Methane (CH\textsubscript{4})-bearing fluid inclusions in the Myanmar jadeitite

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A combined hydrogen-carbon-isotope and microthermometric study has been carried out on CH\textsubscript{4}-bearing fluid inclusions in high-pressure jadeites from the famous jadeite tract Myanmar. Two types of fluid inclusions were found in jadeites, large H\textsubscript{2}O-rich and CH\textsubscript{4}-poor inclusions and small H\textsubscript{2}O-poor and CH\textsubscript{4}-rich inclusions, thus indicating a possible entrapment of CH\textsubscript{4}-H\textsubscript{2}O fluids under unmixing conditions. Microthermometric results yield lower temperature limits for the entrapment of these fluid inclusions of ca. 300 to 400 °C. The bulk composition of the fluid inclusions is mostly H\textsubscript{2}O (87 to 94 mol.% H\textsubscript{2}O) and the isotopic composition of methane and water in the inclusions is characterized by δ\textsubscript{13}C(CH\textsubscript{4}) values ranging from –30.1 to –25.5‰, and δ\textsubscript{D}(H\textsubscript{2}O) values ranging from –56.3 to –49.8‰. The stable isotope data would be indicative of an abiogenic mechanism of CH\textsubscript{4} formation; the occurrence of the jadeite veins in this paleo-subduction zone thus most likely point to the formation of these CH\textsubscript{4}-bearing fluid inclusions by abiogenic thermal maturation of subducted organic carbon. These data not only provide evidence for cycling of organic carbon in paleo-subduction zones but also show that CH\textsubscript{4} not only occurs as shallow CH\textsubscript{4}-rich plumes in accretionary prisms of recent subduction zones but also occurs in deeper portions of at least the upper 20 km of paleo-subduction zones.

Keywords: fluid inclusions, jadeitite, methanogenesis, methane (CH\textsubscript{4}), Myanmar

INTRODUCTION

Jadeite commonly occurs in high-pressure or ultra-high pressure metamorphic rocks such as jadeite-bearing quartzites and gneisses (Schertl et al., 1991), jadeite-K-feldspar bearing schists (Okay, 1997) and occasionally in jadeites, which are primarily composed of jadeite. Jadeiteites are relatively rare and have been described from a handful of localities such as northwestern Myanmar; Kamuikotan area, Hokkaido, Japan; San Benito County, California; Pay-Yer massif, Polar Urals, Russia; northern near-Balkash region, Kazakhstan; Montauk Fault and Guatemala (Chhibber, 1934; Iwao, 1953; Morkovkino, 1960; Coleman, 1961; Dobresov and Ponomareva, 1965; McBriney et al., 1967; Takajama, 1986; Kobayashi et al., 1987; Harlow, 1994). Jadetite petrogenesis has been interpreted to be either metamorphic or metasomatic (Harlow, 1994; Miyazaki et al., 1998; Radvanec et al., 1998). Jadeiteites commonly occur as lenses, pods and veins or as tectonic blocks within a serpentinitized ultramafic matrix such as dunites and are thought to form under high-P/low-T conditions (Harlow, 1994; Okay, 1997). Due to their often common vein-like texture, which implies a possible metasomatic origin, jadeiteites can be thought of as petrological recorders of the fluid history of a subduction environment (Sorensen and Harlow, 1999, 2001; Harlow and Sorensen, 2001, 2005).

Fluid inclusion studies in jadeiteites therefore provide important constraints on the composition of the metamorphic fluid present during formation of the jadeiteites in deep subduction zone environments. In recent studies, Harlow (1986) and Johnson and Harlow (1999) report aqueous fluid inclusions containing up to 8.7 wt% NaCl in coarse-grained jadeites from the Guatemalean jadeiteites. The authors proposed that the fluid is probably of seawater origin and was entrapped during subduction in the minerals. Fluid inclusion studies on high pressure rocks such as eclogites show that fluids in subduction zones are variably brines with minor CO\textsubscript{2} and N\textsubscript{2} and rarely contain some CH\textsubscript{4} (Sorensen and Barton, 1987; Kastner et al., 1998; Scambelluri and Philippot, 2001). Studies of oceanic rocks and of eclogitized ophiolites indicate that prior to subduction, the slab can be considerably altered by surficial fluids (Scambelluri and Philippot, 2001). Metamorphic processes such as devolatilization produce in the initial stages low-salinity, CO\textsubscript{2}- and CH\textsubscript{4}-
bearing fluid inclusions (Kastner et al., 1998). At higher grades, CH4 seems to disappear and CO2 forms instead as reported from fluid inclusions from eclogite-facies rocks (Xiao et al., 2000; Scambelluri and Philippot, 2001; Yang et al., 2001; Fu et al., 2003a). But recent studies show that ultrahigh pressure (UHP) metamorphic eclogites and associated gneisses from the Dabie-Sulu terranes, despite most of them having carbonic fluid (CO2-rich) inclusions, also contain CH4-rich fluid inclusions coexisting with high-salinity brine inclusions (Fu et al., 2001, 2002, 2003b) and organically derived carbonate in whole-rock and apatite (Zheng et al., 2000, 2003a; Li et al., 2000). CH4 is a common fluid species in hydrothermal systems in the oceanic crust and commonly forms either by reactions involving magmatic CO2 or during serpentinization of olivine and/or other mafic phases involving CO2-rich fluids (Kelley and Früh-Green, 1999). So far there is only indirect evidence for the presence of CH4 in subduction zones in shallow (1–3 km depth) CH4-rich plumes emanating from the accretionary prisms in convergent margins (Craig et al., 1987; Watanabe et al., 1994; Tsunogai et al., 1998). Recent investigations to constrain the retention and loss of volatile elements such as CH4 during subduction showed that fluxes of carbon into subduction zones are larger than returned to the surface, thus indicating that CH4 could also occur in deeper levels of subduction zones (see Sadofsky and Bebout, 2003; Shaw et al., 2003).

CH4-bearing fluid inclusions in jadeite have not been reported before the investigations of Shi et al. (2000) on the jadeitites from the famous Myanmar jadeite Tract and thus could help to put constraints on the flux of CH4 in paleo-subduction zones. In this paper we describe CH4-bearing fluid inclusions in jadeite crystals from the Myanmar jadeitites, their composition including the δD(H2O) and δ13C(CH4) isotopes and the implications for carbon cycling in paleo-subduction zones.

SAMPLE SELECTION AND DESCRIPTION

The outcrops are located in an area in the western part of the Sagaing strike-slip fault belt in the Parkhan (or called Hpakan or Pharkan) city, Kachin state, Myanmar (central location at N25°36.9′, E96°18.6′) determined by GPS measurements, Figs. 1A and B. The famous Burmese (Myanmar) jadeite occurs in this belt. Primary jadeite deposits occur as veins cross-cutting the serpentinitized peridotite bodies called the Pharkan-Tammaw ultramafic body, which is part of the Indo-Burma range of ophiolites, and Chhibber (1934) described these veins as a “dike”. As there is no strong evidence for a magmatic origin, we prefer the term “vein”. The jadeite veins are almost vertical, strike N-S, and have a width of 0.5 m up to 5 meters and a length of about 10–100 meters. According to Chhibber (1934), the boundary to the surrounding serpentinites is marked by a zone that consists of a mixture of chlorite, occasionally with calcite, actinolite, and talc. In contrast, we only observed jadeite veins with complex sodic- and sodic-calcic amphibole boundary zones on both sides (Shi et al., 2003). In a few outcrops, thin veins of albite, commonly less than 5 mm wide, cross-cut the jadeite veins and thus obviously

Fig. 1. (A) Geological overview of the jadeite area located in the west of Sagaing strike-slip faults belt within the collision zone between the Indian Plate and the Yangtze Block, modified after Bender (1983); (B) Geological sketch map of the Myanmar jadeite area, the primary jadeitites occur in ultra-mafic rock bodies, modified after Chhibber (1934).
formed at a later stage. The amphibole boundaries have a width of 1–50 cm and fragments of the amphibole fels can be found within jadeite veins due to later deformation. Within or adjacent to the amphibole boundary zones, jadeite fragments containing kosmochlor or chromian jadeite can often be found as roundish aggregates or blocks wrapped by amphiboles. Outside the ultramafic bodies, high-pressure metamorphic rocks such as phengite-bearing glaucophane schists, and amphibolite-facies rocks such as garnet-bearing amphibolites, diopside-bearing marbles and stilpnomelane-bearing quartzites occur (Shi et al., 2001).

We examined about 150 specimens of jadeities, jadeite-kosmochlor-bearing jadeities, and rocks from the amphibole boundary zones (Shi et al., 2003). Through microscopic observations on thin sections of these samples, about 10 samples of undeformed coarse-grained jadeities were found containing a sufficient number of fluid inclusions.

Polished thin- and thick sections were made for petrographic examination, microthermometry using the heating/cooling stage, and micro-Raman spectroscopy to obtain the chemical composition of individual fluid inclusions. In a second step, rock chips were crushed to obtain jadeite grains which were then prepared for bulk chemical analyses of the fluid inclusions. The fluid inclusions in the jadeite grains were then decrepitated to obtain their bulk composition. The bulk chemical methods performed in this study were ion chromatography to obtain the ionic content of the fluid inclusions and quadrupole mass spectrometry (QMS) to obtain the bulk H2O contents of the fluid inclusions. In addition, six samples were also used for bulk stable isotope measurements of δD and δ13C by gas-source mass spectrometry.

**Analytical Techniques**

The chemical composition of the jadeites was determined by electron microprobe analysis (EMPA) in the electron microprobe laboratory of the Institute of Geology and Geophysics, Chinese Academy of Sciences (IGGCAS). The EMPA data were obtained with a CAMECA CAMEBAX SX51 system with the analytical conditions of 15 kV and 12 nA beam current.

Microthermometric investigations of 0.2 mm thick polished sections were performed with a LINKAM THMS600 heating-cooling stage at JICA lab, IGGCAS, with a heating rate of 1 to 5°C/min and a cooling rate of 0.5 to 3°C/min.

Micro-Raman spectroscopy was performed with a Renishaw-1000 Micro Raman spectrometer with an argon laser beam (λ = 514.5 nm) and analytical conditions of 5 mW on thick sections (0.2 mm) of coarse-grained jadeities. The detection limits of the method for the different gases are 0.5 mol%. An objective lens with a magnification of 50 times was used.

Quadrupole mass spectrometry, which is used to obtain information about additional volatiles in the individual aqueous inclusions (Sasada et al., 1992; Küster and Stöckhert, 1997; Röller et al., 2001), was performed to check whether small amounts of additional carbon-bearing compounds besides CH4 exist. Double-polished thick sections with a size of 2 × 2 × 0.2 mm were prepared and inserted in the extraction tube and heated up to 800°C within 150 minutes in high vacuum. During heating, individual fluid inclusions decrepitated due to internal overpressure which leads to the formation of small pressure pulses which are recorded. The released gases were continuously analyzed with a QMS (RG202 at JICA lab, IGGCAS) in a rapid scanning mode (50 msec/amu) for the following molecular masses: 16(CH4), 18(H2O), 28(CO) and 44(CO2). With this method, the composition of individual inclusions in terms of their volatile components, size and temperature of decrepitation can simultaneously be recorded.

In addition to the QMS measurements, we also used an ion chromatograph (SHIMADZU, HIC-6A) at JICA lab, IGGCAS to measure the bulk cation and anion contents of the fluid inclusions. For this purpose, the selected jadeites were crushed and sieved with a 20–80 mesh. One gram of jadeite grains was picked out from each sample and cleaned with highly distilled water until the conductivity of the water used was the same as pure water. The grains were then dried and heated up to 1000°C to decrepitate all fluid inclusions. Afterwards 5 ml of ultrapure water was added to the decrepitated specimens to dissolve any soluble ions, which was then analyzed with the ion chromatograph.

Water was extracted from the fluid inclusions by induction heating in a vacuum extraction line. The material of the line is Pyrex except for the sample tube and the crucible holder, which are made of silica glass. The samples were put into a Pt crucible and were heated stepwise with an induction furnace. The temperature was increased stepwise from room temperature up to about 1400°C. H2O was cryogenically separated from the other volatile species that were first condensed by liquid-N2 trap connecting to sample tube. H2O was then converted into H2 gas by exposing it to hot uranium at 650°C. H2 was then collected with the Topler pump and sealed in a sample tube for mass spectrometry analysis.

CH4 was extracted from the fluid inclusions using sealed tube combustion. The samples together with copper-oxide threads were put into a Pyrex reaction tube and then sealed in vacuum. The sealed tubes were then heated up to 800°C and held within a period of 20 hours to reach complete conversion of CH4 into CO2 according to the reaction CH4 + 4CuO = 4Cu + 2H2O + CO2. Subsequently, Fluid inclusions in Myanmar jadeite...
CO₂ was then collected in high vacuum and sealed in a sample tube for mass spectrometric analysis. D/H ratios and ¹³C/¹²C were measured with a double collector gas source mass spectrometer (MAT 252 and MAT 251) at IGGCAS, and reported in the δD and δ¹³C notations with the references of VSMOW and VPDB, respectively. Analytical uncertainties are about ±3‰ and ±0.5‰ for δD and δ¹³C, respectively.

PETROGRAPHY AND MINERAL CHEMISTRY OF THE JADEITITES

Most jadeitites are strongly deformed and fine-grained...
Fluid inclusions in Myanmar jadeitite and occasionally domains with coarse-grained, less- or undeformed, jadeite crystals occur (Fig. 2). A few jadeitites reveal a primary vein structure, with large euhedral to subhedral grains. Cathodoluminescence images reveal a rhythmic zoning pattern of the coarse-grained jadeite crystals as shown in Fig. 3A, similar to the observations of Harlow (1994) and Sorensen and Harlow (1998) from the Guatemalan jadeitites. The jadeitites of this study are monomineralic and albite and analcite rarely occur as secondary alteration products. The chemical compositions of jadeite are given in Table 1.

Euhedral jadeites are very pure, with jadeite (XJd) contents of more than 98 mol.%. Backscatter electron (BSE) images of some of the coarse-grained jadeite porphyroblasts reveal a chemical zoning pattern as shown in Fig. 3B with rhythmically increasing Ca and Mg contents (light grey zones in the image) ranging from 0.01 a.p.f.u. to 0.08 a.p.f.u. thus leading to variations in XJd from 0.91 to 0.99 (Table 1).

<table>
<thead>
<tr>
<th>Sample</th>
<th>Dc1</th>
<th>Dc2</th>
<th>Dc3</th>
<th>Df1</th>
<th>Df2</th>
<th>Fc1-4</th>
<th>Fc1-6</th>
<th>Fc1-8</th>
</tr>
</thead>
<tbody>
<tr>
<td>SiO₂</td>
<td>58.50</td>
<td>57.9</td>
<td>59.01</td>
<td>58.4</td>
<td>57.62</td>
<td>57.59</td>
<td>58.36</td>
<td>58.37</td>
</tr>
<tr>
<td>TiO₂</td>
<td>n.d.</td>
<td>n.d.</td>
<td>0.04</td>
<td>n.d.</td>
<td>0.01</td>
<td>n.d.</td>
<td>n.d.</td>
<td>n.d.</td>
</tr>
<tr>
<td>Cr₂O₃</td>
<td>n.d.</td>
<td>n.d.</td>
<td>0.06</td>
<td>0.03</td>
<td>0.01</td>
<td>0.05</td>
<td>0.05</td>
<td>0.02</td>
</tr>
<tr>
<td>Fe₂O₃</td>
<td>n.c.</td>
<td>n.c.</td>
<td>n.c.</td>
<td>n.c.</td>
<td>0.69</td>
<td>n.c.</td>
<td>n.c.</td>
<td>n.c.</td>
</tr>
<tr>
<td>FeO</td>
<td>0.03</td>
<td>n.d.</td>
<td>0.07</td>
<td>0.15</td>
<td>n.d.</td>
<td>0.91</td>
<td>0.44</td>
<td>0.76</td>
</tr>
<tr>
<td>MnO</td>
<td>0.07</td>
<td>0.14</td>
<td>n.d.</td>
<td>0.04</td>
<td>0.11</td>
<td>n.d.</td>
<td>0.04</td>
<td>0.01</td>
</tr>
<tr>
<td>NiO</td>
<td>n.d.</td>
<td>n.d.</td>
<td>0.06</td>
<td>0.13</td>
<td>0.04</td>
<td>0.01</td>
<td>n.d.</td>
<td>0.04</td>
</tr>
<tr>
<td>MgO</td>
<td>0.05</td>
<td>0.05</td>
<td>0.09</td>
<td>0.21</td>
<td>0.04</td>
<td>1.64</td>
<td>1.26</td>
<td>1.60</td>
</tr>
<tr>
<td>CaO</td>
<td>0.05</td>
<td>0.12</td>
<td>0.12</td>
<td>0.26</td>
<td>n.d.</td>
<td>2.05</td>
<td>1.59</td>
<td>1.96</td>
</tr>
<tr>
<td>K₂O</td>
<td>0.02</td>
<td>0.01</td>
<td>n.d.</td>
<td>n.d.</td>
<td>n.d.</td>
<td>0.03</td>
<td>0.01</td>
<td>0.02</td>
</tr>
<tr>
<td>Σ</td>
<td>99.94</td>
<td>99.17</td>
<td>100.64</td>
<td>100.3</td>
<td>99.75</td>
<td>99.72</td>
<td>99.31</td>
<td>99.57</td>
</tr>
</tbody>
</table>

The pyroxene formulae were calculated on the basis of 6 oxygens and 4 cations. Fe³⁺ was calculated from charge balance considerations. The analyses Dc1, Dc2, Dc3 are from euhedral-subhedral coarse-grained jadeites whereas the analyses Df1 and Df2 are from fine-grained jadeites. Fc1-4, Fc1-6 and Fc1-8 are from a zoned jadeite porphyroblast. n.d.: not detected; n.c.: not calculated; XJd = Na/(Na+K+Ca).

Table 2. Microthermometric results of individual fluid inclusions

<table>
<thead>
<tr>
<th>Gas content (vol.%)</th>
<th>Tm (°C)</th>
<th>Th (°C)</th>
</tr>
</thead>
<tbody>
<tr>
<td>20</td>
<td>-4.3</td>
<td>365</td>
</tr>
<tr>
<td>15</td>
<td>-1.5</td>
<td>315</td>
</tr>
<tr>
<td>38</td>
<td>-1.8</td>
<td>318</td>
</tr>
<tr>
<td>28</td>
<td>-3.7</td>
<td>325</td>
</tr>
<tr>
<td>23</td>
<td>-5.5</td>
<td>321</td>
</tr>
<tr>
<td>31</td>
<td>-3.0</td>
<td>330</td>
</tr>
<tr>
<td>20</td>
<td>-3.3</td>
<td>318</td>
</tr>
<tr>
<td>30</td>
<td>-2.1</td>
<td>328</td>
</tr>
<tr>
<td>21</td>
<td>-3.1</td>
<td>378</td>
</tr>
</tbody>
</table>

Tm = melting temperature, Th = homogenization temperature.

**OPTICAL DESCRIPTION OF THE FLUID INCLUSIONS**

Two-phase, gas/liquid fluid inclusions were only found in coarse-grained jadeite crystals. Large fluid inclusions (ca. 10 µm), which are about 10% of all inclusions, con-
tain small visible bubbles (Figs. 4A and B), while the bubbles of the small ones, especial the very small ones (<3 µm), are not visible under the polarization microscope. The fluid inclusions appear to be of primary origin, since they are elongated along the c-axis of the crystals, show a random distribution throughout the jadeite grains, and no occurrence of inclusions along healed fractures can be found. The volumetric contents of gaseous components in the larger fluid inclusions ranges from 15 to 38 vol.% as shown in Table 2.

CHEMICAL ANALYSIS OF INDIVIDUAL FLUID INCLUSIONS

Microthermometry

The investigations were carried out only on fluid inclusion in size large enough to permit accurate measurements. The fluid inclusions were brought to temperatures of ~180°C and then subsequently heated up. The final melting temperatures of fluid inclusions show a bimodal distribution. Some small fluid inclusions show freezing temperatures below ~180°C, indicating CH₄-rich compositions, while most fluid inclusions show melting temperatures ranging from ~1.5 to ~5.5°C, which indicates H₂O-rich fluid inclusions, containing small amounts of dissolved additional components such as salts (e.g., NaCl, KCl, CaCl₂) (Table 2). Homogenization temperatures (L + V = L) of the fluid inclusions range from 315 to 378°C. Based on the homogenization temperatures and the volumetric amount of gaseous components it was possible to calculate the chemical composition of the H₂O-rich fluid inclusions in terms of X(CH₄) and X(H₂O) with the program BULK by Bakker (2003). The data show for the H₂O-rich fluid inclusions calculated X(H₂O) of 0.995 and X(CH₄) of 0.005. Calculations of X(CH₄) and X(H₂O) for the CH₄-rich fluid inclusions was not possible since the CH₄ homogenization temperature (L + V = V) could not be determined due to the small size which prevented clear observation of CH₄ homogenization.

Micro-Raman spectroscopy

About 30 inclusions were measured with a laser beam with a diameter of ca. 20 × 20 µm and representative spectra of the detected components in the fluid inclusions are shown in Figs. 5A, B and C. According to the Raman spectra, H₂O and CH₄ are the only components which were identified in these fluid inclusions, other chemical compounds such as carbon dioxide (CO₂), carbon monoxide (CO) or ethane (C₂H₆) have not been detected. The spectra clearly indicate two groups of fluid inclusions, H₂O-rich/CH₄-poor fluid inclusions (Figs. 5B and C). The CH₄-rich/H₂O-poor fluid inclusions occur only as very small fluid inclusions.

BULK CHEMICAL ANALYSIS OF THE FLUID INCLUSIONS

Quadrupole mass spectroscopy

The composition of individual fluid inclusions in terms of their CH₄, CO and CO₂ contents indicates that the fluid inclusions contain CH₄ as the only carbon-bearing species. The data suggest that decrepitation of fluid inclusions is also a function of size since most of the large fluid inclusions decrepitate within a temperature range of 190–275°C whereas most of the smaller ones decrepitated between 475–550°C.

In order to obtain bulk molar ratios of H₂O versus other gaseous components, the crushed and sieved jadeite grains (about 0.3 g weight of 20–40 mesh grains) were put into a Silex glass tube connected to the QMS, heated up to
suggest that the listed in Table 4. The Raman and QMS measurements ratios (XH$_2$O) ranging from 87 mol.% to 94 mol.% H$_2$O.

The occurrence of reactions among the different gas species. After the hot tube cooled down to room temperature, liquid nitrogen was removed and the total velamen pressure. The decrepitated gases were then condensed in traps cooled by liquid nitrogen to prevent the fluids were possibly entrapped under immiscible conditions (Kelley, 1996). QMS measurements of the bulk chemical composition of the fluid inclusions have shown that the bulk fluids contain 87–94 mol.% H$_2$O. Experimental investigations of CH$_4$-H$_2$O-fluids indicate that at temperatures below 400°C a homogeneous CH$_4$-H$_2$O-fluid will unmix into a CH$_4$- and a H$_2$O-rich fluid, thus putting an upper temperature constraint on the entrapment temperature of the fluid inclusions (Holloway, 1984). The range of the homogenization temperatures of the fluid inclusions of 315–378°C thus yields a lower temperature limits for the formation of these fluid inclusions. The real trapping temperatures are expected to be higher if a pressure correction of the homogenization temperature is applied. Huizenga (2001) shows in a thermodynamic evaluation of the C-O-H system that CO$_2$-poor fluids are stable only at temperatures below 400 to 450°C, depending on the fO$_2$ and whether graphite was present. Calculation of isochores, based on the composition of the fluid inclusions has not been attempted for the following reasons: (1) the lack of microthermometric data of the CH$_4$-rich/ H$_2$O-poor fluid inclusions, and (2) the high probability of density modification during uplift, which leads to considerable pressure underestimations as shown in many studies of fluid inclusions in high pressure rocks (see Philippot and Selverstone, 1992; Selverstone et al., 1992; Sterner and Bodnar, 1989). Despite this discrepancy, fluid inclusions from high-pressure rocks still give insight on the fluid/rock interactions at high pressure conditions (Philippot and Selverstone, 1992; Selverstone et al., 1992). The determination of the pressures of the formation of the jadeites is hampered by the lack of appropriate equilibria and the lack of quartz. Equilibria such as jadeite = albite + nepheline and jadeite + H$_2$O = analcite thus provide only lower pressure limits since the low-P phases are absent in the rocks. For the temperature range of the investigations, application of the analcite breakdown reaction according to the calculations of Harlow (1994) yields lower pressure limits of 0.6 to 0.7 GPa between 250 and 400°C for the formation of the Guatemala jadeites. But this pressure is below the lower stability limit of jadeite + H$_2$O relative to analcite at 0.8 to 1.0 GPa (Boeticher and Wyllie, 1969), even slightly lower than a lower limit of formation of omphacite + quartz of 0.6 to 0.8 GPa at 200 to 300°C in Californian metamorphic rocks.

### Table 3. Bulk ionic contents of the fluid inclusions

<table>
<thead>
<tr>
<th>Sample</th>
<th>F</th>
<th>Cl</th>
<th>SO$_4$</th>
<th>Na$^+$</th>
<th>K$^+$</th>
<th>Mg$^{2+}$</th>
<th>Ca$^{2+}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fc3</td>
<td>0.03</td>
<td>0.38</td>
<td>0.19</td>
<td>3.92</td>
<td>0.16</td>
<td>0.05</td>
<td>0.18</td>
</tr>
<tr>
<td>Fc5</td>
<td>n.d.</td>
<td>0.02</td>
<td>0.02</td>
<td>3.00</td>
<td>0.14</td>
<td>0.05</td>
<td>0.14</td>
</tr>
<tr>
<td>Fr1</td>
<td>0.07</td>
<td>0.18</td>
<td>0.19</td>
<td>3.21</td>
<td>0.20</td>
<td>0.19</td>
<td>0.22</td>
</tr>
<tr>
<td>Fc1</td>
<td>0.02</td>
<td>0.20</td>
<td>0.09</td>
<td>4.40</td>
<td>n.d.</td>
<td>0.07</td>
<td>0.08</td>
</tr>
<tr>
<td>Fc8</td>
<td>n.d.</td>
<td>0.12</td>
<td>0.19</td>
<td>3.58</td>
<td>n.d.</td>
<td>0.02</td>
<td>0.11</td>
</tr>
</tbody>
</table>

The contents ($\mu$g/g) of ions were measured with detection limits ranging from 2.4–3.9 ng/ml (Zhu, 1999); n.d.: not detected.

### Table 4. Bulk stable isotope compositions of fluid inclusions

<table>
<thead>
<tr>
<th>Sample</th>
<th>$\delta D$(H$_2$O)‰</th>
<th>$\delta^{13}$C(CH$_4$)‰</th>
</tr>
</thead>
<tbody>
<tr>
<td>FC1</td>
<td>-51.8</td>
<td>-25.5</td>
</tr>
<tr>
<td>FC2</td>
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<tr>
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<tr>
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<td>-51.5</td>
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</tr>
</tbody>
</table>

about 700°C and held at this temperature for about 10 minutes in high vacuum. The decrepitated gases were then condensed in traps cooled by liquid nitrogen to prevent occurrence of reactions among the different gas species. After the hot tube cooled down to room temperature, liquid nitrogen was removed and the total velamen pressure of all gaseous components was recorded. Afterwards, H$_2$O was frozen by using dry ice slush and subsequently the H$_2$O-absent velamen pressure was then recorded. The molar ratio of H$_2$O versus the other components in the gas can be calculated by using the ratio of pressure reduction due to the removal of H$_2$O versus the total velamen pressure. The results show that the bulk composition of the fluid inclusions is very H$_2$O-rich with molar ratios (XH$_2$O) ranging from 87 mol.% to 94 mol.% H$_2$O.

### Ion chromatography

The samples are characterized by very low ionic contents, the most abundant cation being Na$^+$ and the most abundant anion is Cl$^-$. Due to the very low Cl$^-$, Ca and Mg contents, only very small amounts of salts such as NaCl, CaCl$_2$ and MgCl$_2$ can thus be present in the fluid inclusions (Table 3).

### Stable isotope analysis

Results of hydrogen and carbon isotope analyses are listed in Table 4. The Raman and QMS measurements suggest that the $\delta^{13}$C/$^{12}$C values reported here can be attributed to CH$_4$ from the decrepitated fluid inclusions. The isotopic composition of the fluid inclusions from this study, is characterized by $\delta D$(H$_2$O) values ranging from -56.3 to -49.8‰ and $\delta^{13}$C(CH$_4$) values ranging from -30.1 to -25.5‰ (Table 4).

### DISCUSSION

**P-T constraints on the entrapment of the fluid inclusions**

From the coexistence of CH$_4$- and H$_2$O-rich, vapor and liquid-dominated fluid inclusions, it is supposed that the fluids were possibly entrapped under immiscible conditions (Kelley, 1996). QMS measurements of the bulk chemical composition of the fluid inclusions have shown that the bulk fluids contain 87–94 mol.% H$_2$O. Experimental investigations of CH$_4$-H$_2$O-fluids indicate that at temperatures below 400°C a homogeneous CH$_4$-H$_2$O-fluid will unmix into a CH$_4$- and a H$_2$O-rich fluid, thus putting an upper temperature constraint on the entrapment temperature of the fluid inclusions (Holloway, 1984). The range of the homogenization temperatures of the fluid inclusions of 315–378°C thus yields a lower temperature limits for the formation of these fluid inclusions. The real trapping temperatures are expected to be higher if a pressure correction of the homogenization temperature is applied. Huizenga (2001) shows in a thermodynamic evaluation of the C-O-H system that CO$_2$-poor fluids are stable only at temperatures below 400 to 450°C, depending on the fO$_2$ and whether graphite was present. Calculation of isochores, based on the composition of the fluid inclusions has not been attempted for the following reasons: (1) the lack of microthermometric data of the CH$_4$-rich/ H$_2$O-poor fluid inclusions, and (2) the high probability of density modification during uplift, which leads to considerable pressure underestimations as shown in many studies of fluid inclusions in high pressure rocks (see Philippot and Selverstone, 1992; Selverstone et al., 1992; Sterner and Bodnar, 1989). Despite this discrepancy, fluid inclusions from high-pressure rocks still give insight on the fluid/rock interactions at high pressure conditions (Philippot and Selverstone, 1992; Selverstone et al., 1992). The determination of the pressures of the formation of the jadeites is hampered by the lack of appropriate equilibria and the lack of quartz. Equilibria such as jadeite = albite + nepheline and jadeite + H$_2$O = analcite thus provide only lower pressure limits since the low-P phases are absent in the rocks. For the temperature range of the investigations, application of the analcite breakdown reaction according to the calculations of Harlow (1994) yields lower pressure limits of 0.6 to 0.7 GPa between 250 and 400°C for the formation of the Guatemala jadeites. But this pressure is below the lower stability limit of jadeite + H$_2$O relative to analcite at 0.8 to 1.0 GPa (Boeticher and Wyllie, 1969), even slightly lower than a lower limit of formation of omphacite + quartz of 0.6 to 0.8 GPa at 200 to 300°C in Californian metamorphic rocks.
CH₄ is the only hydrocarbon that was found in the fluid inclusions of this study. CH₄ occurring in the Earth’s crust is either formed by abiogenic or by biogenic processes. Abiogenic processes involve outgassing of primordial CH₄ from the mantle, the formation of CH₄ during inorganic chemical reactions involving species such as CO₂, H₂ or other C-H molecules at temperatures >300°C (Fischer-Tropsch-type reactions) or thermogenic processes (thermochemical decomposition of organic matter). Biogenic processes involve the alteration of carbon through bacterial processes (Schoell, 1988). Although CH₄-rich fluid inclusions have been frequently observed in nominally fluid-absent rocks such as peridotites and gabbros (Kelley, 1996; Kelley and Früh-Green, 1999; Su et al., 1999; Yang et al., 2001), their presence is not an unambiguous criteria for abiogenic formation and hence carbon isotopes (δ¹³C) are a widely used tool in distinguishing between biogenic and abiogenic processes responsible for the formation of CH₄, although an attribution to either one or the other process is still a matter of debate (Kelley and Früh-Green, 1999; Whiticar, 1999).

Abiogenic CH₄ is characterized by highly enriched δ¹³C(CH₄) ranging from −25 to −8‰ as indicated by fluid inclusions from plutonic samples from the East Pacific Rise and the Southwest Indian Ridge (Welhan and Craig, 1983; Abrajano et al., 1998). On the other hand, biogenic CH₄ is characterized by highly depleted δ¹³C(CH₄) with values ranging from −110 to −50‰ and low contents of additional hydrocarbons (Whiticar, 1999; Charlou et al., 2002). In contrast to CH₄ from bacterial processes, thermogenic CH₄ is enriched in ¹³C-compound with a δ¹³C(CH₄) of −50 to −20‰.

The isotopic composition of CH₄ in the fluid inclusions of this study is characterized by δ¹³C(CH₄) values ranging from −30.1 to −25.5‰, which could be indicative of an abiogenic origin since these data are very close to abiogenic CH₄ (East Pacific Rise, Welhan and Craig, 1983; Zambales Ophiolite, Abrajano et al., 1988). Although mantle carbon either occurs as CO or as CO₂ with δ¹³C values of −7 to −5‰, several studies have shown that mantle peridotites can contain significant quantities of reduced carbon species such as CH₄ which show distinctly lighter carbon isotope ratios of −30 to −20‰ (see Kelley and Früh-Green, 1999 and references therein).

Moreover, seemingly primordial CH₄-rich fluid inclusions from mantle peridotites are very rare and contain significant amounts of additional components such as hydrocarbons, H₂, N₂, H₂S and CO₂ (Su et al., 1999; Yang et al., 2001). The formation of CH₄ in most mantle peridotites though is probably due to reduction of CO₂ during water-rock interaction since there is no evidence that CH₄ is present in the upper mantle in other than minor concentrations, because the oxidation state of the upper mantle is too high (Apps and van de Kamp, 1993).

In addition, several studies have shown that negative δ¹³C values from alkali basalts and peridotite xenoliths from East China might result from recycled crustal components (e.g., see Kessen and Ringwood, 1989). Yang et al. (2001) found CH₄-rich fluid inclusions in peridotite xenoliths from the Dabie terrane and their investigation indicated that the CH₄ most likely formed due to the influence of CO₂-rich aqueous fluids during serpentinisation prior to or during plate subduction. These observations lead a number of studies to conclude that later-stage abiogenic formation of CH₄ instead of degassing of primordial CH₄ is more common than previously thought (e.g. see Charlou et al., 2002 and references therein). In addition, recent experimental investigations by Berndt et al. (1996) and Horita and Berndt (1999) have shown that CH₄ can be produced by serpentinization of olivine in the presence of Fe-Ni alloys through reactions involving H₂O and CO₂ on the reactant side such as forsterite + H₂O + CO₂ = magnetite + serpentine + brucite + CH₄ + H₂ or forsterite + H₂O = magnetite + serpentine + brucite + H₂ where H₂ reacts with CO₂ to form CH₄ and H₂O according to the reaction CO₂ + 4H₂ = CH₄ + 2H₂O (Fischer-Tropsch synthesis). This process might be able to explain the occurrence of abiogenic CH₄ in shallow plumes in sediment layers above ultramafic rocks on the oceanic floor (Rona et al., 1992; Charlou and Donval, 1993), fluid inclusions in olivine-rich drill cores from the oceanic crust (Kelley, 1996), CH₄-rich gas hydrates in subduction zones (Kastner et al., 1998) and the occurrence of CH₄-rich gases emanating from ophiolite complexes (Abrajano et al., 1988; Sturchio et al., 1989).

Although the jadeitites occur within strongly altered (serpenitized) peridotites and the isotope signature obtained from the fluid inclusions in the jadeitites point to a possible abiogenic origin, fluids which are attributed to the formation by Fischer-Tropsch synthesis should contain significant amounts of H₂ (Kelley and Früh-Green, 1999; Charlou et al., 2002) which has not been found in the fluid inclusions of this study. However, H diffusion out of the system can be quite faster even at low temperatures. A recent study by McGollom and Seewald (2001), on the other hand, has shown that Fischer-Tropsch-type reactions alone cannot account for large-scale reduction
of dissolved CO₂ to CH₄ since the conversion of CO₂ into CH₄ in their experiments was highly insufficient due to kinetic reasons despite extremely long run times (2500 h). In addition, the presence of a Ni-Fe alloy also seems to be of crucial importance in the conversion of CO₂ into CH₄, although the effectiveness of this alloy over time seems to decrease strongly (Horita and Berndt, 1999). No Ni-Fe alloys have been found in the adjacent serpentinitized peridotites surrounding the jadeites in this investigation so far and only magnetite has been found. Several studies also concluded that fluids from peridotite-hosted hydrothermal systems where incomplete conversion of CO₂ into CH₄ took place, should thus contain large fractions of formate (McCollom and Sewald, 2001; Sherwood Lollar et al., 2002). The absence of Ni-Fe alloys in the peridotites therefore does not need to indicate abiogenic formation of these fluid inclusions by serpentinitization of the surrounding ultramafic country rocks.

Thermogenic CH₄ is formed as organic-rich sediments move through progressively higher thermal maturity regimes during increasing depths of burial. The main process of formation is thermochemical decomposition of organic matter. During early maturation (<150°C) thermal CH₄ is accompanied by other hydrocarbons and non-hydrocarbon gases, whereas at highest thermal maturities, CH₄ alone is formed by either breaking C-C bonds in kerogen, bitumen and oils or from the reaction between water and graphite (Wiese and Kvenvolden, 1993). The isotopic composition of these thermal gases depends on: (1) the isotopic composition of the source rock, (2) the degree of fractionation during thermogenesis, and (3) the degree of fractionation after CH₄ formation. Thermal CH₄ has a range of carbon isotopic values from ca. −50 to −25‰ depending on the degree of fractionation of the lightest and/or heaviest source material (Fux, 1979). Therefore, depending on the carbon isotope separation between thermogenic CH₄ and organic matter (δ¹³C(CH₄) – δ¹³C(Corg)) which ranges between 0 and ca. 30‰, thermogenic CH₄ might still retain the isotopic signature of the precursor organic matter which has on average a δ¹³C of −25 ± 1.5‰ (Deines, 1980).

The stable isotopic signature of hydrogen in most cases consists of at least two sources, a mantle-derived magma and/or a secondary source (caused by crustal subduction or fluid venting). The hydrogen isotopic compositions of hydrous minerals or fluid inclusion in high pressure (HP) rocks are rarely homogeneous and reflect local equilibrium (Scambelluri and Philippot, 2001). It is generally assumed that fluids in the upper parts of a subduction zone represent seawater and δD(H₂O) will start changing when the water/rock ratio is sufficiently reduced so that δD(H₂O) of the fluid is influenced with hydrous minerals or by hydrous mineral breakdown (Johnson and Harlow, 1999). At high pressure the H isotopic signatures of serpentinites spread over a large range of values which reflected pre-eclogitic interactions with seawater-derived fluids and with (D-depleted metamorphic fluids released at fore-arc environments (Früh-Green et al., 2001). This will lead to a considerable shift to isotopically lighter δD(H₂O) values due to the contribution of metamorphic water which has a δD(H₂O) of −70 to −20‰. The isotopic composition of the fluid inclusions from our study is characterized by δD(H₂O) values ranging from −56.3 to −49.8‰, suggesting that the water in the fluid inclusions in the jadeites might reflect a strong component of metamorphic water released by dehydration reactions within the metamorphosed rocks. This differs from the Guatemala jadeites which are isotopically heavier with a more distinct seawater signature (Johnson and Harlow, 1999). Johnson and Harlow (1999) also concluded that their isotopic shift to heavier δD(H₂O) values was due to serpentinitization which also was suspected to lead to the formation of H₂, which is completely absent in our fluid inclusions.

On the other hand, δD(H₂O) values of hydrous minerals from ultramafic mantle rocks are also in a similar range from −90 to −40‰. Typical signature of meteoric water was observed in hydroxyl-bearing minerals from UHP eclogites and gneisses from the Dabie–Sulu terranes, with δD values of −127 to −61‰ for micas, −100 to −72‰ for amphiboles, −75 to −37‰ for epidotes (Rumble and Yui, 1998; Zheng et al., 1998, 1999; Fu et al., 1999; Xiao et al., 2002; Li et al., 2004). Compared to fluid inclusions from the eclogites, the δD(H₂O) values from our study not only fall within the δD range of hydroxyl-bearing minerals, but also are very similar to the δD(H₂O) obtained from the Monviso eclogites from the Western Alps (Nadeau et al., 1993), although both localities show a much larger variation in δD due to small scale heterogeneities. Overall, the δD(H₂O) data favor a crust-derived fluid with insignificant input from seawater.

The origin of carbon and CH₄ in high-pressure rocks and implications for carbon cycling in subduction zones

Recent investigations to constrain the retention and loss of volatile elements during subduction showed that fluxes of carbon into subduction zones are larger than returned to the surface in arcs (Sadofsky and Bebout, 2003; Shaw et al., 2003). If this is true, then large amounts of carbon would be transported into the deeper mantle and could contribute to mantle carbon budgets and thus to carbon (CO₂, CH₄) fluxes in ocean-island basalts and at mid-oceanic ridges (e.g., see Marty and Tolstikin, 1998; Charlou et al., 2002 and references therein) as well as in HP and UHP metamorphic rocks from continental subduction zones (e.g., see Xiao et al., 2001; Zheng et al., 2000, 2003a, 2003b). For instance, several fluid inclusion studies from the UHP Dabieshan terrane have shown
that organic carbon seems to play an important role in the carbon budgets of paleo-subduction zones (Yang et al., 2001; Zheng et al., 2000, 2003b). Stable isotope studies on fluid inclusions in peridotites, enclosed in eclogites, revealed $^{13}$C-depleted CO$_2$ (down to $-25.1\%$) present in this UHP terrane, which is most likely derived from oxidation of organic matter during interaction with surface fluids during prograde UHP metamorphism. On the other hand, fluid inclusions from eclogites also showed enrichment in $\delta^{13}$C ($-18.5$ to $4.6\%$), indicating that decarbonisation of marbles also contributes to the $\delta^{13}$C signature of CO$_2$ (Yang et al., 2001). Unfortunately, no such data on CH$_4$-rich fluid inclusions in paleo-subduction zones, yet exist.

Most of the evidence for the presence of CH$_4$ at deep crustal levels (10 to 35 km) comes from graphite-bearing metasedimentary rocks where reactions with graphitic crustal levels (10 to 35 km) comes from graphite-bearing metasedimentary rocks except for some occurrences in granulites (Hall and Bodnar, 1989; Horai et al., 2000) and very rare occurrences in eclogite-facies rocks (Klemd et al., 1992, 1995; Iakub et al., 1999). Several studies from the Dabie-Sulu terrane have also shown CH$_4$-bearing, CO$_2$-rich fluid inclusions in the eclogites and felsic granulites (Xiao et al., 2000, 2002; Fu et al., 2001, 2002, 2003a, 2003b; Yang et al., 2001) and the low $\delta^{13}$C signature in the UHP eclogites and gneisses (Zheng et al., 2000; 2003a, 2003b; Li et al., 2000). These studies show that the highest-grade fluids are comprised of N$_2$ + CO$_2$ and CH$_4$-bearing fluid inclusions only occur at the retrograde portion of the P-T path. Thus no data on the presence of CH$_4$-rich fluid inclusions during growth of high-P minerals such as jadeite or omphacite yet exist. Our data thus indicate that CH$_4$-bearing fluids can still occur during the prograde path of high-P rocks such as jadeitites.

Unfortunately, in the literature there is only indirect evidence for the presence of CH$_4$ in subduction zones so far, namely in CH$_4$-rich plumes emanating from oceanic troughs in convergent margins (Craig et al., 1987; Watanabe et al., 1994; Tsunogai et al., 1998). These plumes form in the accretionary prisms of the subduction zones and rise from depths of 1 to 3 km and their isotopic signatures ($\delta^{13}$C) indicated either CH$_4$ production by microbial activity in shallow sedimentary layers or thermogenic production of CH$_4$ from organic sediments (Haggerty, 1991; Tsunogai et al., 1998). Thus the occurrence of CH$_4$-bearing fluid inclusions in high-P rocks from subduction zones helps to put constraints on the stability limits of CH$_4$-bearing fluids as well as constraints on the flux of CH$_4$ in subduction zones. Sadofsky and Bebout (2001, 2003) investigated low-T high-P rock suites from the Franciscan Complex and the Western Baja Terrane (Mexico) to study the influence of forearc devolatilization on geochemical cycling in convergent margins. Their study showed that rocks from the Franciscan Complex contained carbonate veins with $\delta^{13}$C values very close to marine carbonates (most between 0 and $-10\%$) and reduced graphite with organic-like $\delta^{13}$C (ca. $25\%$) thus indicating that most of these rocks have not experienced significant decarbonisation. In cases where decarbonisation took place, shifts to higher $\delta^{13}$C of the reduced carbon occurred. Sadofsky and Bebout (2001, 2003) showed that carbon in the Franciscan Complex originated from marine sediments, thus organic material was the source of the carbon.

Assuming that thermogenesis of organic carbon is the most likely mechanism of CH$_4$ production in the fluid inclusions of this study, Fuex (1979) showed that a source rock with organic carbon having $\delta^{13}$C of $-19\%$, which experience minimum fractionation of the heaviest source material, is able to produce CH$_4$ which has a $\delta^{13}$C of $-25\%$. Considering that average marine organic matter has a $\delta^{13}$C of $-25\%$ (Deines, 1980), formation of CH$_4$ from a mixture of organic carbon and carbon which evolved from carbonates, would allow the formation of thermogenic CH$_4$ with a $\delta^{13}$C similar to the isotopic values yielded from our fluid inclusions. Figure 6 shows a simple ideal calculation of the evolution of $\delta^{13}$C(CH$_4$) by Rayleigh distillation in a closed system when it evolves...
either from graphite (starting $\delta^{13}C = (25\%)$ or calcite (starting $\delta^{13}C = 0\%)$). The calculations and the temperatures of formation of these fluid inclusions indicate that the stable isotope signatures of the fluid inclusions from the jadeitites rather point to an origin from graphite after high fractionation of CH$_4$ out of it instead of forming from calcite alone by decarbonisation, which is very similar to the formation of the CO$_2$-rich fluids in the Dabie-Sulu terranes (Zheng et al., 2000; Yang et al., 2001). On the other hand, mixtures of both sources allows $\delta^{13}C(CH_4)$ values which are between these two end-member scenarios and possibly close to our obtained data. Although we do not have any isotopic data of carbon (graphite) from whole-rock samples in this study, but diopside-bearing marbles do occur in the vicinity of these veins, fractionation data indicate that the $\delta^{13}C(CH_4)$ values of the fluid inclusions from these rocks from a similar low-P high-T setting may have evolved either by (1) gradual fractionation from an isotopically similar protolith source (organic carbon from marine sediments) or (2) an isotopically heavier protolith such as a mixture of marine sediments and marine carbonates as indicated in Fig. 6.

**Conclusions**

The $\delta^{13}C(CH_4)$ values and the absence of Fe-Ni alloys in adjacent serpentinites and additional components in the fluid inclusions such as hydrocarbons and H$_2$ indicate that CH$_4$ production due to biogenic processes such as bacterial alteration and abiogenic processes such as serpentinization or the release of primordial CH$_4$ from the mantle might be the unlikely sources for CH$_4$ in these fluid inclusions. Nonetheless, if these processes occurred they must have been subordinate and hence other abiogenic processes such as thermogenic processes (thermal decomposition of organic matter) in subducted organic carbon from marine sediments must be considered to be of primary importance in the formation of CH$_4$-rich fluids in these rocks. These data indicate that in addition to CH$_4$ release in plumes from sediments from accretionary prisms in convergent margins, CH$_4$ might also be stable to at least the upper 20 km of a subduction zone where jadeite veins formed under low-T and high-P conditions.

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