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# Revealing the link between A-type granites and hottest melts 19 from residual metasedimentary crust 20 21 22 B.B. Carvalho<sup>1,\*</sup>, O. Bartoli<sup>1</sup>, B. Cesare<sup>1</sup>, M. Satish-Kumar<sup>2</sup>, M. Petrelli<sup>3</sup>, T. Kawakami<sup>4</sup>, T. 23 Hokada<sup>5</sup>, M. Gilio<sup>6</sup> 24 25 <sup>1</sup> Dipartimento di Geoscienze, Università di Padova, Via G. Gradenigo, 6. 35129, Padua, Italy. 26 <sup>2</sup> Faculty of Science, Niigata University, 8050 Ikarashi 2-no-chi, Nishi-ku, Niigata-shi 950-2181, Japan. 27 <sup>3</sup> Dipartimento di Fisica e Geologia, Università di Perugia, Piazza dell'Università 1, 06123, Perugia, Italy. 28 <sup>4</sup> Department of Geology and Mineralogy, Kyoto University, Kitashirakawa-Oiwake-cho, Sakyo-ku, Kyoto 606-29 8502, Japan. 30 <sup>5</sup> Geoscience Group, National Institute of Polar Research, 10-3 Midori-cho, Tachikawa, Tokyo, 190-8518, Japan. 31 <sup>6</sup> Dipartimento di Geologia, Università di Pavia, Via Adolfo Ferrata 1, 27100, Pavia, Italy. 32 \*corresponding author: bruna.borgescarvalho@unidp.it 33 34 ABSTRACT 35 Among S-, I-, and A-type granites, the latter are the most debated in terms of origin, and 36 metasedimentary crust is usually discarded as potential source. Here we tackle this issue by

37 adopting an in-source perspective, rather than focusing on the final product (granite), documenting 38 the occurrence of pristine melt inclusions (MI) in garnet from residual metapelitic ultrahigh 39 temperature (UHT) granulite from East Antarctica. Coexistence of sapphirine + quartz, phase 40 equilibria calculations and Zr-in-rutile thermometry indicate that MI trapped UHT melts formed 41 at peak conditions (930–1000 °C) from a residual metapelitic source. MI are granitic with weakly 42 peraluminous to weakly peralkaline affinity, ferroan character, high alkali contents, high K/Na and 43 Ga/Al, and low Ca, Ba, Sr, and H<sub>2</sub>O concentrations. These features and geochemical modelling 44 indicate that MI represent primary melts for high-SiO<sub>2</sub> A-type granites. Therefore, MI reveal the 45 missing link between A-type granites and the hottest metasedimentary crust. Voluminous amounts 46 of slightly peraluminous, high-SiO<sub>2</sub> A-type granites can be produced in large, residual UHT 47 terranes such as those of eastern Gondwana. Our results provide a wider view of processes 48 responsible for granite formation and show that a larger variety of granites must be considered in 49 models on the effects of UHT anatexis on crustal differentiation.

50

## 51 INTRODUCTION

52 A-type granites form at high temperature and show high FeOt/(FeOt+MgO), high Ga/Al, high 53 abundances of alkali, and high field strength elements, low CaO, H<sub>2</sub>O, Ba, and Sr, ranging from 54 metaluminous to slightly peraluminous, sometimes peralkaline (Collins et al., 1982; Whalen et al., 55 1987; King et al., 1997; Frost and Frost, 2011). Although less abundant than the classic S- and I-, 56 the A-type granites occur in a variety of geodynamic contexts, including extraterrestrial materials 57 (Bonin, 2007). Their origin is controversial and subject of debate (Collins et al., 2021; Eby, 1990; 58 Creaser et al., 1991; Eby, 1992; King et al., 1997; Patiño-Douce, 1997; Huang et al., 2011; Frost 59 and Frost, 2011). Petrogenetic models proposed for their formation are i) anatexis of 60 (meta)tonalitic to granodioritic crust, ii) differentiation of basaltic melts, and iii) differentiation 61 coupled with assimilation of crustal material (Collins et al., 1982; Eby, 1990; Frost and Frost, 62 2011). When felsic igneous protoliths are invoked as a source, the debate focuses on its possible 63 residual (Collins et al., 1982; 2021) or fertile nature (Creaser et al., 1991; Patiño-Douce, 1997; 64 Frost and Frost, 2011). Metasedimentary sources are not considered an option, despite some 65 occurrences may indicate the contrary (Huang et al., 2011; Zhang et al., 2022).

Since the formation of A-type granites may require T >900 °C (Clemens et al., 1986; Collins et al., 2021), debating their crustal origin means entering the domain of ultrahigh temperature (UHT) metamorphism, where longer duration might be instrumental in generating larger volume of melts (Harley, 2016). Most studies on UHT granulites have focused on their strongly residual assemblages (Kelsey and Hand, 2015), whereas characterization of melts produced, and their impact on crustal differentiation remain poorly known (Clark et al., 2011).

Here we present new data on exceptionally preserved melt inclusions (MI) in garnet of granulites from Antarctica. We demonstrate that the MI formed at UHT conditions from a residual metapelitic source and that their melts share key geochemical features with many A-type granites. Our study provides the first in-source evidence linking high-SiO<sub>2</sub> A-type granites to the anatexis 76 of residual metasedimentary crust at UHT conditions.

77

# 78 GEOLOGICAL BACKGROUND AND RESIDUAL SOURCE ROCK

79 Metapelitic Mg-Al granulites occur as lenses within garnet-sillimanite and garnet-biotite gneisses 80 at Rundvågshetta, Lützow-Holm Complex (East Antarctica) where a clockwise *P*–*T* path with peak 81 T of 920–1040°C is well constrained (Durgalakshmi et al., 2021 and references therein). This UHT 82 event, also recorded in Madagascar, Sri-Lanka and southern India, is related to continental 83 collision and amalgamation of the Gondwana supercontinent at 550-520 Ma (Durgalakshmi et al., 84 2021; see Data Repository). As older (>600 Ma) high-grade metamorphic events are also recorded, 85 the UHT metamorphism at Rundvågshetta has been interpreted to result from crustal pre-86 conditioning before Gondwana amalgamation (Durgalakshmi et al., 2021). The studied Mg-Al 87 granulite is coarse-grained and layered, with predominant mafic domains and less abundant felsic 88 ones (Fig. 1A, see Data Repository).

89

### 90 UHT CRUSTAL MELTS PRESERVED IN GRANULITIC GARNET

91 In the felsic domain (Fig. 1B) garnets contain clusters of inclusions with zonal arrangement, 92 indicating entrapment during garnet growth (Cesare et al., 2015). Inclusions consist of quartz and 93 accessory zircon, rutile and apatite, and both glassy, and crystallized MI (i.e., *nanogranitoid*; 94 Cesare et al., 2015). The exceptionally preserved glassy inclusions (GI) are very small (5 to 25 95 µm) and contain isotropic glass (Fig. 1 C), whereas the coexisting nanogranitoids (NI) are larger 96  $(5-150 \,\mu\text{m})$  and contain feldspars, quartz and biotite (Fig. 1D-F). Accidentally trapped phases in 97 MI are apatite, rutile and occasionally touching sapphirine + quartz (hereafter Sa + Qz) (Fig. 1D-98 E). The Sa + Qz pair is also found as composite inclusions in MI-bearing garnet. As this sapphirine has low  $Fe^{3+}/Fe_{total}$  (0.11-0.25; Table DR1), the Sa + Qz association is a strong evidence that garnet 99 100 in felsic domains grew at UHT.

101 A *P*–*T* phase diagram was calculated (Figure 2A; see Data Repository for modelling 102 strategy), considering the path proposed by Durgalakshmi et al. (2021) for the last UHT event at 103 Rundvågshetta (Fig. 2A). According to the calculations, garnet may grow in the stability field of 104 Sa + Qz, melt and rutile (i.e., garnet can trap these phases) when following an isobaric cooling at 105  $\approx$ 8 kbar. The model predicts that  $\approx$ 12 vol.% melt is present at peak conditions ( $\approx$ 1040 °C and 8 kbar) and that ≈15 vol.% garnet can form by cooling down to 925°C, trapping inclusions of UHT
melt (Fig. 2A).

Rutile is associated with MI in garnet (Fig. 1B-C) and coexists with zircon and quartz both in garnet and matrix. Figure 2B shows the Zr contents of rutile sorted by textural position: trapped in NI, included in MI-bearing garnet and in the matrix. All three rutile types display a high variability of Zr content, a common feature in high-grade rocks. Considering the highest Zr values as representative of the peak *T* (see Luvizotto and Zack, 2009), the *T* range between the production of melt and its subsequent entrapment in garnet is from >1000° to 930°C (Fig. 2B), indicating that the interpretation of MI as former UHT melts is robust.

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# 116 **GEOCHEMISTRY OF UHT MELTS**

Only GI containing fresh glass have been analysed for major elements and H<sub>2</sub>O. The melts are granitic, weakly peraluminous [ASI (molar Al/(Ca+Na+K)) = 0.95-1.12], sometimes peralkaline (Fig. 3A), typically ferroan (Fig. 3B), with high SiO<sub>2</sub> (76–78 wt.%) and K<sub>2</sub>O + Na<sub>2</sub>O (8.6–10.5 wt.%; Fig. 3C) and with K<sub>2</sub>O/Na<sub>2</sub>O from 2.5 to 7.5. They exhibit low maficity (FeO + MgO = 0.5-1.1 wt.%), Al<sub>2</sub>O<sub>3</sub> (11–12.6 wt.%) and very low CaO (0.06–0.4 wt.%). All GI have low H<sub>2</sub>O contents, usually <1.3 wt.% (measured by Raman; see Data Repository).

Trace elements measured in both NI and GI show that MI are rich in Cs (10–89 ppm), Rb (146–711 ppm) and Ga (20–51 ppm), but contain low Ba (225–890 ppm), Sr (7–48 ppm) and intermediate to low Zr (32–376 ppm; Fig 3D). Zr saturation temperatures (Crisp and Berry, 2022) of the melts vary from 712 to 931°C (see discussion below).

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## 128 THE LINK BETWEEN ANATECTIC UHT MELTS AND A-TYPE GRANITES

Most A-type suites exhibit a large range in silica (55–78 wt.%) and products having  $\geq$ 75 wt.% SiO<sub>2</sub> are considered subordinate (Frost and Frost, 2011). The investigated UHT MI all have SiO<sub>2</sub>  $\geq$ 76 wt.%, but several of their geochemical features (e.g., ferroan character, ASI 0.95-1.1, high alkali and K/Na, Rb and Ga, low Al<sub>2</sub>O<sub>3</sub>, Ba, Sr, and H<sub>2</sub>O) are typical of A-type granites (Fig. 3A-C). Indeed, in the classification diagrams from Whalen et al. (1987), MI plot in the field of A-type granites, together with other high-SiO<sub>2</sub> occurrences described in landmark papers on these rocks (Fig. 3C-D).

136 Zirconium > 200 ppm is considered diagnostic of many A-type granites, and attributed to 137 the dissolution of zircon at HT-UHT conditions (Collins et al. 1982); yet most of our MI show 138 lower Zr values (Fig. 3D). Such discrepancy can be due to the fact that, unlike predictions of 139 equilibrium solubility models, zircon in rocks from Rundvågshetta also grew during prograde path, 140 surviving UHT conditions (Durgalakshmi et al., 2021). Moreover, the fast garnet growth rates, 141 necessary to trap MI, probably prevented melt-zircon equilibrium to be achieved, as documented 142 in other MI studies (Acosta-Vigil et al., 2012). This complexity of anatexis mechanisms imposes 143 caution when converting the Zr content of MI into magmatic T estimates.

144 The restricted, high-SiO<sub>2</sub> content of MI in this study finds analogies in occurrences of A-145 type granites dominated by felsic rocks: Topsails igneous terrane, Western Newfoundland (43% 146 and 57% of analyzed rocks with 70–75 and 76–78 wt.% SiO<sub>2</sub>, respectively; Whalen et al., 1987), 147 Gabo (all with 72–75 wt.% SiO<sub>2</sub>) and Mumbulla (all with 77–79 wt.% SiO<sub>2</sub>) suites of southern Australia (Collins et al., 1982) and White Mountains suite (67% with 70-75 wt.% SiO<sub>2</sub>, 16% >75 148 149 wt.% SiO<sub>2</sub>; Eby, 1990). In the large dataset of Dubray et al. (2018) on ferroan granites from United 150 States, 68% of the samples are >70 wt.% SiO<sub>2</sub>, and some localities (e.g., Dells granite) are almost 151 totally characterized by rocks with  $\geq$ 73 wt.% SiO<sub>2</sub>.

152 As the primary composition of most granites is modified by multiple processes including 153 magma mixing/mingling and/or accumulation of early crystallized feldspars (e.g., Brown et al., 154 2016), we performed geochemical modelling considering the large-ion lithophile elements (Rb, 155 Sr, and Ba) which are sensitive to these processes. Results show that the composition of less felsic 156  $(SiO_2 < 70 \text{ wt.}\%)$  A-type magmas – whose hybrid origin has been proposed (Smithies et al., 2011) 157 - is well reproduced by mixing UHT MI and a mantle-derived component (Fig. 4A and B). 158 Conversely, high-SiO<sub>2</sub> rocks (> 70 wt.%) do not show a clear trend toward the mafic component; 159 rather, there is a partial overlap of MI with some A-type granites and most of compositional 160 variability in the latter can be explained by accumulation of K-feldspar and plagioclase, starting 161 from parental melts represented by UHT MI (Fig. 4A and B). Considering granitoids as crystal 162 cumulates from which interstitial melt may be extracted to form high-SiO<sub>2</sub> volcanic rocks (Deering 163 and Bachmann, 2010), it is not surprising that the predicted composition of evolved model melt is 164 similar to A-type rhyolites. Accumulation of feldspars, as suggested by trace element modelling 165 (Fig. 4A and B), can also extend the silica range to lower values (Fig. 4C). This reinforces the view that the MI from this study can represent the primary melts of high-SiO<sub>2</sub>, weaklyperaluminous A-type granites.

168

#### 169 IMPLICATIONS

170 A-type granites have not been considered to derive from melting of metasediments probably 171 because most experiments of metapelites produced melts that are insufficiently ferroan and/or too 172 strongly peraluminous (Fig. 3A and B; Gao et al., 2016). However, experiments have reproduced 173 only closed systems behaviour, where the composition of UHT melts produced from fertile sources 174 is the integrated mixture of the melts generated over a *T* interval >200 °C.

175 The pristine natural UHT melts investigated in this study, together with in-source context 176 in which they occur, show instead that residual metasedimentary crust can produce melts 177 representing high-SiO<sub>2</sub> end-members in the wider compositional spectrum of A-type igneous 178 suites. Therefore, our findings are also consistent with a metasedimentary origin of A-type granites 179 suggested on the basis of zircon, whole-rock and isotopic data (e.g., Huang et al., 2011; Zhang et 180 al., 2022) and indicate that anatexis of metasediments should not only be correlated to S-type 181 granite magmatism. Indeed, a weakly peraluminous character (ASI=1) is expected for melts with 182 <1 wt.% H<sub>2</sub>O, despite the strongly peraluminous source (Acosta Vigil et al., 2003).

183 Other MI studies (Cesare et al., 2015) of less residual metasedimentary sources report 184 strongly peraluminous S-type granite compositions at T approaching UHT conditions (Cesare et 185 al., 2015), whereas in this study a residual source produces melts akin to high-SiO<sub>2</sub> A-type granites. 186 Hence, our results question the fertile-source model (Creaser et al., 1991; Frost and Frost, 2011). 187 Rather, we argue that pre-conditioning of the crust (i.e., when a large amount of melt has 188 previously been extracted and thermal buffering of melting reactions is less efficient; Vielzeuf et 189 al., 1990) may be essential in the formation of A-type granites. This would allow the achievement 190 of UHT conditions (with or without the mantle involvement), promoting further low-fraction 191 melting of a residual protolith (12 vol.% melt at 1000 °C; this study). The latter can have a 192 metasedimentary origin (this study; Huang et al., 2011; Zhang et al., 2022) or an igneous one 193 (Collins et al., 1982), resulting in the formation of S- and I-type melts during previous anatexis.

A-type granites are less voluminous than S- and I-types in the crust, and when present they often post-date them (Collins et al., 1982; 2021). This is consistent with the residual character of the crustal source and with the fact that UHT conditions in the crust are subordinate. Nonetheless,

considering the size of exposed UHT terranes (largest being >10000 km<sup>2</sup>; Qi et al., 2022), the 197 198 inferred thickness of crust that could undergo UHT melting (ca. 15 km, Zhang et al., 2022) and 199 the often long-lived duration of UHT metamorphism (Harley, 2016), it is possible that even small 200 degrees of melting can generate significant volumes of A-type granites. For example, the low-201 degree (10%) melting of a UHT terrane of 10000 km<sup>2</sup> by 10 km thickness could feed a 1 km-thick 202 intrusion of A-type granite covering the entire terrane. This conceptual model is supported by the 203 spatial and temporal association of UHT metamorphism and A-type granite magmatism 204 throughout eastern Gondwana terranes in Antarctica, Madagascar, and southern India (Jacobs et 205 al., 2008; Sato et al., 2010; Tsunogae and Santosh, 2010). These findings call for a revaluation of 206 the commonly accepted view that only S-type granites image the geodynamic evolution of the 207 high-grade metasedimentary crust.

208

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Figure 1. A) Macroscopic view of the metapelitic UHT granulite from Rundvågshetta. B) Garnet (Grt) with abundant
inclusions in its core, C) Negative crystal shaped glassy melt inclusion (GI) associated to rutile (Ru) in Grt. Inset:
Raman spectra (cm<sup>-1</sup>) of the glass (blue) and bubble (pink). D) Nanogranitoid (NI) containing trapped Ru + sapphirine
(Sa) + quartz (Qz). E) BSE image of the NI in (D). F) BSE image of a NI containing K-feldspar (Kfs), biotite (Bt) and
Qz.



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Figure 2. A) *P*–*T* phase diagram showing stability fields of key minerals, with garnet and melt modes. D21: path from Durgalakshmi et al. (2021). B) T (°C) and Zr contents (ppm) of rutile in nanogranitoid (in NI), in garnet (in Grt) and in the matrix. Temperatures calculated at 8 kbar (Table DR4). Horizontal bars are median, and colored boxes highlight the whole variability in each microstructural position. T>900°C (UHT metamorphism) is highlighted in grey.



Figure 3. Geochemical features of UHT melts from this study compared with A-type granites (SiO<sub>2</sub> > 70 wt%) and UHT experimental melts from metasedimentary protoliths (Gao et al., 2016). A) Plot of alkalinity index (AI = molar Al-Na+K) vs. aluminium saturation index (ASI), and B) Fe-index (FeO<sub>t</sub>/FeO<sub>t</sub> + MgO) vs. SiO<sub>2</sub> (from Frost and Frost, 2011). C and D) Classification diagrams from Whalen et al. (1987). Maxima and minima of K<sub>2</sub>O + Na<sub>2</sub>O in (C) are used for different Ga/Al ratios, since only MI below garnet surface were analysed for trace elements. Data from the literature: C1982 (Collins et al., 1982); W1987 (Whalen et al., 1987); E1990 (Eby, 1990); W2002 (Wu et al., 2002); S2011 (Smithies et al., 2011).





Figure 4. Plots of A) Sr vs. Rb, B) Sr vs. Ba and C) SiO<sub>2</sub> vs. ASI for UHT MI and A-type granites. Rayleigh fractional
crystallization models for trace elements use the average MI as starting composition and track the evolution of
cumulate and residual melt, considering different partition coefficients and crystallization of 70% Kfs and 30% Pl (see
Data Repository). A mixing model is also reported. Squares (steps of 10%) indicate the fraction of melt remaining or

- the amount of mafic magma in the model. A-type granites are described in Fig. 3, A-type rhyolites >75 wt.% SiO<sub>2</sub>
- from Mc2008 (McCurry et al., 2008), intermediate to mafic lithotypes (<65 wt.% SiO<sub>2</sub>, S2011) from Smithies et al.
- 241 (2011) and mafic component N1999 from Naumann and Geist (1999).
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