1	Whole-rock and zircon evidence for evolution of the Late
2	Jurassic high Sr/Y Zhoujiapuzi granite, Liaodong Peninsula,
3	North China Craton
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15	Abstract: Middle-Late Jurassic high Sr/Y granitic intrusions are extensively exposed
16	in the Liaodong Peninsula, in the eastern part of the North China Craton (NCC).
17	However, the genesis of the high Sr/Y signature in these intrusions has not been studied
18	in detail. In this study, we report results of zircon U-Pb dating, Hf isotopic analysis and
19	zircon and whole-rock geochemical data for the Late Jurassic Zhoujiapuzi granite in
20	the middle part of the Liaodong Peninsula. The Zhoujiapuzi granite is high-K (calc-
21	alkaline) and peraluminous in nature, with high SiO <sub>2</sub> (68.1–73.0 wt %) and Al <sub>2</sub> O <sub>3</sub>
22	(14.5–16.8 wt %), low in TFe <sub>2</sub> O <sub>3</sub> (1.10–2.49 wt %) and MgO (0.10–0.44 wt %), and

with high Sr/Y (19.9–102.0) and La<sub>N</sub>/Yb<sub>N</sub> (14.59–80.40), characteristic of high Sr/Y I-23 type granite. The geochemical signatures, in combination with the presence of a large 24 25 number of Paleoproterozoic inherited zircons, indicate that the Zhoujiapuzi granite was most likely derived from partial melting of the basement in the region, and specifically 26 the Liaoji granites. The high Sr/Y signature is inherited from these source rocks. LA-27 ICP-MS zircon U-Pb dating of the autocryst zircons from two samples (from different 28 localities) yielded consistent weighted average ages of 160.7±1.1 Ma (MSWD=1.3) and 29 159.6 $\pm$ 1.1 Ma (MSWD=1.2), with  $\epsilon$ Hf(t) values in the range of -26.6– -22.8. 30 31 Morphological and chemical studies on autocrystic zircon grains show that there are two stages of zircon growth, interpreted as magmatic evolution in two distinct stages. 32 The light-CL core reflects a crystallization environment of low oxygen fugacity and 33 34 high T<sub>Zr-Ti</sub>; the dark-CL rim formed with high oxygen fugacity and lower T<sub>Zr-Ti</sub>. Based on the geochemical features and regional geological data, we propose that the Liaodong 35 Peninsula in the Late Jurassic was part of a mature continental arc, with extensive 36 37 melting of thick crust above the Paleo-Pacific subduction zone.

Keywords: Liaodong Peninsula; Late Jurassic; Zircon U-Pb-Hf isotopes; Two stages
of crystal growth; High Sr/Y granite

#### 40 1. Introduction

The Liaodong Peninsula is located in the northeast of the North China Craton (NCC). The northeast NCC was influenced by three main tectonic regimes in the Mesozoic, related to the subduction of the Paleo-Asian, Paleo-Pacific and Mongol-Okhotsk oceans (Tang et al., 2018). The superposition of these different regimes resulted in changing tectonic and magmatic patterns over time. Middle-Late Jurassic
granitic rocks are extensively exposed in the northern parts of the Liaodong Peninsula,
such as the Yutun mylonitic granite, Xiaoheishan granodiorite, Heigou monzogranite
(Wu et al., 2005), Wulong two-mica monzogranite (Yang et al., 2018), and Huangdi
biotite monzogranite (Xue et al., 2020). Most of these rocks are characterized by high
Sr /Y, and plot within the adakite field on Sr/Y-Y and La<sub>N</sub>/Yb<sub>N</sub>-Yb<sub>N</sub> diagrams (Wu et
al., 2005a; Yang et al., 2015a, 2018).

The geodynamic settings and petrogenesis of adakite and geochemically similar 52 53 high Sr/Y igneous rocks have been widely discussed. The high Sr/Y rocks were originally proposed to be formed by melting of young (<25 Ma) and hot subducted 54 oceanic slab in an arc setting (Defant and Drummond, 1990). However, later studies 55 56 have shown that the high Sr/Y rocks can form in both arc and non-arc settings by other processes, such as continental interior settings (Wang et al., 2007), cold subduction 57 zones (Nakamura and Iwamori, 2013), collision or post-collision processes (Schwartz 58 et al., 2011). In addition, numerous studies have suggested that the lower continental 59 crust can also be the source of the high Sr/Y rocks (Gao et al., 2004; Ou et al., 2017). 60 However, it is debated whether crustal thickening is necessary for their formation (e.g. 61 Moyen, 2009; Kamei et al., 2009; Zhan et al., 2020). In recent years, some studies have 62 proposed that the high Sr/Y ratio in granitic rocks can be inherited from a high Sr/Y 63 crust source, regardless of pressure (Kamei et al., 2009; Ma et al., 2015; Zhan et al., 64 2020). 65

The Middle-Late Jurassic granitic rocks in the Liaodong Peninsula are commonly proposed to be the products of partial melting of thickened mafic crust with garnet in the residue (Wu et al., 2005a; Yang et al., 2015a, 2018; Tang et al., 2018). However, the source composition has not been fully considered in the petrogenesis of the high Sr/Y rocks in the Liaodong Peninsula. Hence, the petrogenesis of the Middle-Late Jurassic
high Sr/Y rocks needs to be re-evaluated, based on more detailed work and a
consideration of possible sources. This petrogenesis is of significance for understanding
the Jurassic tectonics of the Liaodong Peninsula, and the NCC in general.

In this paper, we examined the high Sr/Y Zhoujiapuzi granite from the Xiuyan area, 74 in the middle of the Liaodong Peninsula. Zircons are analysed for U-Pb-Hf isotopes 75 76 and trace element geochemistry, and by Raman spectroscopy. These results are integrated with whole-rock geochemistry. We focus on the zircons, because of their 77 78 potential to reveal the origins of the pluton (Belousova et al., 2002; Wang et al., 2007; Breiter et al., 2014; Zhao et al., 2014), and so provide a case study for the evolution of 79 plutonic magma systems in general. Based on observations of the CL images and 80 chemical analysis, two zircon growth stages can be distinguished. We first determine 81 the crystallization environments of the two zircon growth stages, and then decipher the 82 petrogenesis, source characteristics and origin of the high Sr/Y signature of the pluton 83 as a whole. Integrated with previous studies, our study provides insights into the 84 tectonic evolution of the Liaodong Peninsula in the Late Jurassic. 85

# 86 2. Geological setting

The Zhoujiapuzi granite is located in the middle of the Liaodong Peninsula, at the northeastern margin of the NCC (Fig. 1). The Paleoproterozoic Liaohe Group and Liaoji granite are the basement in the study area. The Liaohe Group includes the Lieryu, Gaojiayu, Dashiqiao and Gaixian formations. Although stratigraphic terms are used, these rocks are metamorphic, and the group consists of leptynite, leptite, granulite, amphibolite, marble and phyllite. The protoliths of the Liaohe Group include marine volcanics, clastics, carbonates and claystones. The formation age of the
metasedimentary rocks in the Liaohe Group is 2.0–1.9 Ga (Wan et al., 2006; Li et al.,
2015). It is in unconformable contact with the overlying strata of the Mesoproterozoic
Cuocaogou Formation and Xiaoling Formation.

The study area experienced strong magmatic activity in the Paleoproterozoic, 97 which can be divided into two stages of 2.2–2.1 Ga and  $\sim$  1.85 Ga. The 2.18–2.14 Ga 98 Liaoji granites (also called gneissic granites), which lie within an area measuring 300 99 km × 70 km, are dominated by A- and I-type granites (Li and Zhao, 2007; Yang et al., 100 101 2016; Wang et al., 2020a). Metamorphosed volcanic rocks (leptynite, leptite and granulite) in the Liaohe Group also formed at 2.2–2.1 Ga (Li et al., 2015). The ~1.85 102 Ga granites mainly consist of I- and S-type porphyry granites and alkaline syenites 103 104 (Yang et al., 2007; Yang et al., 2015b). In addition, there were small amounts of mafic magmatic activity at ~2.17 Ga, ~2.1 Ga and ~1.8 Ga (Meng et al., 2014; Yuan et al., 105 2015). There are a variety of viewpoints on the Paleoproterozoic tectono-magmatic 106 107 evolution in the Liaodong Peninsula, such as an intracontinental rift opening-closing model (Li et al., 2005) and an arc-continent collision model (Faure et al., 2004). 108

In the Mesozoic, the region of the Liaodong Peninsula was influenced by the circum-Pacific tectonic regime, the Mongol-Okhotsk tectonic regime and the Paleo-Asian Ocean tectonic regime. The joint influence of multiple tectonic regimes resulted in intensive magmatism during the Mesozoic (Fig. 1b). These Mesozoic magmatic rocks can be divided into three stages, namely: Triassic (233–212 Ma), Jurassic (180–

114 156 Ma) and Early Cretaceous (131–117 Ma) (Wu et al., 2005b).

The Triassic magmatic rocks are less exposed, mainly alkaline rocks, diabase, 115 diorites and granites (Wu et al., 2005b). Among them, the granites mainly have A-type 116 affinity, and may have formed in an extensional setting (Tang et al., 2018; Wang et al., 117 2019). Magmatism has been related to either the subduction of the Paleo-Pacific slab, 118 closure of the Paleo-Asian Ocean, or the collision between the NCC and the Yangtze 119 Craton (Tang et al., 2018; Wang et al., 2019). The majority of the Jurassic magmatic 120 rocks are monzogranite and granodiorite, which are generally calc-alkaline I-type 121 granites, and show characteristics of adakite-like rocks. Some of them, exposed near 122 123 later extensional structures, have undergone regional ductile deformation. These Jurassic magmatic rocks are generally considered to relate to the subduction of the 124 Paleo-Pacific slab (Wu et al., 2005a; Zhai et al., 2004). In the Early Cretaceous, basic-125 126 acidic-alkaline rocks were widely developed. Among them, the granites have mainly A- and I-type affinities. These rocks are generally considered to have formed in an 127 intense extensional environment, which is connected with either the rollback or low-128 angle subduction of the Paleo-Pacific slab (Wu et al., 2005c; Zheng et al., 2018). 129

#### 130 **3. Samples and petrography**

The Zhoujiapuzi granite is located to the east of Xiuyan City, in the middle of the
Liaodong Peninsula (Fig. 1b). It intruded into the Lieryu Formation of the Liaohe Group.
Eight samples of the Zhoujiapuzi granite were collected at locations shown in Fig. 1c.
The Zhoujiapuzi granite is generally grey in colour and with fine-grained texture
(Fig. 2a). The mineral assemblage contains K-feldspar (~50 %), quartz (~25 %),

plagioclase (~20 %) and biotite (~5 %) as well as accessory minerals such as zircon,
ilmenite, magnetite and apatite. K-feldspar grains are euhedral or subhedral, and always
exhibit cross-hatched twinning (Fig. 2b). Quartz grains are usually xenomorphic, and
have indented boundaries and wavy extinction (Fig. 2b-d). Plagioclase always exhibits
polysynthetic twinning and have sericitization in places (Fig. 2c). Biotite mainly fills
in the interstices between the other minerals (Fig. 2c, d).

#### 142 4. Analytical methods

The cathodoluminescence (CL) images of zircon were obtained by the Chengpu 143 geological Testing Co. Ltd, Langfang, China using the TIMA analysis. The LA-ICP-144 MS zircon U-Pb analyses were performedusing an Agilent Technologies 7700x ICP-145 MS with a Teledyne Cetac Technologies Analyte Excite laser-ablation system at 146 Nanjing FocuMS Contract Testing Co. Ltd. The analyses were carried out with a 35 µm 147 spot size at 8 Hz repetition rate for 40 seconds. The ICP-MS detector has dual modes: 148 149 pulse for lower signal, and analog for higher signal. Pulse-analog cross calibration was performed before the measurement of U-Pb isotopes, delivering a wider linear dynamic 150 range – up to 10 orders of magnitude. For a signal of  $^{238}$ U higher than 1.2–1.4 Mio cps, 151 equivalent zircon contains U concentrations higher than 600 ppm, and are measured in 152 analog mode. 91500 was used as external standard. GJ-1 (600Ma, Jackson et al., 2004) 153 and Plešovice (337Ma, Sláma et al., 2008) were treated as quality control for 154 geochronology. During our analyses, the weighted mean age of GJ-1 and Plešovice 155 were  $606.0 \pm 4.8$  Ma (n=16, MSWD = 0.50) and  $340.9 \pm 4.0$  Ma (n=7, MSWD = 1.0), 156 respectively. Trace elements abundance of zircon were externally calibrated against 157 NIST SRM 610 with Si as the internal standard. The raw ICP-MS data were processed 158

using ICPMSDataCal software (Liu et al., 2010). No common-Pb correction was
applied to the data. Data reduction was completed using the Isoplot4.15 (Ludwig, 2003).
The instrument description and analytical procedure are described in detail by Zeng et
al. (2018).

The in-situ Lu-Hf isotopic analyses of zircon were performed by LA-MC-ICP-MS 163 using a Teledyne Cetac laser-ablation system and a Nu Plasma II MC-ICP-MS at 164 Nanjing FocuMS Contract Testing Co. Ltd. The 193 nm ArF excimer laser was focused 165 on zircon surface with fluence of 6.0J/cm<sup>2</sup>. The ablation protocol employed a spot 166 diameter of 50 um at 8 Hz repetition rate for 40 seconds. Three standard zircons, GJ-1, 167 91500, and Penglai, were analysed for quality control at every ten unknown samples. 168 In the experiment, standard zircon GJ-1, 91500, and Penglai were analyzed, and the 169  $^{176}\mathrm{Hf}/^{177}\mathrm{Hf}$  ratios were 0.282002–0.282013, 0.282305–0.282315 and 0.282901– 170 0.282914 respectively, in accordance with their recommended values (GJ-1: 0.282012, 171 Yuan et al., 2008; 91500: 0.282307 ± 0.000031, Wu et al., 2006; Penglai: 0.282906 ± 172 0.000010, Li et al., 2010). For the calculation of  $\varepsilon$ Hf(t) values, we have adopted the 173  $^{176}\text{Lu}$  decay constant of 1.867  $\times$   $10^{-11}$  (Söderlund et al., 2004), the present-day 174 chondritic values of  ${}^{176}Lu/{}^{177}Hf = 0.0332$  and  ${}^{176}Hf/{}^{177}Hf = 0.282772$  (Blichert-Toft and 175 Albarède 1997). To calculate one-stage model ages (T<sub>DM1</sub>) relative to a depleted-mantle 176 source, we have adopted the present-day depleted-mantle values of  ${}^{176}Lu/{}^{177}Hf =$ 177 0.0384 and  ${}^{176}\text{Hf}/{}^{177}\text{Hf} = 0.28325$  (Vervoort and Blichert-Toft 1999). To calculate two-178 stage modal ages (TDM2), 'felsic crust' model ages are calculated using average 179 continental crust  ${}^{176}Lu/{}^{177}Hf = 0.015$  (Griffin et al., 2004) 180

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Zircon Raman analyses were carried out using an RM2000 laser Raman

spectrometer at the State Key Laboratory of Nuclear Resources and Environment, East
China University of Technology. The selected incident wavelengths were 532 and 785
nm in order to clearly identify the luminescence bands due to low concentration
impurities. The beam power was 20 mW. The Leica 50× objective was employed.

Six fresh rock samples were selected for geochemical analysis. The elemental 186 analyses were conducted at Analytical Chemistry & Testing Services (ALS) Chemex 187 (Guangzhou) Ltd. Major oxides were analyzed using wave-dispersive X-ray 188 fluorescence (XRF) (ME-XRF26). Analytical precision was better than  $\pm$  0.01%. 189 190 Trace element abundances were measured by the lithium borate dissolution method and ICP-MS (ME-MS81). The analytical uncertainties of the rare earth element (REE) and 191 high field strength element (HFSE) are <5%. Analytical uncertainties are in the range 192 193 of 5%–10% for the other elements. Detailed analytical procedures refer to Zhang et al. (2019) and Nash et al. (2020). 194

#### 195 5. Analytical results

The data for major and trace elements, Raman microprobe data, zircon trace elements, zircon U-Pb ages, and zircon Hf isotopes are shown in Tables S1, S2, S3, S4 and S5, respectively.

## 199 5.1. Whole-rock major and trace element compositions

SiO<sub>2</sub> contents range from 68.11 wt.% to 73.02 wt.% (average 71.71 wt.%). Contents of Na<sub>2</sub>O and K<sub>2</sub>O are 3.81 - 4.65 wt.% and 4.32 - 4.71 wt.%, respectively, with Na<sub>2</sub>O/K<sub>2</sub>O ratio of 0.82 - 1.08 and total alkalis (Na<sub>2</sub>O + K<sub>2</sub>O) of 8.38 - 8.97. All

samples plot in the field of granite in the TAS classification except one (Fig. 3a). These 203 samples have Al<sub>2</sub>O<sub>3</sub> contents of 14.49 - 16.83 wt.% (average 15.09 wt.%), CaO 204 contents of 1.04 - 1.98 wt.% (average 1.38 wt.%) and A/CNK values of 1.05 - 1.10 205 (average 1.07). In the A/NK - A/CNK diagram (Fig. 3b), all samples plot in the 206 peraluminous field (Fig. 3b). The granite samples have low  $TFe_2O_3$  ( $TFe_2O_3 = all Fe$ 207 calculated as Fe<sub>2</sub>O<sub>3</sub>) contents and MgO contents ranging from 1.10-2.49 wt % and 208 0.10-0.44 wt %, respectively, with Mg# (Mg#=100\*molar Mg/(Mg+Fe)) values of 15-209 26. 210

The samples of the Zhoujiapuzi granite exhibit variable REEs, with total REEs 211 ranging from 59 to 302 ppm. The La<sub>N</sub>/Yb<sub>N</sub> values of the Zhoujiapuzi granite range from 212 14.59 to 80.40 (average 38.27), showing right-declined REE patterns (Fig. 4a). The 213 214 samples have Eu/Eu\* of 0.62–1.94 and Ce/Ce\* of 0.94–1.16. In the primitive mantlenormalized trace element diagram (Fig. 4b), the samples show negative anomalies of 215 216 HFSEs (e.g., Nb, Ta, Ti and P) and positive anomalies of La and LILEs (e.g., K, Rb, Ba, U, La, Ce). The Zhoujiapuzi granite is characterized by high contents of Sr (309-217 551 ppm) and low contents of Y (5.01–15.5 ppm) and Yb (0.43–1.40 ppm), with high 218 Sr/Y ratios of 19.94–102.04 (average 65.50). 219

# 220 5.2 Zircon CL images, Raman spectra and REE elements

CL images of zircons from the Zhoujiapuzi granite are shown in Fig. 5. Zircons commonly have crystal sizes between 150 and 250 μm, and have length/width ratios of 2:1–4:1, with euhedral, stubby to elongate prisms. According to the CL images, most zircons show an internal division into 2 distinct domains: light-CL core and dark-CL rim. The light-CL core is characterized by bright CL intensity and widely-spaced oscillatory zoning patterns. The dark-CL rim is overgrown continuously by the lightCL core and is characterized by extremely low CL emission and narrowly-spaced
oscillatory zoning patterns. In addition, some zircons have inherited cores, which have
corroded and rounded shapes in contact with the light-CL core, such as 1# and 37# in
XY-001 and 6# and 41# in XY-008 (Fig. 5). These inherited zircons have oscillatory
zoning in CL images.

Six light-CL core spots and six dark-CL rim spots were analyzed for Raman spectra. The light-CL cores have antisymmetric stretching vibration ( $B_{1g}$ ) of the SiO<sub>4</sub> tetrahedra ( $v_3$  (SiO<sub>4</sub>)) Raman band of 1005–1007 cm<sup>-1</sup> and half-width of the  $v_3$  (SiO<sub>4</sub>) Raman band (*b*) values of 6.0–8.1 cm<sup>-1</sup>, while the dark-CL rims have  $v_3$  (SiO<sub>4</sub>) Raman band of 1004–1007 cm<sup>-1</sup> and *b* values of 5.4–9.0 cm<sup>-1</sup>.

Twenty light-CL core spots, eighteen dark-CL rim spots and six inherited zircon 237 spots were analyzed for trace and rare earth elements. The light-CL core spots have 238 lower U content (28–677 ppm) than the dark-CL rim spots (U=641–3842 ppm). In the 239 chondrite-normalized REE element diagram (Fig. 6a, b), both the light-CL core and 240 dark-CL rim are characterized by HREE enrichment relative to LREE with positive Ce 241 anomalies and negative Eu anomalies. The light-CL core spots have  $\Sigma REE$  of 49–1115 242 ppm (average 390 ppm),  $\Sigma LREE$  of 3–72 ppm (average 14 ppm) and  $\Sigma HREE$  of 46– 243 1100 ppm (average 377 ppm), whereas the dark-CL rim spots have  $\Sigma REE$  of 327–1632 244 ppm (average 895 ppm),  $\Sigma LREE$  of 2–14 ppm (average 6 ppm) and  $\Sigma HREE$  of 325– 245 1627 ppm (average 889 ppm). Hence, the REE content of the light-CL core is 246 significantly lower than that of the dark-CL rim, and the difference between the two is 247 mainly in HREE content. The light-CL core spots have Eu/Eu\* of 0.07-0.60 (average 248 0.28) and Ce/Ce\* of 1.89–24.27 (average 10.03). Because the contents of La and Pr are 249 typically present very low, Ce\* in this study is obtained by the formulation  $(Nd_N)^2/Sm_N$ 250

(Loader et al., 2017). The dark-CL rim spots have Eu/Eu\* of 0.08–0.24 (average 0.13) and Ce/Ce\* of 6.57–200.31 (average 79.23). These results indicate that the light-CL core have a weaker negative Eu anomaly and a weaker positive Ce anomaly than those of the dark-CL rim. The inherited zircon spots have  $\Sigma$ REE of 602–1517 ppm, and show depletion of LREE, enrichment of HREE, a positive Ce anomaly (Ce/Ce\* of 1.52– 216.08) and a negative Eu anomaly (Eu/Eu\* of 0.07–0.13) (Fig. 6c).

#### 257 **5.3 Zircon U–Pb and Hf isotope composition**

Seventy-seven spots were analysed for U-Pb isotope composition from samples 258 XY-001 and XY-008. In the U-Pb Concordia diagram (Fig. 7a, c), both the light-CL 259 core and dark-CL rim spots overlap within uncertainty on the Concordia curve. There 260 is a large degree of overlap between the dark-CL rim and light-CL core in terms of 261 <sup>206</sup>Pb/<sup>238</sup>U age although the mean value for <sup>206</sup>Pb/<sup>238</sup>U age is higher in the light-CL core 262 (Fig. 7e). On a single zircon, the <sup>206</sup>Pb/<sup>238</sup>U age of the light-CL core is older than that 263 of the dark-CL rim (Fig. 5). In sample XY-001, 33 spots define a weighted mean 264  $^{206}$ Pb/ $^{238}$ U age of 160.7±1.1 Ma (2 $\sigma$ , MSWD=1.3; Fig. 7b). In sample XY-008, 28 spots 265 define a weighted mean  ${}^{206}$ Pb/ ${}^{238}$ U age of 159.6±1.1 Ma (2 $\sigma$ , MSWD=1.2; Fig. 7d). The 266 other 10 spots with distinctly older ages (207Pb/206Pb ages ranging from 2500 to 2173 267 Ma) were obtained on inherited cores. Their ages are discordant, suggesting that these 268 inherited cores were variably influenced by lead loss. Among these, 9 spots define a 269 discordia line with an upper intercept age of  $2163 \pm 13$  Ma (MSWD=0.45) (Fig. 7f). 270

Twenty-four zircons were analyzed for Lu-Hf isotope composition. The variation in Hf isotopic data is limited, between 9 spots from light-CL core and 9 spots from dark-CL rim. 18 spots exhibit a range of  $^{176}$ Hf/ $^{177}$ Hf ratios from 0.281921 to 0.282030, which converts to  $\epsilon$ Hf(t) values between -26.6 to -22.8 (Fig. 8), and two-stage Hf model (T<sub>DM2</sub>) ages of 2650 to 2889 Ma by using the U-Pb age for each zircon. Six analytical spots, which define the Concordia upper intercept age of 2163 Ma, show  $^{176}$ Hf/ $^{177}$ Hf radios and  $\epsilon$ Hf(t) values of 0.281443 to 0.281496 and -0.7 to 1.5, respectively, with T<sub>DM2</sub> age of 2648 Ma to 2791 Ma by using the upper intercept age.

279 6. Discussion

#### 280 6.1 Significance of the two stages of zircon

Generally, zircon with high U content can easily break down into the metamict 281 state because of the radiation damage to the lattice caused by  $\alpha$ -particles originating 282 from the decay of uranium (Mezger and Krogstad, 1997). The physical and structural 283 changes often lead to the loss of Pb and addition of trace elements such as LREE. In 284 this study, the dark-CL rim spots have high U content, which is significantly higher than 285 the median value of zircon U content in granitic magma (350 ppm, Wang et al., 2011). 286 Hence, the metamictization degree of the zircons must be taken into consideration. Data 287 from dark-CL rim spots plot on the Concordia curve, indicating no obvious Pb loss. The 288 internal structure of dark-CL rim is relatively intact, with obvious oscillatory zoning, 289 and few cracks, implying that the physical and structural of the dark-CL rim remained 290 unchanged. Nasdala et al. (1998) suggested that the metamictization of zircon can be 291 well characterized by Raman spectroscopy. The half-width of the  $v_3(SiO_4)$  Raman band 292 (b) of  $10 \text{ cm}^{-1}$  and  $20 \text{ cm}^{-1}$  are proposed to approximately distinguish well-crystallized, 293 intermediate and metamict zircons (Nasdala et al., 1998). The dark-CL rim have b 294 values of 5.4-9.2, characterizing them as well-crystallized. Therefore, the above 295

features indicate that the dark-CL rim are not metamict. Consequently, it can be concluded that the U-Pb isotope and trace element systematics of the dark-CL rim have not been changed by metamictization.

Both the light-CL core and dark-CL rim have oscillatory zoning patterns, and their 299 chondrite-normalized REE patterns are characterized by steeply positive slopes from 300 the LREE to HREE with strong negative Eu anomalies and pronounced positive Ce 301 anomalies. The above characteristics are consistent with those of igneous zircon 302 (Hoskin and Schaltegger, 2003). Although hydrothermal zircon can also have 303 304 oscillatory zoning patterns similar to magmatic zircons, there are obvious differences in trace elements between the magmatic and hydrothermal zircon (Hoskin et al., 2005). 305 In the discrimination diagram (Fig. 9), both the spots of light-CL core and dark-CL rim 306 307 fall in or near the magmatic field, which is obviously different from hydrothermal zircon. Hence, the above characteristics indicate that both the light-CL core and dark-308 CL rim have a magmatic origin. 309

310 The light-CL core was overgrown continuously by the dark-CL rim. In addition, the contact between the light-CL core and dark-CL rim is euhedral. Such core-mantle 311 overgrowth relationships indicate that the light-CL core domains are not inherited 312 zircons. The similar Hf isotopic data of the light-CL core and dark-CL rim is also 313 consistent with this interpretation. For the age population, the samples of XY-001 and 314 XY-008 have MSWD of 1.3 and 1.2, respectively, which are both within the expected 315 range for 95 % confidence interval (Mahon, 1996). Although the <sup>206</sup>Pb/<sup>238</sup>U age of dark-316 CL rim is generally younger than that of light-CL core, the ages of these two distinct 317

domains have the characteristics of continuous variation, and do not show two or more 318 distinct age populations (Fig. 7b, d). These phenomena do not support the presence of 319 320 antecrystic zircons (Siégel et al., 2018). Hence, both the light-CL core and dark-CL rim are most likely autocrystic zircon formed in one distinct pulse of magma. The weighted 321 mean U-Pb ages of 160.7±1.1 Ma and 159.6±1.1 Ma can be interpreted as the 322 emplacement age of the Zhoujiapuzi granite. The obvious difference in internal 323 structure and trace element composition between the light-CL core and dark-CL rim 324 could be due to significant changes in their crystallization environments (Wang et al., 325 326 2007).

The Zr/Hf ratio in zircon has a negative correlation with the degree of fractionation 327 in the parent melt (Claiborne et al., 2006). In this study, the Zr/Hf ratios of the dark-CL 328 329 rim (21-40) are obviously lower than those of the light-CL core (39-56) (Fig. 10a). In addition, incompatible elements such as U and REE will become enriched in the highly 330 evolved magma (Zhao et al., 2014). In this study, the contents of U and REE of dark-331 CL rim are significantly higher than those of light-CL core (Fig. 10a). Overall, the 332 above features reflect that the dark-CL rim crystallized from a later and more evolved 333 magma. 334

Watson and Harrison (2005) found that the Ti content of zircon has a strong dependence on temperature (T), and obtained a Ti-in-zircon thermometer ( $T_{Zr-Ti}$ ). Since then, Ferry and Watson (2007) suggested that the solubility of Ti in zircon depends not only on T and activity of TiO<sub>2</sub> (aTiO<sub>2</sub>) but also on the activity of SiO<sub>2</sub> (aSiO<sub>2</sub>), and revised the  $T_{Zr-Ti}$ . We use the  $T_{Zr-Ti}$  from Ferry and Watson (2007) and the recommended

340	values ( $aSiO_2=1$ , $aTiO_2=0.5$ ) for the activity of $SiO_2$ and $TiO_2$ (Schiller and Finger,
341	2019), due to the presence of ilmenite and quartz in the Zhoujiapuzi granite. The $T_{Zr-Ti}$
342	from the light-CL core and dark-CL rim are 684-830 °C (average 761 °C) and 509-
343	712°C (average 635 °C), respectively, i.e. the light-CL core formed at higher
344	temperatures than the dark-CL rim. The $T_{\text{Zr-Ti}}$ value shows a significant positive
345	correlation with Zr/Hf (a tracer of fractional crystallisation), and shows continual
346	fractionation and cooling (Fig. 10b). As the light-CL core and dark-CL rim formed in
347	different magmatic evolution stages, it is problematic to use the same $aSiO_2$ and $aTiO_2$
348	values to calculate both $T_{Zr-Ti}$ values for both. For ilmenite bearing granites, Schiller
349	and Finger (2019) suggested that the variation of $aTiO_2$ values corresponding to
350	different zircon crystallization stages is small. In addition, Schiller and Finger (2019)
351	showed that the aSiO <sub>2</sub> value of the ilmenite-bearing granites at the onset of magmatic
352	zircon crystallization was more than 0.75. Even if the aSiO <sub>2</sub> value of the light-CL core
353	is changed from 1.0 to 0.75, the temperature will only drop by about ~27 $^\circ C$ , which is
354	significantly lower than the 126 °C difference between the average $T_{Zr-Ti}$ value of the
355	light-CL core and dark-CL rim. Therefore, it is certain that the light-CL core formed at
356	higher temperatures than the dark-CL rim, although we cannot calculate the specific
357	temperature difference.
358	Cerium exists in magmas as both $Ce^{3+}$ and $Ce^{4+}$ Because the 0.84-Å radius of the

Cerium exists in magmas as both  $Ce^{3+}$  and  $Ce^{4+}$ . Because the 0.84-Å radius of the Zr<sup>4+</sup> ion is more closely matched by the Ce<sup>4+</sup> (0.97-Å radius) than the Ce<sup>3+</sup> (1.143-Å radius) (all ionic radii are from Shannon, 1976), Ce<sup>4+</sup> is more compatible in the zircon structure than the Ce<sup>3+</sup>. Hence, the magnitude of Ce anomaly is a useful tool for

evaluating the oxygen fugacity condition of crystallization environment (e.g. Ballard et 362 al., 2002; Trail et al., 2012). Loader et al. (2017) suggested that the Ce/Ce\* ratio is 363 likely to be the most robust measure of magma redox conditions, although it is only a 364 semi-quantitative measure. In this study, the Ce/Ce<sup>\*</sup>ratio of the light-CL core and dark-365 CL rim are 6.30-153.36 (average 32.51) and 21.81-5773.06 (average 787.39), 366 respectively. This result suggests that the dark-CL rim formed in a higher oxygen 367 fugacity environment than the light-CL core. As shown in the Ce/Ce\*-Zr/Hf diagram 368 (Fig. 10c), Ce/Ce\* has a significant negative correlation with Zr/Hf, showing that the 369 370 oxygen fugacity condition is increasing with the evolution of magma.

The absence of enclaves and disequilibrium textures in the Zhoujiapuzi granite 371 and uniform EHf(t) values of the light-CL core and dark-CL rim do not support magma 372 373 mixing and wall-rock assimilation. Consequently, the abrupt change between the crystallization environment of the light-CL core and dark-CL rim is not due to the 374 magma mixing or contamination during magma evolution. Therefore, we propose that 375 376 the light-CL core was formed in a relatively deep magma chamber, which had low oxygen fugacity, low Zr saturation and higher temperature. The low Th, U and REE, 377 and widely-spaced oscillatory zoning patterns indicate a low growth rate of zircon 378 (Hoskin and Schaltegger, 2003; Wang et al., 2011). In contrast, the dark-CL rim was 379 formed during the ascent and/or at the emplacement location of the magma. At this 380 stage, the oxygen fugacity significantly increased, the temperature decreased, and Zr 381 saturation increased due to the crystallization differentiation. In this environment, the 382 crystallization rate of zircon significantly increased, forming the zircons with a higher 383

content of Th, U and REE elements, low CL emission and narrowly-spaced oscillatoryzoning patterns.

386 Zircon U-Pb dating is the most commonly used method in geochronology, especially dating the emplacement age of magmatic rocks. A weighted mean age or 387 upper intercept age is usually obtained to represent the emplacement time of a magmatic 388 rock. However, the autocrystic zircons in this study record two different magmatic 389 evolution stages. Previous studies, such as Wang et al. (2007), Zhao et al. (2014) and 390 Chen et al. (2020), also show that zircons can crystallize continually or intermittently 391 392 in a single phase of magmatism, showing several growth zones of clearly different internal structure and distinct time difference. Therefore, autocrystic zircon can be 393 formed in two or more evolution stages during one distinct pulse or increment of 394 395 magma. Some scholars even regard that the age difference of different stages can be more than dozens of Ma (Wang et al., 2007). Therefore, if the zircon ages in the same 396 magmatic rock have a large range of variation, this could be caused by the zircons 397 398 recording different stages in magmatic evolution, related to different levels of magma within the crust and/or different temperature regimes. In this paper, although the 399 apparent age of the dark-CL rim is generally younger than that of the light-CL core, the 400 age difference between the two is within the error range of the in-situ LA-ICP-MS 401 analyses (individual spot of  $\pm 3-5\%$  relative precision, Schmitz and Kuiper, 2013). 402 Therefore, further work is needed to verify the actual age difference between the two 403 magmatic evolution stages. Nevertheless, it is notable that the bulk petrology and 404 geochemistry of the host pluton does not record and reveal this two-stage magmatic 405

406 evolution, which can only be detected in the zircon analysis.

# **6.2 Genetic type: I-type affinity**

408	The Zhoujiapuzi granite has low Zr (113 - 242 ppm), Ce (26.5 - 121.5 ppm),
409	Zr+Nb+Ce+Y (152.0 - 382.6 ppm), (Na <sub>2</sub> O + K <sub>2</sub> O)/CaO (4.53 - 8.31) and FeO*/MgO
410	(5.09 - 10.56), distinct from the typical A-type granites (Fig. 11a-d). Furthermore, the
411	Zhoujiapuzi granite does not contain mafic alkaline minerals, such as arfvedsonite,
412	riebeckite, etc., which is also distinctly inconsistent with typical A-type granites (Wu et
413	al., 2003). Wu et al. (2017) suggested that a high formation temperature is one of the
414	most important characteristics of A-type granite. Zircon saturation thermometry $(T_{Zm})$
415	and Ti-in-zircon thermometer $(T_{Zr-Ti})$ are two methods for estimating magma
416	temperatures. As noted above, because the values of $aSiO_2$ and $aTiO_2$ during the early
417	zircon crystallization cannot be accurately obtained, the temperature of this period
418	cannot be accurately obtained through the Ti-in-zircon thermometer. Zircon saturation
419	thermometry was introduced by Watson and Harrison (1983) and is suitable for non-
420	peralkaline crustal source rocks. Since the zircon solubility is mainly affected by
421	temperature, major element compositions have a limited impact on calculated $T_{\rm Zrn}$
422	(Miller et al., 2003). In addition, the errors introduced by crystal-rich composition tend
423	to cancel as changes in Zr concentration and M value during crystallization have
424	opposite effects on the $T_{zrn}$ value (Miller et al., 2003). Therefore, the composition of
425	Zhoujiapuzi granite can be used to estimate the magma temperature. The calculated $Tz_{rn}$
426	values for the Zhoujiapuzi granite are in the range of 803-870 °C (mean=845 $\pm 20^{\circ}$ C).

It was proposed that the  $T_{Zrn}$  suggests an upper limit on the temperature of melt generation for inheritance-rich granitoid (Miller et al., 2003). Hence, the magma temperature of the Zhoujiapuzi granite should be lower than or equal to the  $T_{Zrn}$  value, which is significantly lower than that of typical A-type granite (>900 °C, Skjerlie and Johnston, 1992; Douce, 1997). Thus, the Zhoujiapuzi granite is not an A-type granite.

The samples of the Zhoujiapuzi granite have A/KNC < 1.1, relatively high Na<sub>2</sub>O 432 (3.96–4.65 wt.%) and lack peraluminous minerals (e.g. cordierite, andalusite, 433 muscovite and garnet), which are clearly different from S-type granites (Chappell and 434 435 White, 1992). With the rise of the degree of crystallization,  $P_2O_5$  contents wt.%) increase S-type granites, 436 (generally>0.1 in accompanied by an increase/immutability in SiO<sub>2</sub> (Wolf and London, 1994). However, the Zhoujiapuzi 437 438 granite samples have low P2O5 contents (0.02 - 0.08 wt.%), and decrease with increasing SiO<sub>2</sub> (Fig. 11e), which are features consistent with the I-type granite rather 439 than S-type granite (Chappell and White, 1992). Additionally, Rb has a positive 440 441 correlation with Y (Fig. 11f), which has been considered as an indicator of I-type granite (Jiang et al., 2018). Furthermore, the composition of the Zhoujiapuzi granite fall in the 442 I-type granite field in the discrimination diagrams of granites introduced by Collins et 443 al. (1982) (Fig. 11 c-d). Therefore, we conclude that the Zhoujiapuzi granite is a I-type 444 445 granite.

### 446 **6.3 Petrogenesis of the high Sr/Y granite**

The samples of the Zhoujiapuzi granite have high Sr/Y and (La/Yb)<sub>N</sub> ratios and
low Y and Yb contents (Fig. 12a) consistent with the geochemical signatures of modern

adakites (Defant and Drummond, 1990). However, other geochemical parameters of 449 the Zhoujiapuzi granite, such as the high K<sub>2</sub>O/Na<sub>2</sub>O ratio (0.93 -1.22), low Al<sub>2</sub>O<sub>3</sub> 450 content (14.49–15.02%, except one) and Sr content (in half of the samples lower than 451 400 ppm), are obviously different from typical adakites (K<sub>2</sub>O/Na<sub>2</sub>O $\leq$ 0.42, Al<sub>2</sub>O<sub>3</sub> $\geq$ 452 15 %, Sr>400 ppm, Defant and Drummond., 1990; Drummond et al., 1996, Martin et 453 al., 2005). A variety of petrogenetic models have been proposed for the origin of high 454 Sr/Y magmatic rocks, such as partial melting of subducting oceanic crust (Model A, 455 Defant and Drummond, 1990), delaminated lower continental crust (LCC) (Model B, 456 457 Kay and Kay, 1993; Xu et al., 2002), differentiation of basaltic arc magma (Model C, Castillo et al., 1999), magma mixing between mantle-derived mafic and crust-derived 458 silicic magmas (Model D, Ma et al., 2013a), partial melting of thickened basaltic LCC 459 (Model E, Gao et al., 2004; Ou et al., 2017), or melting of a high Sr/Y (and La/Yb) 460 source (Model F, Kamei et al., 2009; Ma et al., 2015). 461

## 462 6.3.1 Model A: Partial melting of subducting oceanic crust

463 The partial melting of the young, hot and hydrated subducted oceanic slab in the garnet stability field is the classical formation model of adakite (high Sr/Y rock) (Defant 464 and Drummond, 1990). Studies have shown that the rock with this genetic model 465 generally has the characteristics of high mantle components (such as MgO, CaO and 466 Cr) because of the involvement of mantle magma (Wang et al., 2018). However, this 467 phenomenon was not seen in the Zhoujiapuzi granite. In addition, the Zhoujiapuzi 468 granite has high K<sub>2</sub>O/Na<sub>2</sub>O ratios (0.92–1.22, average 1.13), which is inconsistent with 469 the slab-derived adakites (K<sub>2</sub>O/Na<sub>2</sub>O=  $\sim$ 0.4, Martin et al., 2005). Moreover, the low 470 EHf(t) values (-26.6 to -22.8) of the Zhoujiapuzi granite are also inconsistent with the 471 magmas derived from the partial melting of oceanic crust, which generally have 472

depleted isotopic character (Zhan et al., 2020). Furthermore, the Zhoujiapuzi granite
has low Ti/Eu and high Nd/Sm radios (Fig. 13a), and markedly negative Nb-Ta
anomalies (Fig. 4b), which are distinct from those of oceanic basalts (Yu et al., 2012).
In summary, the Zhoujiapuzi granite is difficult to explain by Model A.

#### 477 6.3.2 Model B: Delaminated lower continental crust (LCC)

High-density, garnet-bearing mafic lower crust delaminating or foundering into 478 479 the asthenosphere mantle and subsequent interaction with mantle peridotite could produce high Sr/Y magmas (Kay and Kay 1993). Because the melt formed by partial 480 melting of the delaminated lower crust would interact with mantle peridotite during 481 magma ascent, the high Sr/Y magmas related to this petrogenetic model generally have 482 high MgO, Mg# and TiO<sub>2</sub> (Gao et al., 2004; Ou et al., 2017; He et al., 2021). The MgO 483 (0.10-0.44 wt.%), Mg# (15-26) and TiO<sub>2</sub> (0.09-0.34 wt.%) values of Zhoujiapuzi 484 granite are significantly lower than the above values (Fig. 13b- d). In addition, 485 delamination of the lower crust generally occurs in within-plate extensional settings 486 487 (Gao et al., 2004), and will form a large number of Mg-rich (Mg#>50) rocks due to the partial melting of lithospheric mantle and/or upwelling of asthenosphere (Ou et al., 488 2017). However, these Jurassic magmatic rocks in the Liaodong Peninsula are generally 489 490 considered to be formed in a compressional environment related to the subduction of the Paleo-Pacific slab (Li et al., 2004; Yang et al., 2015a; Zhu and Xu, 2019; Zheng et 491 al., 2018). Furthermore, the middle-late Jurassic granites are generally Mg-poor (Fig. 492 13c). Due to the high temperature of the asthenosphere (1200 °C, Parsons and 493 McKenzie, 1978; King et al., 2015), rocks formed by partial melting of the delaminated 494 lower crust should possess a high-temperature fingerprint. T<sub>Zrn</sub> has been used as a 495 geothermometer to estimate partial melting temperatures (e.g., Miller et al., 2003; 496

497 Collins et al., 2016). As mentioned before, the  $T_{Zrn}$  of the Zhoujiapuzi granite is below 498 900 °C, which is markedly lower than the temperature of the asthenosphere. Therefore, 499 the petrogenetic model of delaminated lower continental crust (Model B) is also 500 inconsistent with the Zhoujiapuzi granite.

#### 501 6.3.3 Model C: Differentiation of basaltic arc magma

Low-pressure fractional crystallization (involving olivine + clinopyroxene + plagioclase + amphibole+ titanomagnetite) or high-pressure fractional crystallization (involving garnet) from basaltic magmas have been proposed as two ways to generate adakitic characteristics (Castillo et al., 1999; Macpherson et al., 2006).

However, the composition of the Zhoujiapuzi granite is relatively uniform, 506 including SiO<sub>2</sub>, MgO and Na<sub>2</sub>O, which does not support major fractional crystallization 507 (Xue et a., 2017). Furthermore, the Zhoujiapuzi granite has abundant inherited zircons 508 and no obvious depletion of Sr, Eu and Ba, showing that this granite has not experienced 509 510 extensive fractionation (Miller et al., 2003). The samples form clear partial melting 511 trends on the La/Yb versus La diagram (Fig. 13e), which also suggests that partial melting was more important than fractional crystallization (Gao et al., 2007; Shahbazi 512 et al., 2021). In addition, crystal fractionation of basaltic melts can only form minor 513 volumes of granitic melts, the ratio of the two is about 9:1 (Zeng et al., 2016). However, 514 for the same age interval, no coexisting mafic-intermediate rocks have been found in 515 the research area. In the wider region of the Liaodong Peninsula, Middle-Late Jurassic 516 magmatism is dominated by felsic compositions; mafic- intermediate rocks are only 517 reported in the Huaziyu area (lamprophyre dikes, Jiang et al., 2005). Therefore, it is 518 unlikely that there are large-scale mafic- intermediate rocks contemporaneous with the 519 Zhoujiapuzi granite at depth according to the rock assemblage of Liaodong Peninsula 520

in this period. Moreover, the zircon Hf isotopic compositions of the Zhoujiapuzi granite are quite different from those of the depleted mantle, but are similar to those of the basement (Liaohe Group and Liaoji granite) in the study area (Fig. 8). The ancient inherited zircons (2500 to 2173) with low  $\epsilon$ Hf(t) values also indicate older crustal material in the Zhoujiapuzi granite. For these reasons, it is highly improbable that Zhoujiapuzi granite was derived by differentiation of basaltic magma (Model C).

# 527 6.3.4 Model D: Magma mixing between mantle-derived mafic and crust-derived 528 silicic magmas

The Zhoujiapuzi granite has high  $K_2O/Na_2O$  ratio (>1) and A/CNK value (>1), 529 together with the absence of mingling textures, mafic microgranular enclaves (MMEs), 530 felsic xenocrysts and melting texture of plagioclase, implying that the mantle-derived 531 magma is unlikely to have played an important role in the genesis of the Zhoujiapuzi 532 granite (Castro et al., 1991). In addition, the Zhoujiapuzi granite is characterized by the 533 development of biotite, but lacks amphibole and pyroxene. These features, coupled with 534 the high A/CNK value, are consistent with an origin as a crust-derived granitoid, but 535 obviously different from the granitoids formed by crust-mantle-derived magma mixing 536 (Barbarin, 1990). Moreover, granites formed by magma mixing generally have high 537 538 MgO, TFe<sub>2</sub>O<sub>3</sub>, CaO and Cr contents and low SiO<sub>2</sub> content (Ma et al., 2013a; Wang et al., 2018). These features are obviously inconsistent with the Zhoujiapuzi granite in this 539 study. Additionally, the  $\varepsilon$ Hf(t) values and trace element composition of the two stages 540 of zircon also do not support magma mixing. Hence, magma mixing of mantle-derived 541 and crust-derived magmas (Model D) is also unlikely to have produced the Zhoujiapuzi 542 granite. 543

#### 544 6.3.5 Model E: Partial melting of thickened basaltic LCC

Experimental studies have shown that the partial melt of basaltic LCC in the garnet stabilization zone (> 40 km, i.e. ~1.2 GPa) can produce magma with a high Sr/Y ratio (Rapp et al., 2003 and references therein). In these scenarios, high Sr/Y and overall adakitic affinity are caused by leaving garnet as residual phases (e.g. Gao et al., 2004). Based on geochemical data for the Zhoujiapuzi granites, partial melting of thickened basaltic LCC is also unlikely to account for the high Sr/Y Zhoujiapuzi granite (Model E). This conclusion is based on the following observations:

(1) This ratio of  $(Gd/Yb)_N$  is the most important feature to judge whether garnet is 552 involvement in magma genesis (Ma et al., 2012). If the  $(Gd/Yb)_N$  ratio of the source is 553 554 similar to the average value of the LCC (1.71, Rudnick and Gao, 2003), partial melting of these crustal materials controlled by garnet at high pressure can produce melt with 555  $(Gd/Yb)_N$  of 5.8 (Huang and He, 2010). In contrast, the  $(Gd/Yb)_N$  values (1.22–5.06, 556 average 2.69) of the Zhoujiapuzi granite are relatively low. (2) Studies of lower-crustal 557 xenoliths show that garnet may not be a common mineral in the lower crust of the NCC 558 559 (Ma et al., 2012). (3) As shown in the discrimination diagrams of granite sources (Fig. 13f, g), all samples fall in the range of metagreywacke-derived melts. Therefore, the 560 Zhoujiapuzi granite was considered to have been derived from crustal anatexis of 561 562 metagraywacke (or intermediate-acid igneous rock with similar mineral composition), rather than basaltic lower crust. 563

# 564 6.3.6 Model F: Melting of a high Sr/Y (and La/Yb) source

565 Studies have shown that when a source rock has a high Sr/Y ratio, the high Sr/Y 566 signature of the derived magma can inherit from their source, regardless of pressure 567 (Kamei et al., 2009; Moyen, 2009; Ma et al., 2015). We suggest that partial melting of high Sr/Y Liaoji granite was most probably the origin of the high Sr/Y Zhoujiapuzigranite, as discussed below (Model F).

570 The Zhoujiapuzi granite has similar mineral assemblages (contains abundant Kfeldspar and lacks hornblende) and geochemical composition (Fig. 13h) to the 571 Tsutsugatake intrusion, which is explained by partial melting of arc-type tonalite or 572 adakitic granodiorite (Kamei et al., 2009). Among the inherited zircons from 573 Zhoujiapuzi granite, the <sup>207</sup>Pb / <sup>206</sup>Pb ages of all the spots are between 2132 and 2200 574 Ma, except one, and yield a Concordia upper intercept age of 2163 Ma. Both 575 576 assimilation of country-rocks and incomplete melting of source rocks can explain the genesis of inherited zircon in granite. Due to the similar T<sub>DM2</sub> of autocrystic zircons 577 (light-CL core and dark-CL rim) and inherited zircons, these inherited zircons most 578 likely come from the source of the Zhoujiapuzi granite. In the study area, meta-579 sedimentary rocks and meta-volcanic rocks of the South Liaohe Group, 580 Paleoproterozoic mafic rocks, as well as the Liaoji granites, have ~2.16 Ga zircon. In 581 spite of an age peak of 2.17–2.16 Ga in detrital zircon age spectra of the metasediments 582 from the South Liaohe Group, melting of a sediment-dominated source is unlikely to 583 have occurred, as it would have also introduced other age peaks such as ~2.03 Ga and 584 ~2.50 Ga (Li et al., 2015; Wang et al., 2020b). In addition, given the I-type 585 characteristics of the Zhoujipuzi granite, derivation from an igneous precursor is more 586 plausible rather than a metasedimentary origin (Chappell and White, 1992). Therefore, 587 these ~2.16 Ga zircons from the Zhoujiapuzi granite are unlikely to come from the 588 South Liaohe Group. As shown in the host rock discrimination diagrams (Fig. 14, 589 introduced by Belousova et al., 2002), all the ~2.16 Ga inherited zircons from 590 Zhoujiapuzi granite fall into the granitoid area (Fig. 14), precluding that these ~2.16 Ga 591 zircon come from the Paleoproterozoic mafic rocks. In addition, the ~2.16 Ga inherited 592

zircons from Zhoujiapuzi granite and the zircons from the Liaoji granites lie in a similar
area in the ɛHf(t)-age (Ma) diagram (Fig. 8). Hence, the ~2.16 Ga inherited zircon most
likely come from the Liaoji granites.

Some of the Liaoji granites, such as the Muniuhe granite (comprising granodiorite and syenogranite with no distinct boundary between the two), have adakitic signatures, and similar REE and trace element patterns as the Zhoujiapuzi granite (Fig. 4). Based on a model of batch melting (Shaw, 1970) using the experiments of Conrad et al. (1988), the high Sr/Y characteristic of the Zhoujiapuzi granite can be explained by partial melting of Muniuhe granitic pluton leaving amphibole as the main residue (Fig. 12b).

In our modelling, we choose the XY-005 sample to approximately represent the 602 primitive melt composition. The reasons are as below: as mentioned above, the high 603 Sr/Y characteristics of the Zhoujiapuzi granite are not caused by the fractional 604 crystallization of amphibole. Furthermore, the lack of positive correlation between 605 Dy<sub>N</sub>/Yb<sub>N</sub> ratios and La<sub>N</sub>/Yb<sub>N</sub> ratios (Fig. 13i) also suggests that fractional 606 crystallization of amphibole was not a significant process for the Zhoujiapuzi granite. 607 On the other hand, the samples of Zhoujiapuzi granite display variable Eu and Sr 608 contents, implying that plagioclase is likely a fractional phase. The separation of 609 titanomagnetite could explain the positive trend in TFe<sub>2</sub>O<sub>3</sub> with increasing TiO<sub>2</sub> content, 610 consistent with the occurrences of magnetite in some studied rocks. This possible 611 mineral assemblage of fractional crystallization is also reflected by the chemical 612 variations in the Sr/Y-Y diagram (Fig. 12b). Hence, the sample XY-005, which has 613 highest Sr/Y, was chosen to represent a primitive melt composition. To find the best 614 matching experimental melts, we have compared the major elements of the XY-005 615 sample with that of experimental melts and the characteristics of no garnet residue 616 discussed above. Results are shown in Fig. 12b. The Sr and Y compositions of the 617

starting material used in these experiments resemble those of the average composition of the Muniuhe granitic pluton (Sr=475 ppm, Y=9.77 ppm), if the residue contains a large volume of amphibole (>90 %). However, if more plagioclase is retained in the residue (e.g. 18.3 %), a source region with a higher Sr content is required. Therefore, a similar high Sr/Y Liaoji granite to the Muniuhe granitic pluton can produce the high Sr/Y signatures of the Zhoujiapuzi granite.

624 A large number of Yanshanian adakites (or high Sr/Y rocks) are developed in the NCC, which are generally considered to be derived from the thickened basaltic LCC 625 626 (e.g. Gao et al., 2004; Wu et al., 2005a; Ma et al., 2013b). Zhang et al. (2001, 2003) suggested that these so-called "C-type adakites" indicated a large-scale crustal 627 thickening event. However, according to the studies on the Triassic and Jurassic adakitic 628 rocks near the Pingquan area, the northern part of the NCC, Ma et al. (2012, 2015) 629 suggested that the adakitic signatures of these rocks are inherited from their source 630 rocks. The research of the Zhoujiapuzi granite in this study also shows that among the 631 widely distributed Jurassic high Sr/Y granites in the Liaodong Peninsula, there is at 632 least one pluton with a high Sr/Y signature inherited from the source. Therefore, we 633 suggest that melting of a high Sr/Y (and La/Yb) source is one of the important processes 634 for the generation of Yanshanian high Sr/Y rocks in the NCC. This kind of high Sr/Y 635 granite does not need to be formed in the garnet stability field. 636

#### 637 6.4 Tectonic implications

A large number of Early Jurassic arc-like igneous rocks occur in the northeast part
of NCC- Korean Peninsula-Hida belt, which belong to the middle-high K calc-alkaline
series and are characterized by enrichment in LILE and depletions in HFSE (Wu et al.,
2007; Tang et al., 2018 and references therein). In addition, the Early Jurassic

accretionary complexes in the eastern margin of the Eurasian continent and the Japan
islands, such as the Heilongjiang complex, the Khabarovsk complex and the MinoTamba complex, are considered to be related to subduction (Wu et al., 2007; Tang et al.,
2018 and references therein). It is generally accepted that the Paleo-Pacific slab
subducted westwards in the Early Jurassic (Tang et al., 2018; Zhu and Xu, 2018).

In the middle-late Jurassic, I-type granites are dominant in the Liaodong Peninsula, 647 such as the Zhoujiapuzi granite (this study), Heigou pluton, Gaoliduntai pluton (Wu et 648 al., 2005a), Waling granite (Yang et al., 2015a) and Wulong granite (Yang et al., 2018). 649 650 There are not A-type granites, and mantle derived magmatism is extremely rare. These granites were formed by partial melting of crustal materials without obvious 651 contribution of mantle derived magma (Wu et al., 2005a; Yang et al., 2015b, 2018; Xue 652 653 et al., 2020). In addition, WNW-ESE compression during 157-143 Ma was widespread in the Liaodong Peninsula (Yang et al., 2004; Zhang et al., 2020). It not only 654 mylonitized the granite plutons in middle-lower crust levels, but also intensely 655 656 deformed the thick sedimentary cover in the upper crust (Qiu et al., 2018; Ren et al., 2020). Hence, Late Jurassic magmatism in the Liaodong peninsula is most likely to be 657 related to subduction of the Paleo-Pacific plate in a mature continental arc, with crust 658 previously thickened by compressional tectonics, related to both the oceanic subduction 659 and the earlier Mesozoic collisions at the north and south margins of the NCC. This 660 setting would produce the conditions required for extensive crustal melting of pre-661 existing basement. There is a potential resemblance to the modern arc of the Central 662 Andes (Allmendinger et al., 1997), where crustal thickening and plateau growth has 663

developed over the Cenozoic (Scott et al., 2018), and melting of older basement has
taken place during subduction of the Nazca plate (Miller and Harris, 1989). This model
is also consistent with the idea that much of eastern China was a high orogenic plateau
during the Mesozoic, before widespread Early Cretaceous extension and core complex
development (Meng, 2003; Chu et al., 2020).

#### 669 7. Conclusions

670 (1) LA-ICP-MS zircon U-Pb dating indicates that the Zhoujiapuzi granite in the
671 Liaodong Peninsula formed at ~160 Ma.

(2) Zircon growth in Zhoujiapuzi granite can be divided into two distinct stages.
The light-CL core was formed in a deeper, hotter, magma chamber, which had low
oxygen fugacity and high temperature. The dark-CL rim formed from later, more
evolved, magma. Oxygen fugacity significantly increased and the temperature
decreased at this stage. The Zhoujiapuzi granite is a case study of multistage generation
and emplacement of magma, revealed by zircons, where no signals are discernible in
the bulk petrology or geochemistry.

(3) The I-type Zhoujiapuzi granite originated from partial melting of the
Paleoproterozoic Liaoji granites. The high Sr/Y compositions are inherited from their
source rocks, rather than being a direct indication of deep crustal melting, or any other
common mechanism for generating adakitic signatures.

(4) The Late Jurassic tectonic setting of the Liaodong Peninsula and the easternNCC resembled the modern orogenic plateau of the Central Andes, where silicic

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magmatism may occur by partial melting of older continental crust in a compressional
environment, related to the subduction of the Paleo-Pacific plate.

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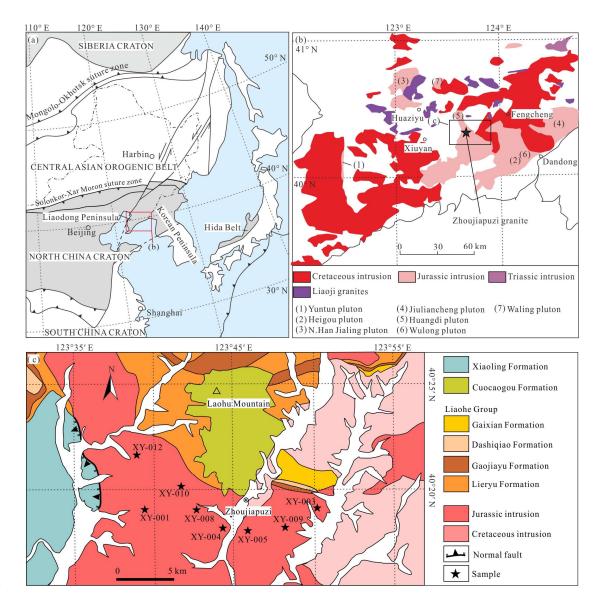
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- **1163 Table captions**
- Table S1. Major element (wt. %) and trace element (ppm) compositions of theZhoujiapuzi granite
- 1166 Table S2. Raman microprobe data
- 1167 Table S3. The zircon major element (wt. %) and trace element (ppm) from the

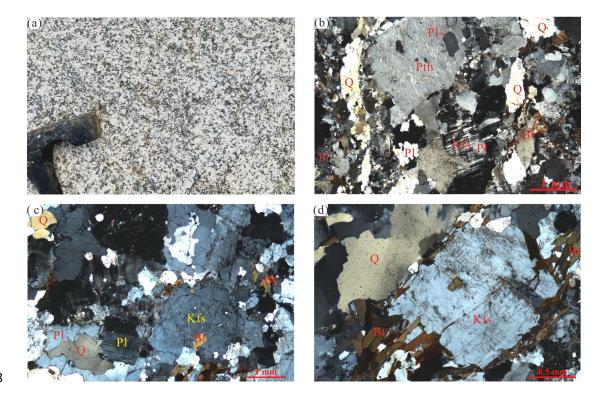
- 1168 Zhoujiapuzi granite
- 1169 Table S4. Zircon La-ICP-MS U-Pb isotopic data and ages of the Zhoujiapuzi granite
- 1170 Table S5. Zircon Hf isotopic data of the Zhoujiapuzi granite
- 1171
- 1172 Figure captions



1174 Figure 1. (a) Simplified geological map of Northeast China (Modified from Li et al.,

1175 2016); (b) distribution of Mesozoic intrusions in the Liaodong Peninsula (Modified

1176 from Wu et al., 2005a); (c) geological map of the Zhoujiapuzi granite.

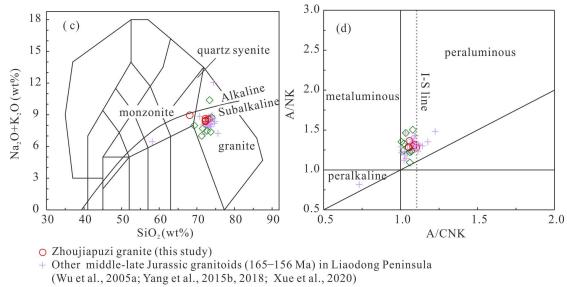




1179 Figure 2. Outcrop photograph (a) and corresponding micrographs (b, c, d-

- 1180 perpendicular polarized light). Q quartz; Kfs feldspar; Pl plagioclase; Pth perthite; Bt
- 1181 biotite

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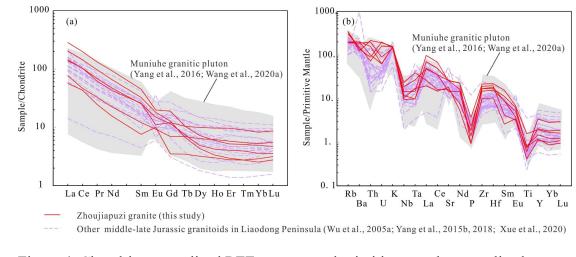
♦ Muniuhe granitic pluton (Yang et al., 2016; Wang et al., 2020a)

1184 Figure 3. Geochemical classification diagrams for the Zhoujiapuzi granite. (a) TAS

1185 diagram (after Frost et al., 2001); (b)A/CNK-A/NK diagram (after Maniar and

## 1186 Piccoli, 1989)

## 1187



1189 Figure 4. Chondrite-normalized REE patterns and primitive mantle-normalized trace

element patterns of the Zhoujiapuzi granite (chondrite and primitive mantle values are

1191 from Sun and McDonough, 1989).

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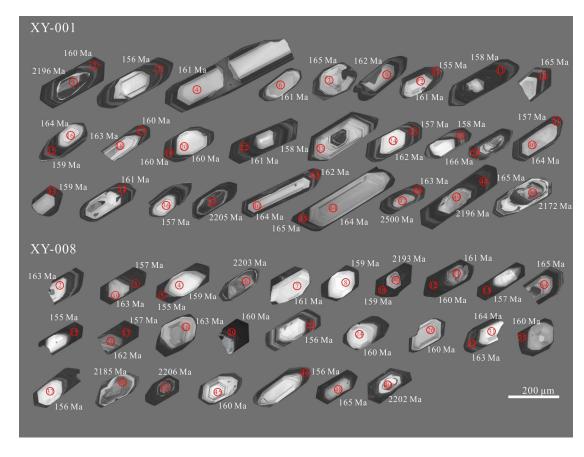
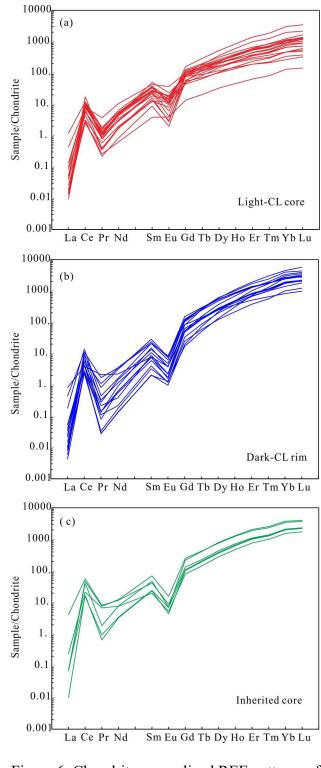


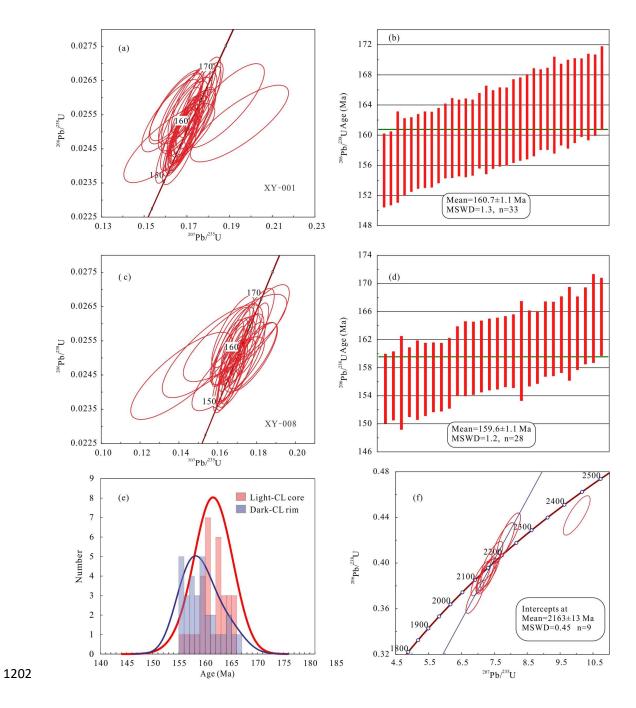
Figure 5. CL images of zircons. Circles denote U-Pb analysis spot. Numbers in the circles are the spot numbers. Numbers near the analytical spots are the U–Pb ages (Ma).



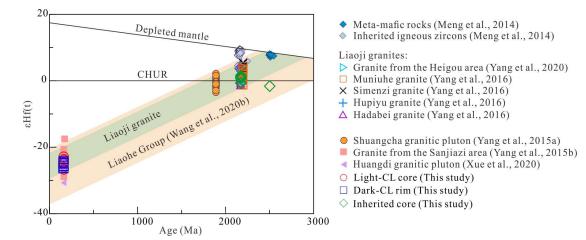
1199 Figure 6. Chondrite-normalized REE patterns of zircon (chondrite values are from

1200 Sun and McDonough, 1989).

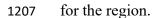
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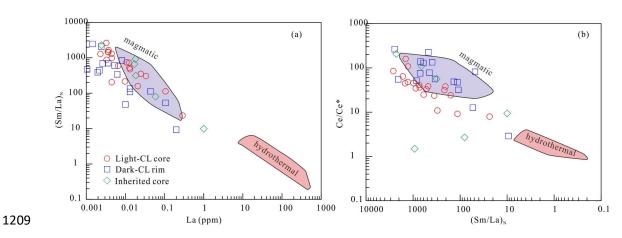


1203 Figure 7. Concordia diagrams for zircon LA-ICP-MS U-Pb analyses.

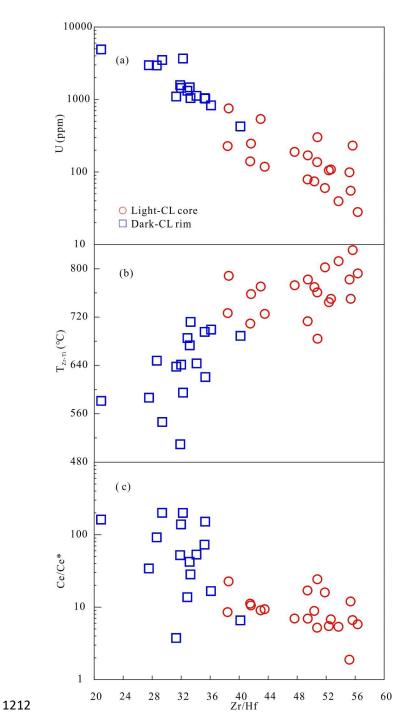


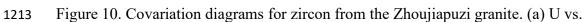
1206 Figure 8. Zircon ɛHf(t)-age (Ma) diagram for samples in this study and published data



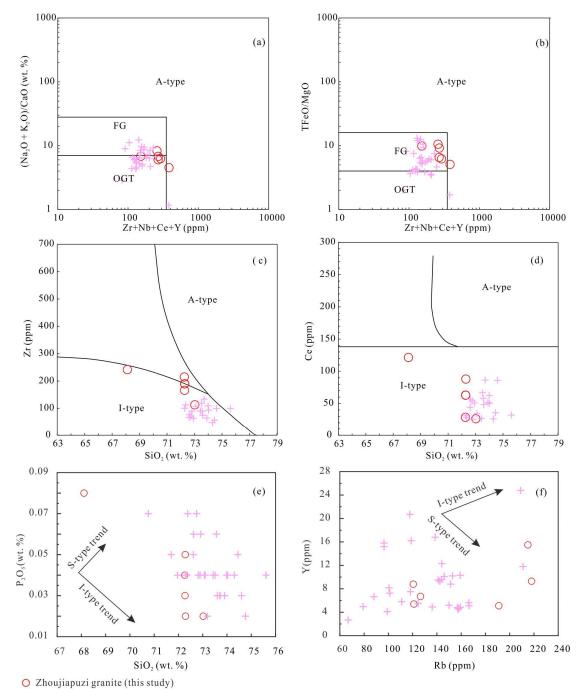


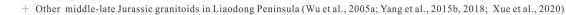
1210 Fig. 9. Discrimination plots for magmatic and hydrothermal zircon (Hoskin, 2005).





1214 Zr/Hf; (b)  $T_{Zr-Ti}$  vs. Zr/Hf; (c) Ce/Ce\* vs. Zr/Hf.



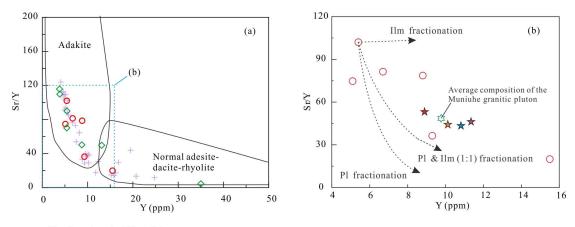


1217 Figure 11. Chemical variation diagrams for the Zhoujiapuzi granite. (a and b)

1218 Zr+Nb+Ce+Y vs. (Na<sub>2</sub>O + K<sub>2</sub>O)/CaO and TFeO/MgO (after Whalen et al., 1987); (c

1219 and d) SiO<sub>2</sub> vs. Zr and Ce (after Collins et al., 1982); (e) SiO<sub>2</sub> vs. P<sub>2</sub>O<sub>5</sub> diagram; (f)

1220 Rb vs. Y diagram



O Zhoujiapuzi granite (this study)

O Enorgiapari giante (init study)
 O there indide-late jurassic granitoids in Liaodong Peninsula (Wu et al., 2005a; Yang et al., 2015b, 2018; Xue et al., 2020)
 Muniuhe granitic pluton (Yang et al., 2016; Wang et al., 2020a)
 Average composition of the Muniuhe granitic pluton

★ Simulated source (Sr=473 ppm; Y=8.9 ppm) based on the residual mineralogy of Amp (F=85.5 %)

Simulated source (Sr=447 ppm; Y=10.1 ppm) based on the residual mineralogy of Amp : Ilm= 98.9: 1.2 (F=80.8 %)

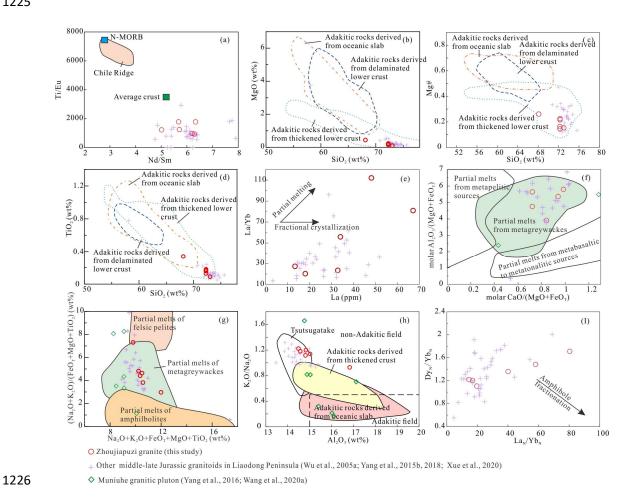
\* Simulated source (Sr=469 ppm; Y=10.8 ppm) based on the residual mineralogy of Amp: P1 : Ilm= 92.7: 6.9: 0.5 (F=78.1 %)

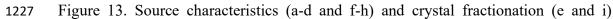
\* Simulated source (Sr=524 ppm; Y=11.3 ppm) based on the residual mineralogy of Amp: Pl : Ilm= 82.4: 18.3 (F=72.2 %)

## 1223 Figure 12. Adakite discrimination diagrams for the Zhoujiapuzi granite (after Defant

and Drummond, 1990). 1224

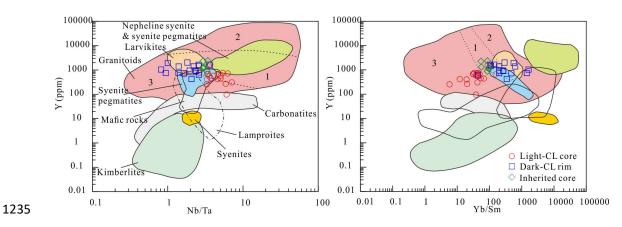
1225





discrimination diagrams for the Zhoujiapuzi granite. Plots of (a) Nd/Sm vs. Ti/Eu (Yu 1228 et al., 2012); (b-d) SiO<sub>2</sub> vs. MgO, Mg# and TiO<sub>2</sub> (after Wang et al., 2006); (e) La vs. 1229 La/Yb (Gao et al., 2007); (f) molar Al<sub>2</sub>O<sub>3</sub>/(MgO+FeO<sub>T</sub>) vs. molar CaO/(MgO+FeO<sub>T</sub>) 1230 (after Altherr al., 2000); (g) (Na<sub>2</sub>O+K<sub>2</sub>O)/(FeO<sub>T</sub>+MgO+TiO<sub>2</sub>) 1231 et VS. Na<sub>2</sub>O+K<sub>2</sub>O+FeO<sub>T</sub>+MgO+TiO<sub>2</sub> (after Patiño Douce, 1999); (h) K<sub>2</sub>O/Na<sub>2</sub>O vs. Al<sub>2</sub>O<sub>3</sub> 1232 diagrams (after Kamei et al., 2009); (i) La<sub>N</sub>/Yb<sub>N</sub> vs. Dy<sub>N</sub>/Yb<sub>N</sub>. 1233





1236 Figure 14. The fields of zircon compositions used as discriminants for different rock

1237 types (after Belousova et al., 2002). 'Granitoids' include: 1 aplites and leucogranites;

1238 2 granites; 3 granodiorites and tonalities

1239