CH_4, moderate *T*, or absent	Some later low to moderately saline, or only low to moderately salinity	No associated volcanic rocks, fluid *P* estimated 700–2000 bar	Some F-rich	Schwartz and Surjono (1990, 1991)

inclusions. Where tin deposits are associated with high-level intrusions, indicated by porphyritic textures and the presence of coeval volcanic rocks, cassiterite is invariably associated with, or was preceded by, high temperature hypersaline fluids e.g. Bolivia (Kelly and Turneaure 1970), Erzgebirge in Germany (Thomas 1983), and in Australia, New England (Eadington 1983) and Herberton-Mount Garnet (Witt 1988). Boiling is interpreted to have occurred in many of the sub-volcanic systems, e.g. Bolivia (Kelly and Turneaure 1970) but like porphyry copper systems, it may also be possible that hypersaline fluids were produced without fluid immiscibility (Webster 1997).

By contrast, in deeper plutonic belts coeval volcanism is lacking and typically at least some mineralized pegmatites are present. The granites in these belts can have equigranular textures, but mineralized intrusions typically display textures indicative of a magmatic or hydrothermal overprint (e.g. Main Range granites, Pitfield et al. 1990). Some examples of deeper plutonic belts are the Central Iberian Zone in Spain (Mangas and Arribas 1987) and Portugal (Gouanvic and Gagny 1987), the Main Range in Malaysia (Cobbing et al. 1992) and western Thai granite belt (this study). The Sn-W deposits in plutonic belts are commonly dominated by aqueous-carbonic and/or low to moderately saline aqueous fluids, e.g., the Portuguese, French and Spanish Hercynian (Kelly and Rye 1979; Bril 1982; Mangas and Arribas 1987), Main Range of Malaysia (Schwartz and Askury 1989) and western Thailand deposits (this study). Some of the deeper plutonic belts such as the French Hercynian contain late-stage high level intrusions, but the fluids associated with these late intrusions are similar to other high level systems, e.g. hypersaline fluid inclusions are present at the Beauvoir granite (Aïssa et al. 1990). There are also exceptional occurrences of mineralized pegmatites associated with high-level intrusions, e.g. Ehrenfriedersdorf (Webster et al. 1997).

It is interesting that the deposits in Cornwall and western Tasmania have some of the fluid inclusion characteristics of both sub-volcanic and plutonic environments (Table 4). In both cases the depth of emplacement is interpreted to be relatively shallow, but coeval volcanism is lacking. These regions therefore seem to be intermediate between sub-volcanic and plutonic environments both in terms of depth of emplacement and fluid composition. Deposits associated with Main Range granites in Indonesia may have also formed in an intermediate level. Schwartz and Surjono (1991) estimate that contact metamorphism at the Pemali deposit occurred at a pressure less than 2 kbar. The same authors report that the fluid inclusions at this deposit have low to moderate salinities and contain only trace amounts of CO_2. By contrast abundant aqueous-carbonic fluid inclusions are observed at the Tikus Sn-W deposit. However, this deposit is associated with an Eastern Province granite, and it may have been emplaced at a slightly deeper level since the minimum pressure estimates from fluid inclusions range from 0.7 to 2 kbar (Schwartz and Surjono 1990).

Experimental constraints on tin mineralization

Experimental studies on the solubility of tin in granitic melts and aqueous fluids place constraints on the conditions at which magmatic, orthomagmatic-hydrothermal or remobilization processes can result in the formation of cassiterite mineralization. The solubility of cassiterite in water-saturated granitic melts at 850 °C and 2 kbar was determined by Linnen et al. (1995, 1996), using a diffusion profile technique in order to avoid the problem of tin forming alloys with the capsules material which hampered previous studies. SnO_2 solubility is highly dependent on fo_2. At these conditions, and at an fo_2 corresponding to the fayalite-magnetite-quartz buffer, approximately 1.0 wt.% SnO_2 is required for cassiterite saturation in metaluminous to peraluminous granitic melts. With increasing fo_2 cassiterite solubility decreases, and at a log fo_2 of Ni-NiO + 0.5 saturation occurs at only 400–700 ppm SnO_2. A late-stage oxidation event thus can trigger cassiterite crystallization in a highly evolved granitic melt. The latter values are consistent with the tin contents in the late-stage granitic melts at Nong Sua (700 ppm Sn) estimated by Linnen et al. (1992), and are similar to the analyses of melt inclusions (average 800 ppm Sn) from the Erzgebirge by Webster et al. (1997).

The crystallization of magmatic cassiterite (or a different tin phase such as wodginite) from a granitic melt is inevitable if the bulk distribution coefficient of tin between crystals + vapour and melt is <1. Typically only small amounts of Fe-Ti oxide phases crystallize from late-stage peraluminous granitic melts, particularly for the ilmenite series of granites. It follows that if the fluid-melt distribution coefficient is <1, the bulk distribution coefficient is also likely to be <1 (see Kovalenko et al. 1988 for mineral-melt partition coefficients from natural occurrences). It is difficult to determine experimentally vapour-melt partition coefficients of tin because of the alloying problem. Nevertheless, it appears that tin is partitioned in favour of the melt when the chlorinity of the coexisting aqueous fluid is less than approximately 1 M, at least for fo_2 conditions near that of the Ni-NiO buffer (Durasova et al. 1986; Taylor 1988; Keppler and Wyllie 1991). Consequently in systems where low salinity aqueous fluids exsolve from a granitic melt, magmatic cassiterite is possible but orthomagmatic-hydrothermal mineralization is less likely (but not impossible, note that orthomagmatic-hydrothermal mineralization is present at Nong Sua). If on the other hand a hypersaline fluid coexists with melt, tin will be strongly partitioned into the aqueous fluid phase and orthomagmatic-hydrothermal mineralization is more probable. In the latter case magmatic cassiterite is unlikely to be preserved if it crystallized, unless the fluid to rock ratio is low, e.g. at the Beauvoir granite (Fouillac and

Rossi 1991; Raimbault et al. 1995). By contrast, during the fractionation of I-type and magnetite series granites, tin is removed from the melt because the greater abundance of oxide phase crystallization. In addition, a greater proportion of the tin in I-type relative to S-type granitic melts is in the 4^+ valence, due to the higher oxidation state. This should increase the substitution for Ti^{4+} in titanite and magnetite, i.e., increases the compatibility of tin in I-type melts (see also the discussion in Lehmann 1990).

The solubility of tin in fluids coexisting with melt depends on fluid composition, hence it is instructive to examine some of the salient points of the solubility of H_2O, CO_2 and Cl in granitic melts. The experimental data on the solubility of water in granitic melts have recently been summarized by Johannes and Holtz (1996). At pressures less than approximately 3.5 kbar (where water saturation occurs at roughly 8 wt.% H_2O) there is a negative dependence on H_2O solubility with temperature. However, even at low pressure, e.g. 500 bar, at least 2 wt.% H_2O is required for a granitic melt to be saturated with water. By contrast, carbon dioxide solubilities in granitic melts are much lower. At 500 bar the maximum CO_2 solubility is approximately 600 ppm and even at 5 kbar, CO_2 saturation occurs at only a few thousand ppm CO_2 (Blank and Brooker 1994). Thus for an H_2O-CO_2 fluid coexisting with a granitic melt, CO_2 is strongly partitioned in favour of the fluid. The partitioning of Cl is complicated by the fact that it is dependent not only on P and T, but also melt composition and Cl concentration (e.g. Webster 1992) and the presence of CO_2 (Webster and Holloway 1988). The oldest fluid inclusions from Thailand are CO_2-free. Thus, in light of the experimental data there is no basis to propose a magmatic aqueous-carbonic fluid, although that is not to say that none of the CO_2 in the fluid inclusions from western Thailand is magmatic in origin. An additional consideration is that external aqueous-carbonic fluids could have infiltrated into the granites at a relatively early hydrothermal stage, potentially even before the melts were completely solidified. If external fluids did infiltrate magma (melt + crystals) they did not originate by vapour saturation of melt and are not considered in this study to be magmatic fluids (although such fluids may have acquired a magmatic signature). In summary, based on the experimental and geological data it is very likely that the majority of the CO_2 and CH_4 in aqueous-carbonic fluid inclusions from western Thailand are 'metamorphic', where metamorphic fluids are broadly defined as fluids that originate either by prograde metamorphic reactions or by fluid interaction with metamorphic rocks.

Cassiterite solubility in aqueous fluids is largely controlled by temperature, pH and fo_2 (Eugster 1986; Wilson and Eugster 1990). This is illustrated by the reaction:

$$SnO_2 + 2H^+ + Cl^- = SnCl^+ + H_2O + \tfrac{1}{2}O_2$$

Several authors, including Eugster (1986), Wilson and Eugster (1990), Heinrich (1990) and Halter et al. (1996) have proposed increasing pH (acid neutralization) as an effective mechanism of cassiterite deposition. It is apparent from Eugster (1986) and Heinrich (1990) that an order of magnitude change in pH has a larger effect on tin solubility than an order of magnitude change of fo_2. Nevertheless for some cases, such as at Pilok, cassiterite is disseminated in highly altered granite, muscovite greisen (fluid dominated), and is not in the outer alteration selvage (rock dominated) or associated with topaz, where acid neutralization should occur. This argues against an increase in pH as a depositional mechanism and instead, increasing fo_2 or decreasing temperature were more important at this deposit (Linnen and Williams-Jones 1995). Variable CO_2/CH_4 ratios are preserved in fluid inclusions at both Pilok and Nong Sua, reflecting varying fo_2 conditions, which is probably related to volatilization of organic material in the surrounding metasediments. In fact the presence of organic-rich metasedimentary hosts for the granites may be an important factor controlling the formation of tin deposits (Zentilli et al. 1995), providing reduced fluids capable of remobilizing tin.

Discussion

The contrasting fluid evolution of sub-volcanic and deeper plutonic systems may be related to the effect of pressure on the fluid-melt partitioning of chlorine and/or the effect of pressure on fluid immiscibility in the system H_2O-$NaCl \pm CO_2$. At pressures greater than approximately 2 kbar the first aliquot of water exsolved from a melt has the highest salinity and the salinity decreases with successive aliquots (Candela 1989). At pressures less than approximately 1 kbar chlorine exhibits the opposite behaviour; the first aliquot has a relatively low salinity. Successive aliquots of water contain increasingly higher salt concentrations. This partitioning behaviour of chlorine has been used to explain the association of high temperature hypersaline fluid inclusions with high-level porphyry copper deposits (Candela 1989; Cline and Bodnar 1991). This also seems to be a reasonable explanation for the association of high temperature hypersaline fluid inclusions with sub-volcanic tin deposits. However, as pointed out by Candela and Piccoli (1995), the "instantaneous aliquot" model of Candela (1989) and Cline and Bodnar (1991) may not be an accurate model of the fluid evolution in a crystallizing pluton and different fluid batches may mix to produce an "average magmatic fluid". In addition, the experimental data of Webster (1997) indicate that brines as a single fluid phase may exsolve directly from granitic melts with Cl-rich compositions. The phase equilibria and fluid–melt partitioning of H_2O-$NaCl$-CO_2 is not completely understood, but it is clear that aqueous fluid immiscibility (boiling) can produce Cl-rich brines and

Cl-poor vapour ($\pm CO_2$) at low pressure, e.g. 500 bar. By contrast, the conditions of fluid immiscibility become more restricted at higher pressure, e.g. 2000 bar, and two aqueous-carbonic fluids are present only for high Cl concentrations (Bowers and Helgeson 1983; Webster 1997). Whether hypersaline fluids originate by Rayleigh fractionation, fluid immiscibility or a high Cl/H_2O ratio in the source melt, hypersaline fluids are nonetheless a common feature of subvolcanic systems. Because tin is easily transported as a chlorine complex, these systems have the potential to form orthomagmatic-hydrothermal mineralization. This idea is supported by the fact that the tin concentrations in high salinity orthomagmatic fluid inclusions trapped in topaz from the Mole granite, Australia range up to several thousand ppm (Rankin et al. 1992).

In the deeper plutonic systems the pressure at which vapour saturation occurred may also explain the low to moderate salinity of the aqueous fluid inclusions, as the late-magmatic fluids in these systems may have progressively lower salinities (Candela 1989). The common occurrence of aqueous-carbonic inclusions indicates that either the magmatic fluid is aqueous-carbonic or that metamorphic fluids were involved. A metamorphic origin for aqueous-carbonic fluids has been proposed for several of the plutonic systems including this study, and in one case Aberfoyle, Tasmania, the presence of hydrocarbons in fluid inclusions strongly supports a metasedimentary source (Hoffmann et al. 1988). It seems reasonable to propose that, in many plutonic systems, the aqueous-carbonic fluid inclusions are dominantly metamorphic in origin (or fluids that underwent exchange with metamorphic rocks). Only the earliest magmatic fluids should contain CO_2, since with vapour saturation the CO_2 content of the melt will rapidly be depleted.

A relationship between depth of emplacement and fluid composition appears to exist for other granite-related deposits. Molybdenum deposits emplaced at high levels, e.g. Henderson, Colorado, contain abundant hypersaline fluid inclusions (White et al. 1981), whereas those emplaced at a deeper level, e.g. Trout Lake, British Columbia, are dominated by aqueous-carbonic fluid inclusions (Linnen and Williams-Jones 1990). It is possible that the aqueous-carbonic fluid inclusions in the deeper molybdenum systems are also dominantly metamorphic in origin. The similarities of fluid compositions in deeper Sn-W and Mo plutonic environments, associated with ilmenite- and magnetite-series granites, respectively, argues against a magmatic origin for the aqueous-carbonic fluid inclusions in the deeper plutonic environment, i.e. if aqueous-carbonic fluids are magmatic, those from S-type granites should have higher CO_2/H_2O and CH_4/CO_2 ratios compared to I-type granites; the fact that they do not suggests that the aqueous-carbonic fluids are not magmatic.

In conclusion, the depth of emplacement is an important factor in controlling mineralization processes. Magmatic, orthomagmatic-hydrothermal and remobilization processes can all result in tin mineralization at both shallow and deep levels of emplacement, but the relative importance of these different processes changes as a function of pressure. There is a greater tendency for cassiterite-bearing pegmatites to form at depth. Low chlorine contents are characteristic of deeper granitic intrusions, consequently tin is partitioned in favour of granitic melts and magmatic cassiterite can crystallize in highly evolved systems such as pegmatites. Vein and greisen styles of mineralization in deeper plutonic systems tend to form by remobilization, because tin is not strongly partitioned into magmatic aqueous magmatic fluids (fluids exsolved from granitic melts) due to the low chlorine contents of the fluids. The presence of carbon-bearing metasediments may be a key component to deeper mineralized systems, since the metasediments directly or indirectly (by prograde metamorphism or by fluid-rock interaction) provide reduced fluids capable of transporting tin.

By contrast, high level sub-volcanic systems are characterized by hypersaline fluids. Tin is, therefore, partitioned from the melt into the aqueous phase in these systems, which can result in the formation of orthomagmatic-hydrothermal mineralization. Similar relationships between fluid composition and pressure appear to be true for other granite-related deposits such as molybdenum stockwork and porphyry deposits, thus the depth of emplacement is important in controlling granite-related mineralization in general.

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