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REVISES

Cumulates of arc magmas incorporated into the Sulu UHP metamorphic belt, eastern China

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ABSTRACT

The Hujialin ultramafic complex in the central region of the Sulu ultra-high pressure (UHP) metamorphic belt consists of discontinuous lenses of garnet-bearing clinopyroxenite and dunite surrounded by marginal serpentinite. The clinopyroxenite shows relatively low concentrations of compatible elements, such as Cr (≤ 1670 ppm) and Ni (≤ 514 ppm) and Ir-group platinum group elements (IPGE; Ir, Os, and Ru; ≤ 4.8 ppb in total). They show varying ratios (0.02–2.50) of IPGE to Pd-group PGE (PPGE). Their chondrite-normalized rare earth elements (REE) patterns are convex and the total REE concentrations range from 18 to 63 times that of CI chondrite. The bulk rocks show a 'subduction-related' geochemical signature, with high concentrations of fluid-mobile elements (i.e. Sr, Ba) relative to high-field strength elements (i.e. Nb, Y, Zr). Clinopyroxene is diopside and contains low Al_2O_3 (< 2.76 wt.%) and high SiO_2 (54.6–56.9 wt.%). Olivine grains enclosed by clinopyroxene and in the matrix show relatively low Fo (76.6–80.7) and NiO contents (0.18–0.29 wt.%). The bulk rock compositions and mineral chemistry of olivine and clinopyroxene suggest that the unit was a cumulate of a subduction-related melt. On the other hand, dunite and its hydration product, serpentinite, have a different origin. The bulk rock and mineral chemistry suggest that dunite represents a mantle wedge peridotite in a spinel-stable field. Both clinopyroxenite and spinel-bearing dunite were once located in the mantle wedge below the southern margin of the North China craton (NCC), and were dragged by a mantle flow into the continental subduction channel along the interface between the subducting Yangtze craton (YZC) and the overlying NCC. Although clinopyroxenite and dunite are dense (2.8–3.2 g/cm³), the buoyancy-driven exhumation of voluminous granitic rocks of the YZC likely brought clinopyroxenite and dunite to shallow crustal depths. The lack of the evidence for high pressure to ultra-high pressure (HP-UHP) metamorphism in spinel-bearing dunite may be explained by overall low Al and Ca in the bulk rocks. Alternatively, dunite was not subducted to deep levels, but exhumed together with the deeply subducted clinopyroxenites and granite during their exhumation.

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1. Introduction

The Dabie-Sulu ultra-high pressure (UHP) terrane was formed during the subduction of the northern margin of the Yangtze Craton (YZC) beneath the North China craton (NCC) following the collision of the two continents in Triassic time. The terrane is one of the largest and longest continuous ultra-high-pressure metamorphic (UHP) belts in the world (Yang *et al.* 2003). The terrane is primarily composed of granitic gneisses, but contains volumetrically minor ultramafic rocks that form massifs and large lenticular bodies measuring metres to hundreds of metres (Zhang *et al.* 2000; Ye *et al.* 2009). A majority (>90%) of ultramafic rocks contain garnet, suggesting that they underwent deep subduction, but some

rocks contain spinel instead of garnet as an Al mineral (e.g. Chen *et al.* 2009; Xie *et al.* 2013).

Ultramafic rocks in the Dabie-Sulu belt are grouped into three based on field occurrences, bulk rock composition, and mineral chemistry. The first group includes garnet-bearing peridotites in the Rongcheng, Rizhao, Yangkou, and Donghai areas (Figure 1a). They are considered to be fragments of subcontinental lithospheric mantle (SCLM) below the margin of the NCC, subducted deep together with the YZC lithosphere (Zhang *et al.* 2000, 2005; Ye *et al.* 2009). The second group is cumulates of mafic magmas that show well-developed compositional layering and evidence of fractional crystallization from ultramafic to mafic rocks. The rocks were originally in the YZC and subducted to reach the

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Figure 1. (a) Geologic sketch map of the Sulu UHP terrane showing localities of peridotites (black solid circle) and pyroxenites (grey solid pentagon), coesite-bearing eclogites (pentagon), and CCSD-drilling sites (modified after Zhang *et al.* 2005). The Sulu belt is bounded by the Yantai-Wulian fault (YWF) and the Tan-Lu fault to the west and the Jiashan-Xiangshui fault (JXF) to the southeast. CCSD-MH, the main drill hole of the Chinese Continental Scientific Drilling Project; PP1, the pre-pilot hole. (b) Simplified geological map of Hujialin area showing the distribution of ultramafic bodies (filled area) (modified after Zhang and Liou 2003). (c) Sampling locations in the Hujialin ultramafic complex.

peak metamorphic conditions of 760–970°C and 4.0–6.5 GPa (e.g. Zhang *et al.* 2000). Examples include the Bixiling and Maowu ultramafic complexes in the Dabie belt (e.g. Zhang *et al.* 2000; Zheng *et al.* 2008). The third group is spinel-bearing harzburgite and dunite in the Raobazhai area of the northern Dabie belt and in the Yangkou and Suoluoshu areas of the Sulu belt (e.g. Zheng *et al.* 2008; Xie *et al.* 2013). The Hujialin ultramafic complex, the focus of this manuscript, is one of complexes in the Suoluoshu-Hujialin area in the Sulu belt (Figure 1b).

The Hujialin ultramafic complex is located in the central region of the Sulu UHP metamorphic belt and is surrounded by granitic gneisses (Figure 1a). It consists mainly of serpentinized dunite and discontinuous lenses of garnet-bearing clinopyroxenite (Figure 1c). The origin and tectonic evolution of Hujialin clinopyroxenite remain controversial. Proposed origins include: (1) a reaction product of peridotite with an evolved basaltic melt; (2) a pyroxenite from the upper mantle (e.g. Chen and Xu 2005); and (3) a cumulate of mantle-derived magma (e.g. Yang 2006). Most studies agree that Hujialin clinopyroxenite was subducted to the deep mantle and underwent UHP metamorphism, but questions remain how lenses of garnet-bearing

clinopyroxenite came in contact with spinel-bearing lenses and how the Hujialin ultramafic complex was incorporated into the UHP belt and exhumed to the surface.

This paper presents major and trace element compositions of bulk rocks and mineral compositions of clinopyroxenite in the Hujialin ultramafic complex, and evaluates (1) the origins of these two ultramafic rocks; (2) the tectonic evolutions of the area; and (3) the nature of the subduction channel between the YZC and the overlying NCC.

2. Geological setting

2.1. The Sulu belt and ultramafic occurrences

The NCC is an Archaean craton and is underlain by a thick (~250 km) lithospheric mantle, but the eastern part is underlain by a thin (<60 km) lithospheric mantle after the mid-Mesozoic time (e.g. Kusky *et al.* 2007). The Dabie-Sulu terrane represents a suture zone between the NCC and the YZC. The rocks of the Sulu belt are well exposed in the southeastern part of the Shandong Peninsula in eastern China (Figure 1a). The Sulu belt is bounded by the Yantai-Wulian fault to the west and the Jiashan-

Xiangshui fault to the southeast (Figure 1a). Rocks in the belt are mostly granitic gneisses, migmatites, and granitoids, with lenses and layers of eclogites, metasediments, and mafic-ultramafic massifs. U–Pb ages and Hf-isotope data of zircons suggest that the majority of granitic gneisses have middle Neoproterozoic protoliths (780–740 Ma; e.g. Tang *et al.* 2008; Zheng 2008). Sm–Nd and U–Pb dating of the eclogites, UHP gneisses, and garnet peridotites shows the peak metamorphism at 245–220 Ma (e.g. Liu *et al.* 2004; Hacker *et al.* 2006; Ye *et al.* 2009). The presence of exsolved clinopyroxene, rutile, and apatite in garnet from Yangkou eclogites attests that they were subducted to a depth greater than ~200 km (Ye *et al.* 2000). These UHP metamorphic rocks are overlain by young rocks, which include Jurassic siliciclastic and Cretaceous volcanoclastic rocks, and Mesozoic granites.

Ultramafic rocks in the Sulu belt are mostly garnet peridotites and garnet pyroxenites, and they are mostly intercalated with country-rock granitic gneisses and metasediments. Garnet peridotites and garnet pyroxenites also occur in the main drill core of the Chinese Continental Scientific Drilling Project (CCSD-MH), the pre-pilot drill core of hole-1 of CCSD (CCSD-PP1), and the pre-pilot drill core of hole-3 of CCSD (CCSD-PP3) in the Donghai area of the southern Sulu terrane (Figure 1a; Chen *et al.* 2009; Yang *et al.* 2009; Zhang *et al.* 2010). Previous studies suggested that these garnet peridotites and garnet pyroxenites mostly recrystallized at depths within the diamond stability field (e.g. Zhang *et al.* 2000, 2009a). Minor spinel-bearing peridotites (dunite, minor harzburgite, and very minor lherzolite) contain no garnet and show no evidence of UHP metamorphism, and they may have undergone different metamorphic paths (e.g. Chen *et al.* 2009; Xie *et al.* 2013).

2.2. Geological setting of the study area and sampling

The Hujialin area of the Sulu belt contains numerous lenses and rounded bodies of serpentinite and eclogite along faults and mylonite zones of granitic gneissic rocks (Figure 1b). The mafic-ultramafic bodies in the area are considered to be rootless and tectonically emplaced into the gneissic rocks (e.g. Gao *et al.* 2004; Yang 2006). Among these ultramafic rocks, the Suoluoshu ultramafic complex, the largest in the area with a surface exposure of 5.5 km × 0.8 km, has been studied by previous researchers. It contains several lenses of eclogite showing the pseudomorphs of coesite and abundant exsolved rutile needles in garnet (e.g. Zhang and Liou 2003). The evidence suggests that the surrounding gneissic rocks and lenses of eclogites have

undergone UHP metamorphism (e.g. Gao *et al.* 2004). On the other hand, serpentinites with spinel and antigorite show no evidence of UHP metamorphism (Xie *et al.* 2013). Spinel grains in serpentinites are rimmed by thin magnetite, but no development of ferritchromite that is common for metamorphosed serpentinites (Saumur and Hattori 2013).

The Hujialin ultramafic complex is the second largest ultramafic body in the area, with a NNW-elongated surface exposure of 2 km in length with width varying from 0.2 to 1 km (Figure 1c). The north end of the ultramafic body is bent EW, and mainly consists of serpentinitized peridotites, discontinuous lenses of clinopyroxenite, and dunite surrounded by serpentinite and minor harzburgite (Figure 1c), with sharp contacts between clinopyroxenites and peridotites. The abundance of garnet in clinopyroxenite varies widely even within an outcrop. Some areas show no visible garnet grains and other areas contain abundant garnet up to 35 vol.%.

3. Samples and petrology of ultramafic rocks in the Hujialin complex

We examined the total nine samples of clinopyroxenite collected in the northern and southern lenses of the Hujialin complex (Figure 1c). Dunite samples were derived from the central part of the complex and sample locations are shown in Figure 1c.

Clinopyroxenite is divided into three types based on their mineral mode. Type-I (sample HJL6) lacks garnet. It mainly consists of coarse-grained clinopyroxene (subhedral-anhedral; 0.5–1.0 mm in size, 10 vol.%), fine-grained clinopyroxene (anhedral; ≤0.3 mm in size, 80 vol.%), Fe oxide minerals (magnetite and/or ilmenite; 0.1–0.3 mm in size, 10 vol.%), alteration minerals (chlorite and/or amphibole), and minor olivine (0.05–0.15 mm, <1 vol.%) (Figure 2a and b). Coarse-grained clinopyroxene is commonly rimmed by fine-grained clinopyroxene and Fe oxides, and contains fine-grained olivine inclusions (Figure 2e). Type-II (samples HJL8, HJL9, HJLII-1, and HJLII-5) is the predominant type of clinopyroxenite and is characterized by abundant grey and reddish garnet grains in hand specimens. It contains garnet megacrysts (up to 20 mm, 20–40 vol.%), anhedral clinopyroxene (0.2–1.0 mm, 50–60 vol.%), and Fe oxide minerals (0.2–0.8 mm, <15 vol.%). Megacrystic garnet commonly contains fine-grained clinopyroxene and Fe oxides (Figure 2c), and is partially coated by a rim of chlorite, amphibole, and epidote. Type-III (samples HJL7, HJLII-3, HJLII-6 and HJLII-7) is characterized by abundant Fe oxides (30–40 vol.%) and porphyroblastic clinopyroxene (samples HJL7 and HJLII-3), with abundant exsolution lamella of garnet, clinopyroxene, magnetite, and

Figure 2. (a) Photomicrograph of coarse-grained clinopyroxene (Cpx) surrounded by fine-grained Cpx and Fe oxides (magnetite: Mag and ilmenite: Ilm) under crossed polarized light (sample HJL6). (b) Photomicrograph of coarse-grained Cpx surrounded by fine-grained Cpx, amphibole (Amp), and chlorite (Chl) (sample HJL6). (c) Photomicrograph of porphyroblastic garnet (Grt) containing inclusions of Cpx and Fe oxides in sample HJL8. (d) Photomicrograph of porphyroblastic Cpx rounded by fine-grained Cpx, Fe oxides, and relict garnet. (e) Back-scattered electron (BSE) image of olivine (Ol) inclusion in Cpx (sample HJL6). (f) BSE image of porphyroblastic Cpx with exsolved Grt, Cpx, Mg, and Ilm in sample HJLII-3 and exsolved Grt partially altered to Chl.

ilmenite (Figure 2d and e). It also contains fine-grained clinopyroxene (40–50 vol.%) and relict garnet (0.2–0.5 mm in size, ≤10 vol.%) and green, Al-rich spinel (≤2 vol.%). Rims of relict garnet are commonly replaced by chlorite, amphibole, and epidote.

4. Analytical methods

Mineral compositions were determined using a CAMEBAX MBX electron probe by the wavelength dispersive method at Carlton University in Ottawa. Counting times were 20 s per element, and analytical conditions were 15 kV accelerating voltage and 20 nA beam current. The calibration used wollastonite (Si, Ca), synthetic spinel (Al), synthetic Cr₂O₃ (Cr), forsterite (Mg),

synthetic MnTiO₃ (Mn, Ti), vanadium metal (V), albite (Na), fayalite (Fe in silicates), and synthetic Fe₂O₃. Fe³⁺ contents of spinel were calculated assuming a stoichiometric composition. Compositions of individual minerals are similar within individual samples, and representative compositions of minerals are listed in Supplementary Tables 3 and 4.

Weathered surface was removed before bulk rock geochemical analysis. The major element compositions of samples were analysed with a Phillips PW 2400 X-ray fluorescent spectrometer after fusing powder with a flux composed of 78.5% Li₂B₄O₇ and 21.5% LiBO₂ at the University of Ottawa. Precision based on a replicate run of three samples is ±0.35% for Al₂O₃, ±0.48% for MgO, ±1.3% for Cr, and ±9.2% for Ni. The accuracy,

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which was monitored using international references for SY-2 and MRG-1, shows $\pm 0.039\%$ for Al_2O_3 , $\pm 0.28\%$ for MgO , $\pm 3.4\%$ for Cr , and $\pm 4.0\%$ for Ni . Precision and accuracy are less than 1 and 10%, respectively, for other major and minor elements. Contents of PGE in bulk rocks were determined by the isotopic dilution method using a mixed spike of ^{99}Ru , ^{105}Pd , ^{190}Os , ^{191}Ir , and ^{194}Pt after pre-concentration of PGE into a Ni-sulphide bead that was dissolved in 6 N HCl. The filtrate was dissolved in concentrated HNO_3 before analysis with an Agilent HP 4500 inductively coupled plasma mass spectrometer (ICP-MS) at the University of Ottawa. Blanks for this analysis were 0.0032 ng Ir/g flux, 0.0007 ng Os/g flux, 0.0026 ng Pt/g flux, 0.035 ng Pd/g flux, and 0.0049 ng Ru/g flux. These values are negligible compared with those in the samples, and thus blank corrections were not applied to the results. The analytical quality was monitored by running two rock references: TDB-1 diabase from CANMET, Natural Resources of Canada; and JP-1 harzburgite from the Geological Survey of Japan. The values for these are comparable to those obtained by Meisel and Moser (2004) (Supplementary Table 2). The concentrations of S, Cu, and Pb were determined using a Varian VISTA-PRO inductively coupled plasma-optical emission spectrometer (S, Cu) and Agilent HP 4500 ICP-MS (Pb) after digesting samples in *aqua regia* in screw-top Teflon vials for 48 h at 150°C . The precision based on replicate runs of five samples is $<11\%$ for Cu, $<7\%$ for S, and $<12\%$ for Pb. The trace elements of bulk rock samples were analysed after digestion in 1 ml of hydrofluoric acid (HF) and 0.5 ml of HNO_3 in screw-top PTFE-lined stainless steel bombs at 190°C for 12 h, followed by ICP-MS at the Key Laboratory of Ore Deposits, Chinese Academy of Sciences, Guiyang, China. All instrumental operating processes are referenced in Qi *et al.* (2000). Repeated analysis of rock references shows the average standard deviations are less than 10% and the average relative standard deviations are better than 5%.

5. Results

5.1. Bulk rock compositions of clinopyroxenite

Samples contain significantly low concentrations of compatible elements ($\text{Cr} = 52\text{--}1673$ ppm; $\text{Ni} = 98\text{--}514$ ppm) compared with the primitive mantle values (2625 ppm Cr and 1960 ppm Ni; McDonough and Sun 1995), and high contents of CaO (15.0–20.3 wt.%) and Al_2O_3 (3.13–13.3 wt.%) (Supplementary Table 1). The data suggest that clinopyroxenite is not a residue of mantle peridotites after partial melting. Instead, the clinopyroxenite is likely a cumulate of mafic magmas.

The cumulate origin is further supported by low concentrations of Ir-type PGE (IPGE; Os = 0.1–1.1 ppb, Ir = 0.1–2.3 ppb, Ru = 0.1–1.4 ppb) and various ratios of IPGE/PPGE (0.03–3.37) (Supplementary Table 2; Figures 4 and 5). Among PGE, IPGE is compatible with mantle minerals and remains in the residual mantle peridotites during partial melting. A melt and the cumulate, therefore, contain low concentrations of IPGE compared with the residual mantle peridotites. The primitive mantle-normalized Ni–PGE–Cu patterns of clinopyroxenite samples are similar to those of clinopyroxene-rich cumulate rocks in the Sanbagawa metamorphic belt (Hattori *et al.* 2010a), pyroxenites in the Ural-Alaskan-type ultramafic-mafic complexes along subduction zones (Garuti *et al.* 1997; Pettigrew and Hattori 2006), and garnet peridotites of an arc cumulate in the Dominican Republic (Hattori *et al.* 2010b). This pattern is in contrast to the high PGE contents of the ancient subcontinental lithospheric mantle peridotites of the NCC reported by Zhang *et al.* (2005) and Xie *et al.* (2013) (Figure 4).

The bulk rock chemical compositions of clinopyroxenite display a subduction-related geochemical signature, with high concentrations of fluid-mobile elements (i.e. Sr, Ba, and Pb) and low concentrations of high field strength elements, such as Nb, Y, and Zr (Supplementary Table 1; Figure 3a), which reflects the features of the original rocks. The total REE in clinopyroxenite samples are in the range of 18–63-fold CI chondrite (McDonough and Sun 1995), and their chondrite-normalized REE patterns are convex upward (Figure 3b) with $\text{La}_\text{N}/\text{Sm}_\text{N} = 0.23\text{--}0.61$ and $\text{Sm}_\text{N}/\text{Lu}_\text{N} = 3.6\text{--}9.6$. The bulk rock data of type-I and type-II clinopyroxenite show minor positive anomalies of Eu ($\text{Eu}/\text{Eu}^* = 1.08\text{--}1.20$) (Figure 3b), whereas type-III clinopyroxenite lacks the anomalies of Eu ($\text{Eu}/\text{Eu}^* = 0.79\text{--}1.04$). Since plagioclase preferentially incorporates Eu as Eu^{2+} , positive anomalies of Eu in type-I and type-II clinopyroxenite suggest that minor amounts of plagioclase likely crystallized as a cumulate phase. High CaO and Al_2O_3 in bulk rocks also support the presence of plagioclase in the original rocks.

Type-I clinopyroxenite (sample HJL6) is diopsidite with more than 90% diopside, some of which is altered to chlorite and amphibole. It contains lower contents of moderately incompatible elements (Al, Ti, and V) and higher contents of compatible elements (Mg, Cr, and Ni) than Type-II clinopyroxenite. Sample HJL6 shows high Pt and Pd values, attributed to late hydrothermal activity that brought Pt and Pd from the surrounding dunite as these are both relatively mobile in aqueous fluids (e.g. Hattori and Cameron 2004). Type-III clinopyroxenite contains relatively low compatible elements (Cr, Ni, and Mg) and relatively high Fe, Ti, and Co compared with other samples (Supplementary Table 1). High contents of Fe and Ti reflect

Figure 3. (a) Primitive mantle-normalized bulk chemical compositions of clinopyroxenite samples. Primitive mantle values are from McDonough and Sun (1995). The data of cumulates oceanic plateau and islands and Higashi-akaishi clinopyroxenites are from Hattori *et al.* (2010a). The data of garnet peridotites of an arc cumulate in the Dominican Republic (D.R.) are from Hattori *et al.* (2010b) and the data of Bixiling ultramafic-mafic rocks are from Zheng *et al.* (2008). (b) Chondrite-normalized REE patterns of clinopyroxenite samples. Data for C1 chondrite are from McDonough and Sun (1995).

abundant Fe oxides (magnetite and ilmenite, 30–40 vol.%) and suggest that the protolith of Type-III clinopyroxenite is likely cumulates of evolved magmas.

5.2. Mineral chemistry of clinopyroxenite

5.2.1. Clinopyroxene and garnet

Clinopyroxene is diopside and contains high MgO (15.12–17.57 wt.%) and SiO₂ (54.6–56.9 wt.%), low

TiO₂ (0.02–0.29 wt.%) and Na₂O (0.35–0.97 wt.%) (Supplementary Table 3). It differs from metamorphic clinopyroxene in eclogites as it contains low Al₂O₃ (<2.76 wt.%). The contents of Al is mostly less than 2% in the tetrahedral site of clinopyroxene (Figure 6a), which makes our clinopyroxene distinct from those in mid-oceanic ridge basalts and in alkaline igneous rocks common in oceanic plateau and islands.

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Figure 4. Primitive mantle-normalized PGE abundance in Hujialin ultramafic complex: (a) type-I and type-II clinopyroxenite and (b) type-III clinopyroxenite. Comparison of the data for Ural/Alaskan-type pyroxenite from Garuti *et al.* (1997), Higashi-akaishi clinopyroxenites from Hattori *et al.* (2010a), garnet peridotites of an arc cumulate in Dominican Republic (D.R.) from Hattori *et al.* (2010b), the ancient SCLM of the NCC from Zheng *et al.* (2005), and dunites in Hujialin from Xie *et al.* (2013). Primitive mantle values are from McDonough and Sun (1995).

Diopside in this study is the residual product of the precursor clinopyroxene after the removal of exsolved phases. The precursor clinopyroxene likely contained higher Al_2O_3 , FeO and TiO_2 than diopside in this study, because the exsolution of garnet, ilmenite, magnetite, and spinel (e.g. Hiramatsu and Hirajima 1995; Yang 2006). The compositions of the precursor clinopyroxene for Hujialin clinopyroxenites have been estimated by

Yang (2006) based on the modal proportions of the average compositions of minerals. The compositions of this precursor and current clinopyroxene plot in the field of arc cumulates in the SiO_2 - Al_2O_3 discrimination diagram of Loucks (1990) (Figure 6b).

Garnet shows varying proportions of pyrope and grossular components (Supplementary Table 3). Garnet in our samples falls into two distinctly populations. One is

Figure 5. Ratios of Ir/(Pt + Pd) versus (vs.) the contents of Ir (ppb) for our ultramafic rocks. Primitive mantle value is denoted by grey squares, labelled PM (McDonough and Sun 1995). The field for mantle peridotites includes abyssal peridotites (Rehkämper *et al.* 1999); Horoman, Japan (Rehkämper *et al.* 1999); Zabargad, Red Sea, and Ronda, Spain, plus fore-arc mantle serpentinites in the Himalayas (Hattori and Guillot 2007). The field for ultramafic cumulates includes data from the Jijal ultramafic complex of the Kohistan Arc, Pakistan, and Talkeetna Arc (Hattori and Guillot 2007). Cumulates of dunites and pyroxenites in Laiwu are from Wang *et al.* (2012a) and dunites in Hujialin from Xie *et al.* (2013). Higashi-akaishi clinopyroxenites and garnet peridotites of an arc cumulate in Dominican Republic (D.R.) are respectively from Hattori *et al.* (2010a) and Hattori *et al.* (2010b). Note that clinopyroxenites plot within or close to the field for cumulates.

represented by $\text{Prp}_{27-36}\text{Alm}_{20-23}\text{Gro}_{40-53}\text{Spe}_{0-1}$ (Prp: pyrope; Alm: almandine; Gro: grossular; Spe: spessartine) in the cores of garnet porphyroblasts in type-II garnet clinopyroxenites. The second population has high grossular component, $\text{Prp}_{17-22}\text{Alm}_{19-21}\text{Gro}_{55-60}\text{Spe}_{1-1}$, and occurs fine-grained relict garnet and exsolution lamellae in clinopyroxene in type-III clinopyroxenites.

5.2.2. Olivine and spinel in clinopyroxenite

Olivine occurs as isolated grains in the matrix and also as inclusions in clinopyroxene in sample HJL6. Olivine grains in the matrix (0.05–0.15 mm) contain low contents of forsterite ($\text{Fo} = 100 \times \text{Mg}/[\text{Mg} + \text{Fe}]$; 79.1–80.7) and NiO (0.18–0.29 wt.%). Olivine inclusions vary in size from 0.02 to 0.10 mm and show low Fo (76.6–76.8) and NiO contents (0.25–0.26 wt.%) (Supplementary Table 4). The compositions of olivine in this study are similar to those of olivine grains from Hujialin clinopyroxenites reported by Hiramatsu and Hirajima (1995) and Yang (2006), and they all plot on a broad curve expected during the fractional crystallization of a mafic melt (Figure 6c). The data suggest that these olivine grains likely represent the early crystallization products from a melt and that clinopyroxenite is likely a cumulate of a mafic melt.

Spinel grains in clinopyroxenite samples are mostly chromian magnetite and magnetite with minor Al-rich spinel and show a wide compositional variation (Supplementary Table 4). Chromian magnetite commonly contains high Fe^{3+} ($Y_{\text{Fe}^{3+}} = \text{Fe}^{3+}/[\text{Fe}^{3+} + \text{Al}^{3+} + \text{Cr}^{3+}]$, mostly >0.75) and Cr_2O_3 contents (12.2–13.4 wt.%), and low X_{Mg} (atomic ratio of $\text{Mg}/[\text{Mg} + \text{Fe}]$, 0.01–0.06) (Figure 6d). Minor green Al-rich spinel occurs at the boundary or within garnet grains in sample HJL7. The composition of green spinel varies in composition from close to $\text{Spl}_{45}\text{Hrc}_{55}$ (Spl: spinel; Hrc: hercynite; Supplementary Table 4) to $\text{Spl}_{61-73}\text{Hrc}_{39-27}$ (Hiramatsu and Hirajima 1995; Yang 2006). Green spinel with a high hercynite component is common in evolved mafic-ultramafic cumulates (e.g. Melcher *et al.* 2002).

6. Discussion

6.1. Origins of Hujialin ultramafic complex

6.1.1. Origins of clinopyroxenite

Bulk compositions of all three types of clinopyroxenite show low concentrations of IPGE compared with the primitive mantle composition where IPGE are at ~0.1

Figure 6. Compositions of diopside in this study and precursor clinopyroxene obtained by Yang (2006) from the Hujialin body plotted in the clinopyroxene discrimination diagram of (a) Loucks (1990) and (b) Le Bas (1962). Arc cumulate field is marked grey. Dashed areas are the fields for clinopyroxene composition of Ural/Alaskan-type ultramafic-mafic intrusions (Helmy and El Mahallawi 2003; Pettigrew and Hattori 2006). (c) Contents of NiO vs. forsterite (Fo) values of olivine from the Hujialin ultramafic complex. The light grey field for most abyssal peridotites (e.g. Dick 1989; Sobolev *et al.* 2005), and Mariana forearc peridotites (Ishii *et al.* 1992), dunites in Hujialin (Xie *et al.* 2013) and peridotites xenoliths represented ancient SCLM and Cenozoic SCLM in the NCC (Zheng *et al.* 2001, 2006; Ying *et al.* 2006; Xu *et al.* 2010; Wang *et al.* 2012b). The expected fractional crystallization curve is marked by an arrow. (d) Compositions of spinel in Hujialin clinopyroxenite samples compared to Mariana forearc peridotites. Thick grey arrow shows the compositional variation of spinel during ferritchromite evolution (Saumur and Hattori 2013). Other symbols are as in (c).

primitive mantle values (Figure 4). These elements have high partition coefficients (D) between mantle minerals and melt (e.g. Puchtel and Humayun 2001; Richter *et al.* 2004) and remain in the residual mantle during partial melting. Therefore, melt and cumulates have low IPGE and IPGE/PPGE (e.g. Hattori and Hart 1997; Wang *et al.* 2012a). The low concentrations of IPGE in the samples suggest that these rocks are not residual mantle rocks. Instead, they are cumulates of melt (Figures 4 and 5). The interpretation is consistent with overall low contents of Ni and Cr in bulk rock samples (Supplementary Table 1). These elements have high D values and tend to remain in the residual mantle.

The Hujialin clinopyroxenite shows low Nb and Zr compared with fluid-mobile elements (i.e. Sr, Ba) and REE, which produce a fractionated primitive mantle-normalized trace element pattern (Figure 3a). The pattern cannot be explained by fractional crystallization of olivine and clinopyroxene (e.g. Hattori *et al.* 2010a). Therefore, the pattern reflects that of the original primitive melt and the source. Fluid-mobile elements and REE have high solubility in aqueous fluids compared with Zr and Nb. The evidence implies that the source mantle for the parental melt had been enriched in fluid-mobile elements. The geochemical data suggest that the source mantle is most likely mantle wedge below

the NCC, and clinopyroxenite represents a cumulate of a melt in the mantle wedge. Variation contents of MgO, Al_2O_3 , TiO_2 , and $\text{Fe}_2\text{O}_3^{\text{total}}$ in the bulk rocks suggest that the parental magmas had undergone variable degrees of differentiation.

Our interpretation based on the bulk rock compositions is consistent with the mineral chemistry of clinopyroxene and olivine. Clinopyroxene contains low Al and Ti and plots in the arc cumulate field (Figure 6a and b). Olivine shows overall low contents of Ni and Mg (Figure 6c), which are too low to be considered as olivine in the residual mantle peridotite. In addition, olivine inclusions in clinopyroxene are common in cumulates, but very rare in mantle peridotites.

The absence of garnet grains in sample HJL6 is likely attributed to lower contents of Al_2O_3 (3.13 wt.%) in bulk rock than garnet-bearing samples ($\text{Al}_2\text{O}_3 \geq 6.95$ wt.%). The clinopyroxene and spinel occur as granular inclusions in garnet porphyroblasts (Figure 2c), suggesting that the garnet cores formed at the expense of these minerals after the solidification of the rock. Furthermore, if garnet was a magmatic phase, the bulk rock with abundant garnet should contain high concentrations of heavy REE as garnet preferentially incorporates heavy REE (e.g. Green *et al.* 2000). Our samples show very low concentrations of heavy REE (i.e. $\text{Yb} = 0.31\text{--}0.81$ ppm; $\text{La}_\text{N}/\text{Yb}_\text{N} > 1.6$) (Supplementary Table 1; see <http://dx.doi.org/10.1080/00206814.2015.1101623> for supplementary tables), suggesting that garnet was not a magmatic crystallization product. The minor positive anomalies of Eu ($\text{Eu}/\text{Eu}^* = 1.08\text{--}1.20$) in the bulk rock of type-I and type-II clinopyroxenite suggest that minor plagioclase likely crystallized as a cumulate phase. The very low $\text{Mg}^\#$ and high contents of $\text{Fe}_2\text{O}_3^{\text{total}}$ and TiO_2 in bulk rock compositions of type-III clinopyroxenite confirm that they were cumulates of evolved magmas.

There are two possible locations in the mantle wedge for the production of subduction-related melt: fore-arc and sub-arc mantle. We suggest that the parental melt for the garnet-bearing clinopyroxenites likely formed in the sub-arc mantle since melt in the fore-arc mantle is characterized by high Mg boninite. Boninitic melt contains high $\text{Mg}^\#$ (>0.80) and low concentrations of incompatible elements such as REE (e.g. Stern and Bloomer 1992). Reflecting the high $\text{Mg}^\#$ of melt, olivine in boninites is characterized by high Fo, commonly over 90.0 (e.g. Walker and Cameron 1983). These characteristics of boninites and boninitic cumulates differ significantly from the observed mineralogy, mineral chemistry, and bulk compositions of clinopyroxenite samples from the Hujialin area. Our samples show low $\text{Mg}^\#$ (0.45–0.85) and high concentrations of incompatible elements in bulk rock, and olivine grains overall

show low Fo (76.6–80.7). In addition, studied samples contain 18–63 times the CI chondrite values of REE (McDonough and Sun 1995), with significant enrichment in light REE and middle REE. Therefore, we conclude that the melt for our ultramafic cumulates was not likely generated in the fore-arc mantle, but rather in the sub-arc mantle.

The proposed origin for the Hujialin clinopyroxenite is in contrast to the proposed protolith of the Bixiling ultramafic-mafic rocks in the southern Dabie terrane. The Bixiling rocks show incompatible trace element patterns comparable to mid-ocean ridge basalts and essentially no enrichment in fluid-mobile elements (i.e. Ba, Sr) compared with Nb and Zr ($\text{Ba}_\text{N}/\text{Nb}_\text{N} = 1.0\text{--}1.7$ except for sample B4-1) (Zheng *et al.* 2008). However, the Hujialin clinopyroxenites are markedly enriched in fluid-mobile elements relative to Nb and Zr ($\text{Ba}_\text{N}/\text{Nb}_\text{N} = 4.4\text{--}22.7$), confirming the formation of their parental melt in the sub-arc mantle. The geochemical signature of the Hujialin clinopyroxenites is distinctly different from that of the Bixiling ultramafic-mafic rocks (Figure 3), reflecting different protoliths for the two. Furthermore, the U–Pb and Hf–isotope data of the Bixiling complex suggest the crystallization age in Neoproterozoic time (ca. 745 Ma) (Zheng *et al.* 2008). There are many mafic igneous rocks of similar age in the YZC due to a mantle plume related to the break-up of Rodinia (e.g. Li *et al.* 2003). All data suggests that the Bixiling complex was formed as cumulates of an asthenosphere-derived melt in the YZC and later subducted together with the YZC to ca. 120 km (Zheng *et al.* 2008).

In summary, the protolith of Hujialin clinopyroxenite is most likely cumulates of a subduction-related melt, and the primary magmas were probably generated in the sub-arc mantle and underwent various degrees of magmatic differentiation. We suggest the following sequence of mineral crystallization before they were incorporated into the subduction channel together with the down-going slab. Clinopyroxene, spinel, minor olivine, and negligible plagioclase solidified from a mafic melt as cumulate phases at shallow depth of the mantle wedge where spinel peridotite is stable. Once they were incorporated into the subduction channel, they were metamorphosed to form Ca- and Al-rich garnet after spinel and very minor plagioclase. During the exhumation, porphyroblastic clinopyroxene would have exsolved garnet, diopside, and Fe oxides (magnetite and ilmenite).

6.1.2. Origins of peridotites in the Hujialin ultramafic complex

Dunite samples in the Hujialin area, which are partially hydrated (LOI; 6.6–13.2 wt.%), contain high concentrations

of compatible elements (Cr, Co, and Ni) and IPGE (5.0–22.7 ppb in total) with high ratios of IPGE/PPGE in bulk rocks. Cr-spinel shows high Cr[#] (0.68–0.76) and olivine has high Fo (91.7–92.4) (Xie *et al.* 2013). The data confirm that Hujialin hydrated dunite represents mantle wedge peridotite at a relatively shallow depth in a spinel-stable field below the southern margin of the NCC, and that it is most likely a relict of ancient subcontinental lithospheric mantle. Serpentinites and harzburgite in the Hujialin ultramafic complex contain high concentrations of compatible elements (Mg and Cr) and low concentrations of incompatible elements (Al and Ti), with high Mg (Fo = 91.7–92.2) and Ni contents in olivine and high Cr (Cr[#] = 0.68) contents in Cr-spinel in the harzburgite rocks, and they are interpreted to have derived from a depleted mantle wedge (Yang 2006).

6.2. Tectonic setting of Hujialin ultramafic rocks

6.2.1. Tectonic evolutions of Hujialin ultramafic rocks

The Hujialin garnet-bearing clinopyroxenite formed through five stages: (1) a partial melting in the mantle wedge below a continental arc on the southern margin of the NCC; (2) crystallization of clinopyroxenite from the slightly evolved melt; (3) near-horizontal motion by the mantle corner flow towards the subduction channel and incorporation into the subduction channel; (4) UHP metamorphism; and (5) exhumation together with granitic gneisses.

The pressure and temperature conditions of the crystallization of the clinopyroxenite are difficult to evaluate due to the lack of primary clinopyroxene. Yang (2006) speculated that the clinopyroxenites may have crystallized at a depth near the crust–mantle boundary based on the presence of Al-spinel. Stage 3 is most likely to have taken place in the early stage of the subduction of the YZC when the friction between the newly subducting craton and the overlying plate was sufficient to create a strong mantle flow (e.g. Zhang *et al.* 2009a). The cumulates of the parental melt had to be dislodged from the sub-arc lithospheric mantle, and transported towards the subduction plate, which has a horizontal distance of more than 100 km in most zones (e.g. Syracuse and Abers 2006). Such processes require a strong mantle flow between the subducting and overlying plates and a rheologically weak mantle wedge (Arcay *et al.* 2007). Therefore, the flow is faster and stronger where the interface between the subducting and overlying plates is poorly lubricated. Therefore, we suggest that the horizontal transport of cumulate from the sub-arc to the subduction channel likely took place in the early stage of

subduction of the YZC. The proposed interpretation is consistent with the anticlockwise P–T paths for Hujialin clinopyroxenite reported by Yang (2006) and Chen and Xu (2005). An anticlockwise path is not common in subduction zones, but is expected during the initiation of subduction before a subduction zone has reached a thermally steady-state condition (e.g. Krogh *et al.* 1994).

Stage 4 is defined by zircon U–b ages (~220 Ma) of Hujialin garnet-bearing clinopyroxenites as reported by Gao *et al.* (2004) and the exsolution (garnet, clinopyroxene, magnetite, and ilmenite) of clinopyroxene during the exhumation (Zhang and Liou 2003; Li *et al.* 2008). The peak P–T conditions (P ≥ 5.0 PGa; T ≥ 750°C) were estimated by Chen and Xu (2005) and Yang (2006) based on the Fe–Mg exchange of coexisting garnet and clinopyroxene. The conditions are equivalent to a depth close to 150 km. The maximum physical conditions of Hujialin clinopyroxenites are similar to adjacent UHP eclogites and garnet peridotites which recorded ≥700°C and ≥4.0 PGa (e.g. Zhang *et al.* 2000, 2009b), suggesting a common P–T history during deep continental subduction. During their exhumation, Hujialin garnet-bearing clinopyroxenites were subjected to minor degrees of retrogression to form amphibole and chlorite at shallow crustal levels.

The highly refractory geochemical characteristics of Hujialin spinel peridotites suggests their original site in the mantle wedge. Two questions arise: how lenses of garnet-bearing clinopyroxenite came in contact with spinel-bearing peridotite lenses and how the Hujialin ultramafic complex was incorporated into the UHP metamorphic belt and exhumed to the surface. Two possible tectonic evolution models are discussed below.

The first model suggests the same path for clinopyroxenites and peridotites during their dragging to the subduction channel, subduction, and eventual exhumation. The protolith of Hujialin clinopyroxenite, together with the surrounding anhydrous peridotites in the mantle wedge, may have been incorporated into the continental subduction channel along the interface between the subducting YZC and the overlying NCC. Both underwent the deep subduction process. During their exhumation, the spinel-bearing peridotites were partially to totally hydrated by water released from the subducted continent slabs, and formed lenses and surrounded garnet-bearing clinopyroxenite (possibility 1 in Figure 7a). In this model, the lack of metamorphic garnet in peridotites is attributed to very low contents of Al and Ca in the bulk rock compositions (Xie *et al.* 2013). Hattori *et al.* (2010b) found similar phenomena in peridotites in the subduction complex of the northern Dominican Republic. Peridotites with high Al

Figure 7. Schematic of two possible models for the subduction and exhumation of the Hujialin ultramafic complex in the continental subduction channel, based on the numerical models of Warren *et al.* (2008) and Li and Gerya (2009). (a) Clinopyroxenites were mingled with anhydrous peridotites in the mantle wedge, and they underwent HP-UHP metamorphism in the subduction process. The ultramafic complex was partially to totally hydrated by water released from the subducted continent slabs at shallow levels in the exhumation process. (b) Clinopyroxenites were individually brought into the subduction channel and underwent HP-UHP metamorphism to crystallize garnet. Hydrated dunite and the surrounding serpentinized peridotite body were not subducted to deep levels, and were physically mixed with deeply subducted clinopyroxenites and granites at shallow levels during their exhumation. U. crust = upper crust; L. crust = lower crust.

crystallized garnet, but not those with low Al, did not crystallize garnet even though both have undergone HP metamorphism.

An alternative model suggests separate paths for clinopyroxenites and peridotites. The fragments of clinopyroxenite in the sub-arc mantle may have been incorporated into the subduction channel by a mantle flow and underwent HP-UHP metamorphism to crystallize garnet. During its exhumation in the return flow of the subduction channel, it was physically mixed with

serpentinized peridotite lenses off-scraped from the overlying SCLM wedge. Then, both exhumed to shallow levels together with granitic rocks (possibility 2 in Figure 7b).

The Suoluoshu mafic-ultramafic complex, very close to the Hujialin ultramafic complex (Figure 1b), is also dominated by mantle-derived serpentinized peridotites, containing lenses of garnet-bearing pyroxenite, eclogite, and dunite (e.g. Gong *et al.* 2005). Similar lithological association and occurrence suggest that the two had a similar tectonic evolution.

Most ultramafic complexes in the Dabie-Sulu UHP terrane are commonly composed of garnet peridotite with various proportions of spinel peridotite (dunite, harzburgite, and minor lherzolite) and garnet pyroxenite, such as the Donghai and Yangkou areas of the southern-central Sulu belt (e.g. Zhang *et al.* 2000; Ye *et al.* 2009) and the Raobazhai area of the northern Dabie orogen (Tsai *et al.* 2000; Zheng *et al.* 2008). The garnet peridotite and spinel peridotite are mostly interpreted to be fragments of the ancient SCLM beneath the NCC (Zhang *et al.* 2000; Yang *et al.* 2009; Ye *et al.* 2009), whereas garnet-bearing pyroxenites commonly occur as irregular bodies or veins in various sizes of ultramafic complexes, and their protoliths commonly represent the cumulates of mafic magma (Zheng *et al.* 2008; Zhang *et al.* 2009b). In addition, the garnet peridotites and garnet pyroxenites, as discontinuous lenses founded in gneiss and eclogite bodies of CCSD-MH and associated drill holes (Figure 1 in Zhang *et al.* 2010), are considered to be cumulates formed by differentiation of mafic magmas. Similar ultramafic complexes also occurred in the Bixiling and Maowu areas, southern Dabie orogen (e.g. Zheng *et al.* 2008). The tectonic setting and petrogenesis of ultramafic rocks in the Dabie-Sulu UHP terrane suggest the continental subduction channel between the subducting plate and the overlying mantle wedge is mostly composed of a mélangé that consists of ultramafic lithologies such as metamorphic and/or altered peridotite and pyroxenite from the upper part and HP-UHP lithologies (granitic gneisses, eclogites, and metasediment) from the lower part (e.g. Zheng 2012).

6.2.2. Possible causes for the exhumation of Hujialin ultramafic rocks

Oceanic and/or continental lithosphere were subducted to a deep level forming HP-UHP metamorphic rocks in convergent zones (Ernst 2001), and the exhumation processes are still under debate (e.g. Guillot *et al.* 2009; Zheng 2012). The proposed driving forces of exhumation include buoyancy, compression of a soft zone between two rigid blocks, soft serpentinite channel, and coaxial extension associated with a decoupling fault (e.g. Chemenda *et al.* 1995; Guillot *et al.* 2009). The exhumation of HP-UHP metamorphic rocks in oceanic subduction zones is attributed to a wide serpentinite-rich subduction channel in a mechanically weakened mantle wedge (e.g. Guillot *et al.* 2000; Gorczyk *et al.* 2007), but this is not applicable to the Dabie-Sulu UHP terrane because it is mostly composed of granitic orthogneisses (≥ 90 vol.%) with minor (< 1 vol.%) serpentinitized peridotites (e.g. Zhang *et al.* 2000; Zheng 2008).

The southern margin of the NCC was an active continental margin in response to the subduction of Neotethys until the collision of the two continents. The closure of the ocean resulted in the collision followed by the subduction of the YZC below the NCC to form a continental subduction channel between the two plates. The Hujialin garnet-bearing clinopyroxenite and serpentinitized peridotite were incorporated into the subduction channel, a mélangé made up of blocks of different lithologies from the two plates. Numerical models by Warren *et al.* (2008) and Li and Gerya (2009) show the buoyancy of granitic rocks would result in the separation of granitic blocks from the rest of subducted lithosphere and exhumation to the surface in the return flow of the subduction channel. Volumetrically minor eclogites and ultramafic rocks were likely exhumed together with the voluminous granitic gneiss. The interpretation is consistent with the similar isothermal decompression paths observed in the majority of HP-UHP rocks including granitic gneisses, eclogites, garnet peridotites, and garnet pyroxenites in the Dabie-Sulu UHP terrane (e.g. Yao *et al.* 2000; Zheng 2008; Ye *et al.* 2009; Zhang *et al.* 2009a).

Conclusions

Bulk rock and mineral compositions suggest that Hujialin clinopyroxenites are cumulates of a subduction-related melt that underwent various degrees of magmatic differentiation in the sub-arc lithospheric mantle. The primary minerals of clinopyroxenite are clinopyroxene (augite), spinel, minor olivine, and negligible plagioclase before the UHP metamorphism.

Two possible tectonic evolution models are proposed for the Hujialin ultramafic complex. Clinopyroxenite and spinel peridotite were once below the sub-arc mantle and both were incorporated into the continental subduction channel and underwent the UHP metamorphism. The lack of garnet in peridotite may be explained by low Al and Ca in the rocks. Alternatively, clinopyroxenite and peridotites were not subducted together to deep levels. Clinopyroxenite was subducted deeply to form garnet and exhumed together with granitic gneiss. It was physically mixed with peridotite at relatively shallow depths during exhumation.

The density-induced decoupling of voluminous granitic orthogneisses from the rest of the subducted lithosphere resulted in the upward movement of granitic gneisses, which brought the deep clinopyroxenites and spinel peridotites to shallow crustal depths. The process explains the physical mixture of peridotite and pyroxenite within voluminous granitic rocks of the Dabie-Sulu UHP terrane.

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