GEOLOGY
E: geology@geosociety.org
W: geosociety.org/gsa
T: @geosociety

The formation of lithium-rich pegmatites through multi-stage melting

Tracking no: G51633R

Authors:
Lot Koopmans (University of Oxford), Tania Martins (Manitoba Geolgical Survey), Robert Linnen (Western University), Nicholas Gardiner (University of St Andrews), Catriona Breasley (University of British Columbia), Richard Palin (University of Oxford), Lee Groat (University of British Columbia), David Silva (University of British Columbia), and Laurence Robb (University of Oxford)

Abstract:
Lithium-cesium-tantalum type pegmatites - the primary source of lithium - crystallize from highly evolved, volatile- felsic melts that incorporated crustal material in their source. Pegmatites are classically thought to form either from extreme fractionation of a parental granite body or via low-degree partial melting of a metamorphic rock (anatectic-origin). However, the processes that lead to the formation of economic lithium pegmatite deposits remain enigmatic, since precipitation of lithium ore minerals requires melt lithium concentrations in excess of 5000 ppm - approximately 500 times upper crustal abundances. Here, we use petrological modeling to quantify lithium enrichment in an anatectic-origin scenario and show that it is primarily driven by the relative stability of residual biotite and muscovite at medium to high pressures (~8 kbar), and biotite and cordierite at low pressures (~3 kbar). We show anatexis of an average lithium-enriched metasedimentary source cannot sufficiently elevate the lithium content of the ensuing melt to form economic deposits; however, if this first-generation melt - now crystallized as granitic crust - is re-melted, the second-generation melt will be sufficiently concentrated in lithium to crystallize lithium ore minerals. We propose a petrogenetic model for anatectic-origin lithium pegmatites, in which a region experiences at least two stages of partial melting, ultimately generating lithium-rich melts without invoking extensive fractional crystallization. This mechanism can account for both the occurrence of unzoned lithium pegmatites and explains why economic pegmatites in many terranes are younger than their inferred source granites.

The formation of lithium-rich pegmatites through multi-stage melting

Lot Koopmans[1], Tania Martins[2], Robert Linnen[3], Nicholas J. Gardiner[4], Catriona M. Breasley[5], Richard M. Palin[1], Lee A. Groat[5], David Silva[5], and Laurence J. Robb[1]

[1]Department of Earth Sciences, University of Oxford, 3 South Parks Road, Oxford, OX1 3AN United Kingdom

[2]Manitoba Geological Survey, 360-1395 Ellice Avenue, Winnipeg, Manitoba, Canada, R3G 3P2

[3]Department of Earth Sciences, Western University, 1151 Richmond Street N., London, Ontario, Canada, N6A 5B7

[4]School of Earth & Environmental Sciences, University of St Andrews, Bute Building, Queen's Terrace, St Andrews, KY16 9TS, United Kingdom

[5]Department of Earth, Ocean, & Atmospheric Sciences, University of British Columbia, 2207 Main Mall, Vancouver, British Columbia, Canada, V6T 1Z4

ABSTRACT

Lithium-cesium-tantalum type pegmatites – the primary source of lithium – crystallize from highly evolved, volatile- felsic melts that incorporated crustal material in their source. Pegmatites are classically thought to form either from extreme fractionation of a parental granite body or via low-degree partial melting of a metamorphic rock (anatectic-origin). However, the processes that lead to the formation of economic lithium pegmatite deposits remain enigmatic, since precipitation of lithium ore minerals requires melt lithium concentrations in excess of 5000 ppm – approximately 500 times upper crustal abundances. Here, we use petrological modeling to quantify lithium enrichment in an anatectic-origin scenario and show that it is primarily driven

by the relative stability of residual biotite and muscovite at medium to high pressures (~8 kbar), and biotite and cordierite at low pressures (~3 kbar). We show anatexis of an average lithium-enriched metasedimentary source cannot sufficiently elevate the lithium content of the ensuing melt to form economic deposits; however, if this first-generation melt – now crystallized as granitic crust – is re-melted, the second-generation melt will be sufficiently concentrated in lithium to crystallize lithium ore minerals. We propose a petrogenetic model for anatectic-origin lithium pegmatites, in which a region experiences at least two stages of partial melting, ultimately generating lithium-rich melts without invoking extensive fractional crystallization. This mechanism can account for both the occurrence of unzoned lithium pegmatites and explains why economic pegmatites in many terranes are younger than their inferred source granites.

INTRODUCTION

Lithium–cesium–tantalum (LCT) pegmatites are formed from highly fractionated melts (Černý, 1991) and dominate global lithium (Li) production (U.S. Geological Survey, 2023). LCT pegmatites first appeared in the Neoarchean (c. 2.8 Ga), are dominantly hosted in greenschist- to amphibolite-grade supracrustal rocks (Dittrich et al., 2019), and retain geochemical signatures which imply their genesis involved reworking of existing (typically metasedimentary) crust (Černý, 1991). However, fundamental questions remain regarding how these evolved rocks become highly concentrated in Li, since the primary extractable Li ore minerals – the Li-aluminosilicates spodumene and petalite – only crystallize when melt Li contents exceed 5000 ppm (London, 1984; Maneta et al., 2015), representing an enrichment of ca. 500 times above ordinary crustal abundances (Taylor and McLennan, 2003). Furthermore, to be economically viable, an economic Li-pegmatite deposit must contain a sufficiently large mineralized zone

(Bradley and Mccauley, 2017), the extent of which is controlled by the initial Li concentration in the emplaced pegmatitic melt (London, 2014).

In general, concentrating incompatible elements such as Li within a small-volume melt fraction occurs through either (i) high-degree crystal fractionation of a larger body of magma, or (ii) siphoning low-degree (e.g., 7–10%) partial melts away from a source rock undergoing anatexis. These contrasting processes underpin two proposed LCT pegmatite petrogenetic models: (1) extreme fractionation of a granitic magma that carries a typical (crustal) Li concentration, generating a minor volatile- and metal-rich melt fraction which migrates from the source region and ultimately crystallizes as a pegmatite (Cameron et al., 1949; Jahns, 1953; Černý, 1991; Černý et al., 2005), or (2) low-degree partial melting of Li-rich host rocks during prograde metamorphism ('anatectic origin'), generating a volatile-rich melt that does not require significant additional fractionation (Stewart, 1978; Simmons et al., 1995; Shaw et al., 2016; Müller et al., 2017).

Recent research has focused on the anatectic model as a viable mechanism to form economic pegmatite deposits (Müller et al., 2017; Kunz et al., 2022; Knoll et al., 2023). Here, we use petrological modelling to quantify the extent of Li enrichment during crustal melting of metasedimentary protoliths, and then assess how subsequent melting of their crystallized magmatic products might serve to further concentrate Li in newly formed melt fractions. Our results imply that multi-step anatexis in long-lived orogenic systems can act as a highly efficient mechanism to produce economic grade LCT pegmatites.

MODELING LI ENRICHMENT DURING CRUSTAL ANATEXIS

Lithium in metasedimentary rocks is primarily hosted in biotite, muscovite, cordierite, and staurolite (Simons et al., 2017; Kunz et al., 2022; Knoll et al., 2023). During anatexis, the Li content of a melt is controlled both by the breakdown of these minerals, and by the partitioning of Li between restitic minerals and melt as they re-equilibrate. We examined Li mineral–melt partitioning during crustal anatexis via a batch melting geochemical model, which assumes that generated melt remains in equilibrium with the source rock until extraction (Rosenberg and Handy, 2005); an assumption validated in experimental studies (e.g., Acosta-Vigil et al., 2012). The onset of melting, as well as the stability and thus presence of restitic minerals in equilibrium with the melt, depends on the source composition and thermobarometric conditions of melting. As such, we performed Gibbs free energy minimization calculations using the petrological modeling program Theriak-Domino (De Capitani and Petrakakis, 2010) to determine the stable mineral–silicate melt assemblages at different pressure (*P*, i.e. depth) and temperature (*T*) conditions within the continental crust for two putative metasedimentary source rocks: the average metapelite of Ague (1991) and a greywacke (Composition I of Pettijohn (1963)). Although Li contents vary between metasedimentary rocks, for ease of comparison we assume both compositions have a starting Li composition of 125.5 ppm, the average of 11,634 siliciclastic sedimentary rocks worldwide (see Supplementary Material).

We calculated *P*–*T* phase diagrams for both metasedimentary protoliths and examined the crystallized products of extracted melts. Initial anatexis was examined along isobaric heating paths at 3 kbar (~9 km depth, i.e., intrusion-related heating) and 8 kbar (~24 km, i.e., a standard orogenic geotherm intersecting the solidus) under both dehydration melting (minimal saturation) and flux melting scenarios to quantify how Li behaves during partial melting. Our approach allowed the calculation of changes in melt Li content during crustal anatexis and then the

behavior of that melt fraction after extraction (set at 7 vol. % melt, the threshold at which melt can escape its source, after Rosenberg and Handy, 2005).

Melt Li concentration was then modeled for two end-member scenarios: (1) the melt extracted undergoes fractional crystallization in a closed system; and (2) the resulting extracted melt crystallizes to form a granitic body which then undergoes a second (later) stage of anatexis. Bulk Li partition coefficients for fractional crystallization were calculated using stable assemblages present at the solidus and published mineral/melt partition coefficients. All models were run through TDMelts (Koopmans et al., 2023) with the setup described in the Supplementary Material.

During melting, both metasedimentary compositions (Figure 1a and Supplementary Material) stabilize cordierite and biotite at low pressures, whereas muscovite and biotite are stable at higher pressures. Figure 1 (b) shows the predicted partitioning of Li between metasediment-derived melt and residuum for both starting compositions. At fluid-present conditions, the solidus occurs between 660 and 690 °C and the release of Li into the melt is largely controlled by the initial breakdown of muscovite and later breakdown of biotite.

Metapelites are typically enriched in Al compared to greywackes and stabilize higher proportions of cordierite and mica at equivalent *P–T* conditions. At equivalent pressures, greywacke-derived melts contain higher Li contents (by a factor of 1.3–2.1) than metapelite-derived melts (Figure 1b); this difference is amplified at higher pressure due to the extended stability of mica compared to cordierite.

Micas are the main hosts of Li in the subsolidus, and our models confirm that Li enrichment during partial melting of metasedimentary rocks is primarily controlled by biotite and muscovite stability, with cordierite having a minor influence, and feldspar playing an

insignificant role (Figure 1), agreeing with previous work (Kunz et al., 2022; Knoll et al., 2023). Li is released into the melt upon mica breakdown, but importantly, the persistence of micas during melting sequesters Li and serves to inhibit melt Li enrichment.

At a melt proportion of ~7 vol.%, we calculate maximum Li concentrations of 411 ppm at 788 °C and 538 ppm at 785 °C for metapelites and metagreywackes, respectively. These values represent a potential maximum Li enrichment factor of ~4.3 at extraction compared to the source. Additionally, flux melting (i.e., ingress of external H_2O) also serves to depress melt Li enrichment under all conditions, largely due to the higher proportion of micas present at melt extraction conditions that have not broken down during melting reactions, although we note melts are extractable at a significantly lower temperature (Figure 1b).

Fractional crystallization (our scenario 1) during cooling may further enrich the melt phase in incompatible elements including Li. Modeled Rayleigh fractionation trends for melts extracted at the 7 vol. % threshold are shown in Figure 2. In all cases, the concentration of Li reaches 800–1800 ppm at 75% fractionation and 1500–3400 ppm at extreme fractionation conditions (Zhao et al., 2022), still below the minimum saturation point for spodumene. Spodumene saturation is only achieved at low pressure (3 kbar) during the final stages of crystallization (Figure 2), when the melt fraction is reduced below 5 vol. %, a scenario which is not physically extractable (Vigneresse et al., 1996).

Alternatively (scenario 2), the melt produced during anatexis of a metasedimentary protolith may crystallize as a granitic body in the crust and experience a separate, later stage of melting. We modeled the remelting of the granitic compositions produced through primary anatexis (Figure 1c, d) to calculate potential second stage melt Li concentrations. In this scenario, melt first appears between 640 °C and 675 °C, and coexists with a smaller proportion

of micas (<8 wt. %) relative to metasedimentary rocks, and no stable cordierite in any of the modeled scenarios (Figure 1c). Accordingly, the secondary melting stage generates significantly higher melt Li concentrations of 3590–7496 ppm at a 7 vol. % extraction point (Figure 1d), equal to a potential Li enrichment factor of ~14.2 (Figures 1d, 3). These results importantly indicate that a second stage of melting substantially enriches Li; in two stages driving Li concentration from 125.5 to 7496 ppm, with the resultant secondary melts having economic potential without invoking fractional crystallization.

MULTI-STEP DISTILLATION MODEL FOR LI PEGMATITES

Our results show that partial melting of typical metasedimentary sources followed by closed-system fractional crystallization of those melt fractions cannot concentrate Li to the saturation limit required (>5000 ppm) to generate a large zone of Li-aluminosilicates within a pegmatite. Thus, production of a melt capable of saturating Li ore minerals in economic quantities within a single stage melt scenario would require a rare, extremely enriched protolith (e.g., 1160 ppm in the metasedimentary rock). While such concentrations have been documented in reworked Li-rich volcanic sediments (e.g., Kadir et al., 2023), they are typically not documented near Li-rich pegmatite provinces (e.g., Roda Robles et al., 1999) and therefore their role in Li pegmatite formation is equivocal. By contrast, additional processing (i.e., re-melting) of a package of continental crust can increase the Li concentration in a second-generation in order to allow spodumene/petalite saturation very early in the fractionation history, obviating the need for extensive fractional crystallization.

We therefore propose the following petrogenetic model for anatectic-origin Li pegmatites with economic potential (Figure 4):

(1) Partial melting of a metasedimentary succession during prograde metamorphism, which produces a granitic melt that is modestly enriched in Li;

(2) This melt crystallizes as a granitic intrusion structurally above the migmatite zone;

(3) A subsequent melting event, either an extension of the initial metamorphic event or later during an unrelated orogenic cycle, reheats and remelts the granite forming a highly enriched melt that ultimately crystallizes as a Li-rich pegmatite.

Our two-stage anatectic model indicates that the size of the pegmatite generated is approximately 200 times smaller than the metasedimentary protolith; for example, a deposit the size of Tanco (0.021 km^3, Stilling et al., 2006) likely formed from an initial metasedimentary package of c. 4.5 km^3.

This mechanism also accounts for two particular features of pegmatites. Firstly, pegmatites can be unzoned with economic minerals homogenously distributed throughout, requiring Li-bearing minerals to crystallize from the margin inwards (e.g., Kings Mt pegmatites; Swanson, 2012); Mt Cattlin pegmatites, Sweetapple et al., 2019). This suggests the emplaced melt has sufficiently high Li concentrations prior to crystallization.

Secondly, where pegmatite fields have a spatial relationship with granites, they are often significantly younger (e.g., Stilling et al., 2006), which is at odds with arguments for the rapid crystallization of pegmatites from the granitic melt when standard magmatic fractionation is invoked (Simmons and Webber, 2008). Alternatively, many orogenic events are long-lived and experience multiple cycles of melting (e.g., Mulcahy et al., 2014). Together, this suggests the possibility of granites remelting as a mechanism for pegmatite genesis (e.g., Issia Zone, Brou et al., 2022).

A multi-step anatectic model for the formation of Li pegmatites therefore satisfies these geochemical and geochronological constraints, providing an efficient mechanism to elevate Li concentrations and generate economic Li deposits.

ACKNOWLEDGMENTS

We thank delegates at the Maine Pegmatite Workshop, 2022, for fruitful discussions that inspired this work. This work was funded by a National Environmental Research Council, UK grant (no. NE/S007474/1) awarded to LK, a Natural Science and Engineering of Canada Discovery Grant to RL, and a Mitacs Accelerate Award to LG and RL for intern CB. We thank Kathryn Goodenough and two anonymous reviewers for their insightful comments, and Andrew Barth for editorial handling of the manuscript.

REFERENCES CITED

Acosta-Vigil, A., Buick, I., Cesare, B., London, D., and Morgan, G.B., 2012, The Extent of Equilibration between Melt and Residuum during Regional Anatexis and its Implications for Differentiation of the Continental Crust: a Study of Partially Melted Metapelitic Enclaves: Journal of Petrology, v. 53, p. 1319–1356.

Ague, J.J., 1991, Evidence for major mass transfer and volume strain during regional metamorphism of pelites: Geology, v. 19, p. 855–858.

Bradley, D., and Mccauley, A., 2017, A Preliminary Deposit Model for Lithium-Cesium-Tantalum (LCT) Pegmatites: US Geological Survey, doi:10.3133/ofr20131008.

Brou, J.K., Van Lichtervelde, M., Kouamelan, N.A., Baratoux, D., and Thébaud, N., 2022, Petrogenetic relationships between peraluminous granites and Li- Cs-Ta rich pegmatites in south Issia zone (Central-West of Côte d'Ivoire): Petrography, Mineralogy, Geochemistry and zircon U–Pb Geochronology: Mineralogy and Petrology, v. 116, p. 443–471.

Cameron, E.N., Jahns, R.H., McNair, A.H., and Page, L.R., 1949, Internal Structure of Granitic Pegmatites: Society of Economic Geologists.

Černý, P., 1991, Rare-element Granitic Pegmatites. Part II: Regional to Global Environments and Petrogenesis: GSA today: a publication of the Geological Society of America, https://journals.lib.unb.ca/index.php/GC/article/view/3723 (accessed March 2023).

Černý, P., Blevin, P.L., Cuney, M., and London, D., 2005, Granite-Related Ore Deposits: Economic Geology, p. 337–370.

De Capitani, C., and Petrakakis, K., 2010, The computation of equilibrium assemblage diagrams with Theriak/Domino software: The American mineralogist, v. 95, p. 1006–1016.

Dittrich, T., Seifert, T., Schulz, B., Hagemann, S., Gerdes, A., and Pfänder, J., 2019, Introduction to Archean Rare-Metal Pegmatites: , p. 1–21.

Jahns, R.H., 1953, The genesis of pegmatites: I. Occurrence and origin of giant crystals: The American mineralogist, v. 38, p. 563–598.

Kadir, S., Külah, T., Erkoyun, H., Helvacı, C., Eren, M., and Demiral, B., 2023, Mineralogy, geochemistry, and genesis of lithium-bearing argillaceous sediments associated with the Neogene Bigadiç borate deposits, Balıkesir, western Anatolia, Türkiye: Applied clay science, v. 242, p. 107015.

Knoll, T., Huet, B., Schuster, R., Mali, H., Ntaflos, T., and Hauzenberger, C., 2023, Lithium pegmatite of anatectic origin - A case study from the Austroalpine Unit Pegmatite Province (Eastern European Alps): geological data and geochemical model: Ore Geology Reviews, p. 105298.

Koopmans, L., Palin, R.M., and Gardiner, N.J., 2023, TDMelts: A Theriak-Domino Wrapper for 2-dimensional batch melting models, *in* Goldschmidt 2023 Conference, GOLDSCHMIDT, https://conf.goldschmidt.info/goldschmidt/2023/meetingapp.cgi/Paper/20404 (accessed August 2023).

Kunz, B.E., Warren, C.J., Jenner, F.E., Harris, N.B.W., and Argles, T.W., 2022, Critical metal enrichment in crustal melts: The role of metamorphic mica: Geology, v. 50, p. 1219–1223.

London, D., 2014, A petrologic assessment of internal zonation in granitic pegmatites: Lithos, v. 184–187, p. 74–104.

London, D., 1984, Experimental phase equilibria in the system LiAlSiO4–SiO2–H2O: a petrogenetic grid for lithiumx-rich pegmatites: The American mineralogist, v. 69, p. 995–1004.

Maneta, V., Baker, D.R., and Minarik, W., 2015, Evidence for lithium-aluminosilicate supersaturation of pegmatite-forming melts: Contributions to mineralogy and petrology. Beitrage zur Mineralogie und Petrologie, v. 170, p. 4.

Mulcahy, S.R. et al., 2014, Multiple migmatite events and cooling from granulite facies metamorphism within the Famatina arc margin of northwest Argentina: Tectonics, v. 33, p. 1–25.

Müller, A., Romer, R.L., and Pedersen, R.B., 2017, The sveconorwegian pegmatite province - thousands of pegmatites without parental granites: Canadian mineralogist, v. 55, p. 283–315.

Pettijohn, F.J., 1963, Chemical composition of sandstones, excluding carbonate and volcanic sands: USGS, 1–21 p.

Roda Robles, E., Pesquera Perez, A., Velasco Roldan, F., and Fontan, F., 1999, The granitic pegmatites of the Fregeneda area (Salamanca, Spain): characteristics and petrogenesis: Mineralogical magazine, v. 63, p. 535–558.

Rosenberg, C.L., and Handy, M.R., 2005, Experimental deformation of partially melted granite revisited: implications for the continental crust: Journal of Metamorphic Geology, v. 23, p. 19–28.

Shaw, R.A., Goodenough, K.M., Roberts, N.M.W., Horstwood, M.S.A., Chenery, S.R., and Gunn, A.G., 2016, Petrogenesis of rare-metal pegmatites in high-grade metamorphic terranes: A case study from the Lewisian Gneiss Complex of north-west Scotland: Precambrian research, v. 281, p. 338–362.

Simmons, W.B., Foord, E.E., Falster, A.U., and King, V.T., 1995, Evidence for an anatectic origin of granitic pegmatites, western Maine, USA, *in* Geological Society of America Annual Meeting, New Orleans Abstracts, A411, p. 27.

Simmons, W.B. “skip,” and Webber, K.L., 2008, Pegmatite genesis: state of the art: European Journal of Mineralogy, v. 20, p. 421–438.

Simons, B., Andersen, J.C.Ø., Shail, R.K., and Jenner, F.E., 2017, Fractionation of Li, Be, Ga, Nb, Ta, In, Sn, Sb, W and Bi in the peraluminous Early Permian Variscan granites of the Cornubian Batholith: Precursor processes to magmatic-hydrothermal mineralisation: Lithos, v. 278–281, p. 491–512.

Stewart, D.B., 1978, Petrogenesis of lithium-rich pegmatites: The American mineralogist, v. 63, p. 970–980.

Stilling, A., Cerny, P., and Vanstone, P.J., 2006, THE TANCO PEGMATITE AT BERNIC LAKE, MANITOBA. XVI. ZONAL AND BULK COMPOSITIONS AND THEIR PETROGENETIC SIGNIFICANCE: Canadian mineralogist, v. 44, p. 599–623.

Swanson, S.E., 2012, MINERALOGY OF SPODUMENE PEGMATITES AND RELATED ROCKS IN THE TIN–SPODUMENE BELT OF NORTH CAROLINA AND SOUTH CAROLINA, USA: Canadian mineralogist, v. 50, p. 1589–1608.

Sweetapple, M.T., Grigson, M.W., Tornatora, P., and Urgine, S., 2019, The Archean Mt. Cattlin spodumene pegmatite group and 3D geochemical mapping of large “unzoned” pegmatites of economic significance: Canadian mineralogist, v. 57, p. 803–805.

Taylor, S.R., and McLennan, S.M., 2003, Chemical Composition and Element Distribution in the Earth’s Crust, *in* Meyers, R.A. ed., Encyclopedia of Physical Science and Technology (Third Edition), New York, Academic Press, p. 697–719.

U.S. Geological Survey, 2023, Mineral commodity summaries 2023: US Geological Survey, doi:10.3133/mcs2023.

Vigneresse, J.L., Barbey, P., and Cuney, M., 1996, Rheological transitions during partial melting and crystallization with application to felsic magma segregation and transfer: Journal of Petrology, v. 37, p. 1579–1600.

Zhao, P., Chu, X., Williams-Jones, A.E., Mao, J., and Yuan, S., 2022, The role of phyllosilicate partial melting in segregating tungsten and tin deposits in W-Sn metallogenic provinces: Geology, v. 50, p. 121–125.

Figure 1. Representative results of petrologic modeling of a metasedimentary rock (a, b, here the greywacke at 8 kbar) and granite (c, d, sourced from greywacke at 8 kbar) composition undergoing isobaric heating. Modebox diagrams (a, c) show the equilibrium assemblages across the modelled temperature range. The dashed lines highlight the stable assemblage under flux melting conditions. The graph in (b) traces the concentration of lithium in the melt component of the modelled metasedimentary scenarios. (d) as (b), though of the melt components of the corresponding metasedimentary rocks of (b). Stars denote melt extraction points for each condition, where Flux corresponds to flux melting and Sat. corresponds to dehydration melting. Details of the modelling method are provided in the Supplementary Material. Mineral abbreviations: Amph: amphibole; Bt: biotite; Cd: cordierite; Fsp: feldspar; Gt: garnet; Kfs: Alkali feldspar; Mu: muscovite; Pl: plagioclase; Opx: orthopyroxene; Q: quartz.

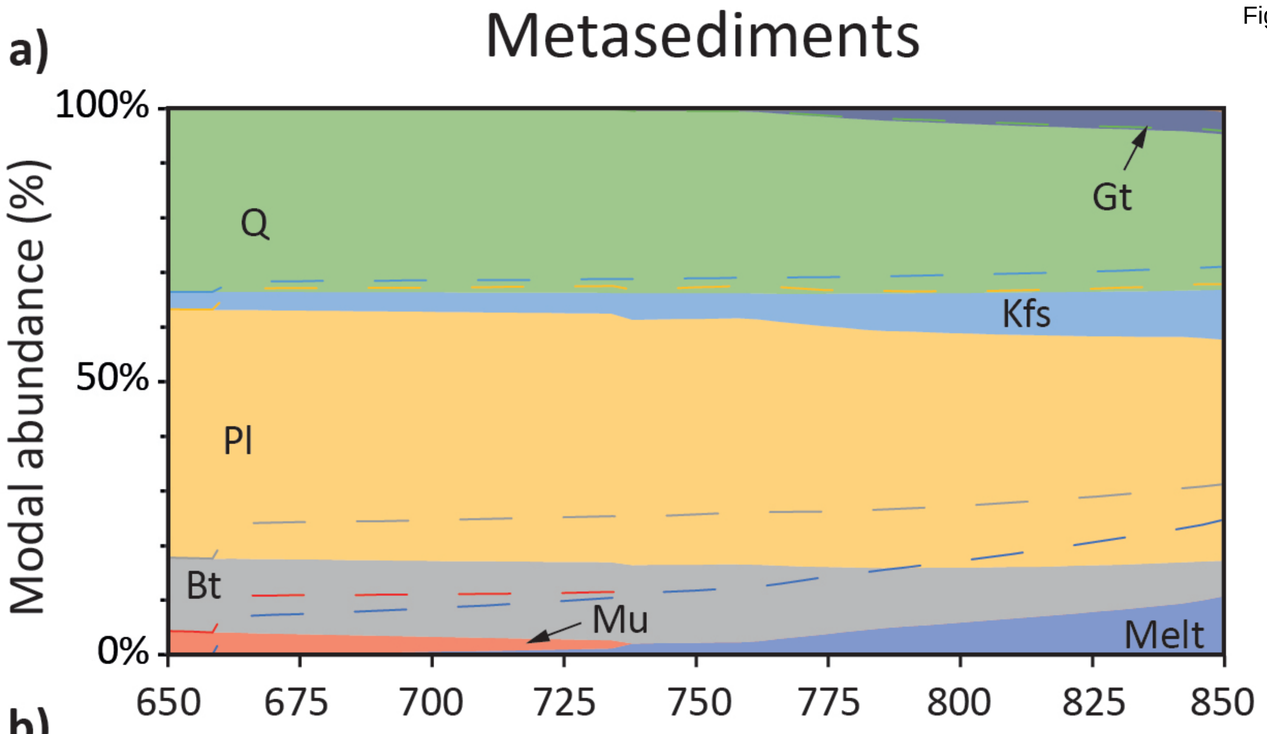
Figure 2. The behavior of lithium (Li) in the extracted melt during Rayleigh fractionation. The initial Li content of each melt corresponds to the concentration of Li at extraction in each of the modelled scenarios. The partition coefficients are taken from the model near the solidus of each individual melt. The particle-locking threshold (PLT) of Vigneresse et al. (1996) and the extreme fractionation boundary of Zhao et al. (2022) are highlighted, as well as the minimum threshold for spodumene saturation (Maneta et al., 2015).

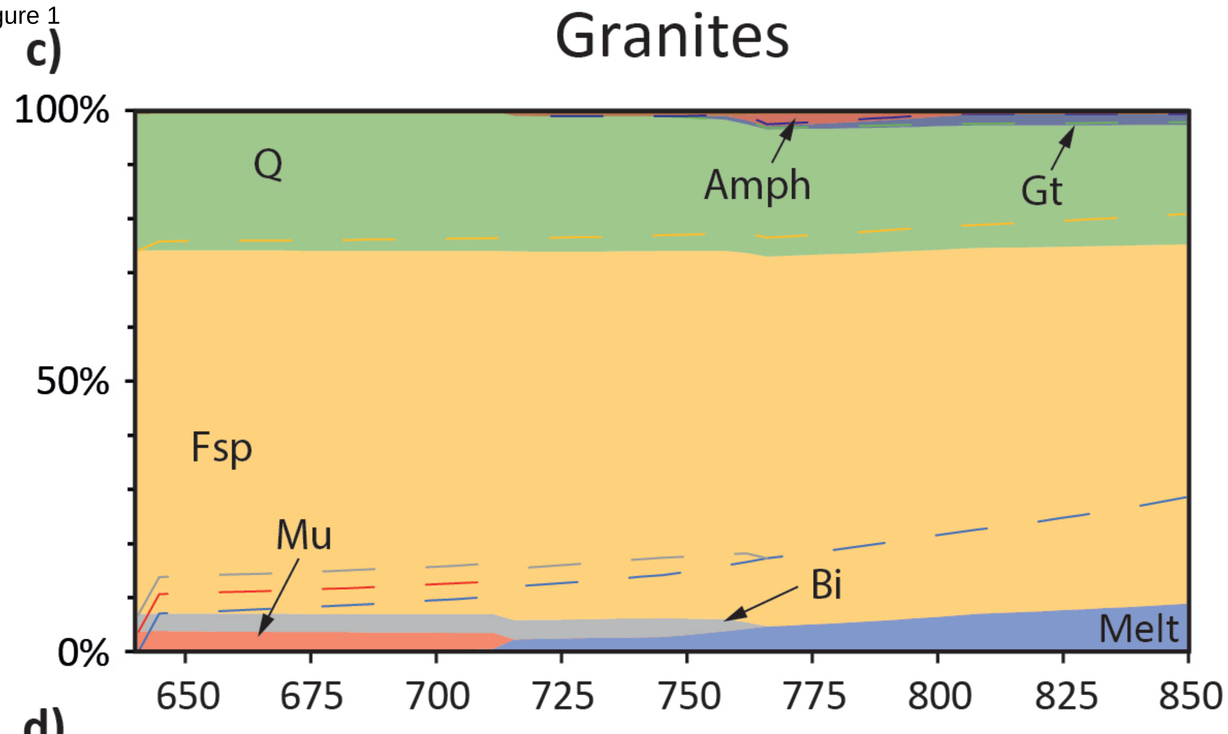
Figure 3. Evolution of Li concentrations during melting and fractionation of melt components generated during crustal anatexis – with the spodumene saturation boundary highlighted (Maneta et al., 2015). After initial melt extraction, the granite either: 1) undergoes fractional crystallization (hollow symbols, the compositions of fractional crystallization was taken as the composition at 90% crystallization); or (2) is assumed to crystallize and then remelted and extracted at the 7% melt (filled shapes).

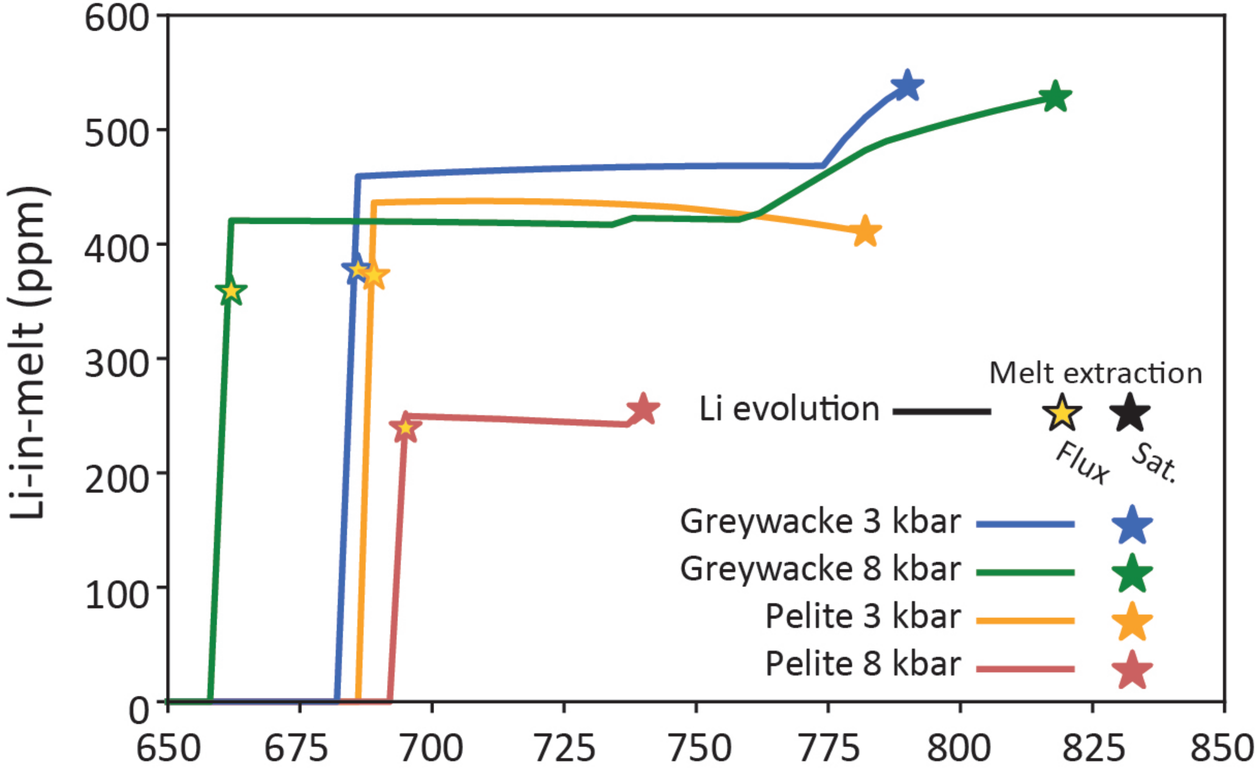
Figure 4. Schematic petrogenetic model for generating lithium (Li)-rich pegmatites in the via crustal anatexis. (A) Partial melting of a metasedimentary rock generates melt lenses, which periodically lose melt. (B) grain-scale diagram visualizing melt films connecting and driving melt out of the system in (A). The breakdown of hydrous phases and mineral-melt diffusion controls the Li budget of the resultant melt. (C) The melt accumulates and crystallizes as a granite structurally above the anatectic zone. (D) A subsequent melting event, either associated with the primary metamorphic event, or during a later orogenic cycle melts the granite, generating Li-rich pegmatites in their vicinity.

[1]Supplemental Material. *Description of model setup and geochemical database used.* Please visit https://doi.org/10.1130/XXXX to access the supplemental material, and contact editing@geosociety.org with any questions.

Figure 1


a)
Metasediments
100%
50%
0%
Modal abundance (%)
Q
Gt
Kfs
Pl
Bt
Mu
Melt
650
675
700
725
750
775
800
825
850


c)
Granites
100%
50%
0%
Q
Amph
Gt
Fsp
Mu
Bi
Melt
650
675
700
725
750
775
800
825
850


b)
Li-in-melt (ppm)
600
500
400
300
200
100
0
650
675
700
725
750
775
800
825
850
Melt extraction
Li evolution
Flux
Sat.
Greywacke 3 kbar
Greywacke 8 kbar
Pelite 3 kbar
Pelite 8 kbar

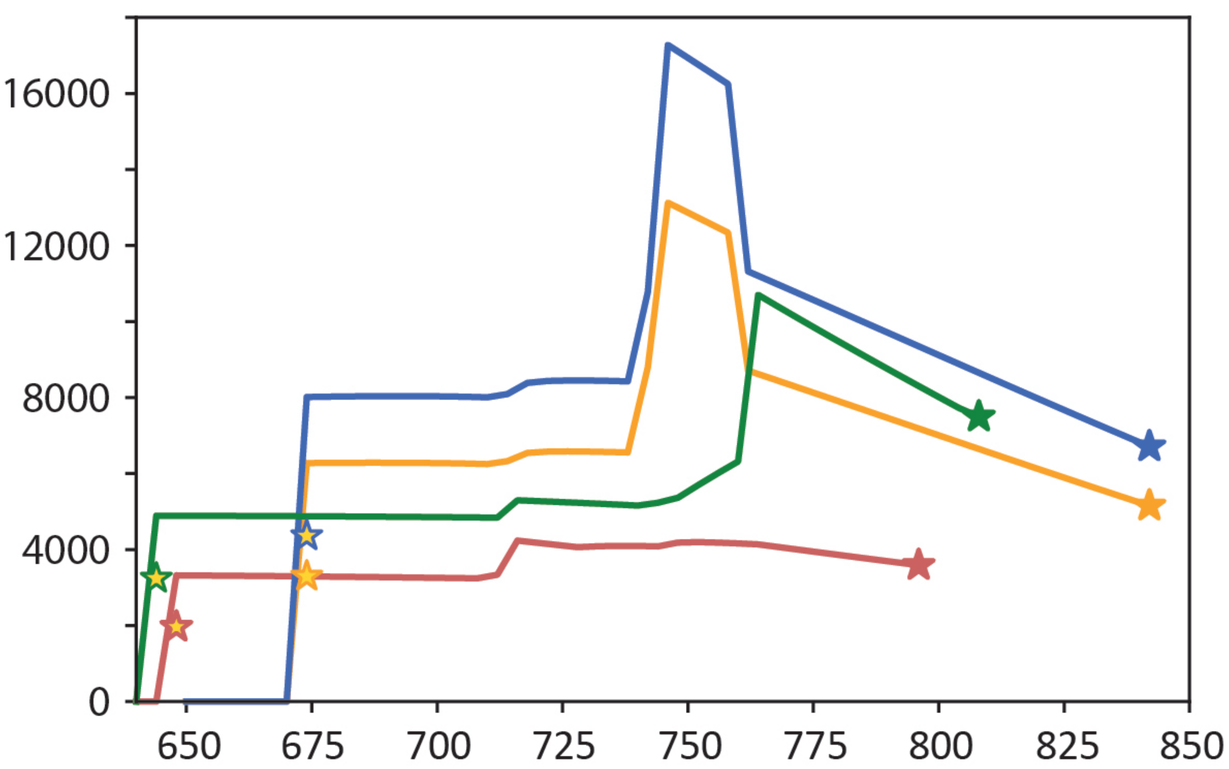

d)
16000
12000
8000
4000
0
650
675
700
725
750
775
800
825
850

Figure 2

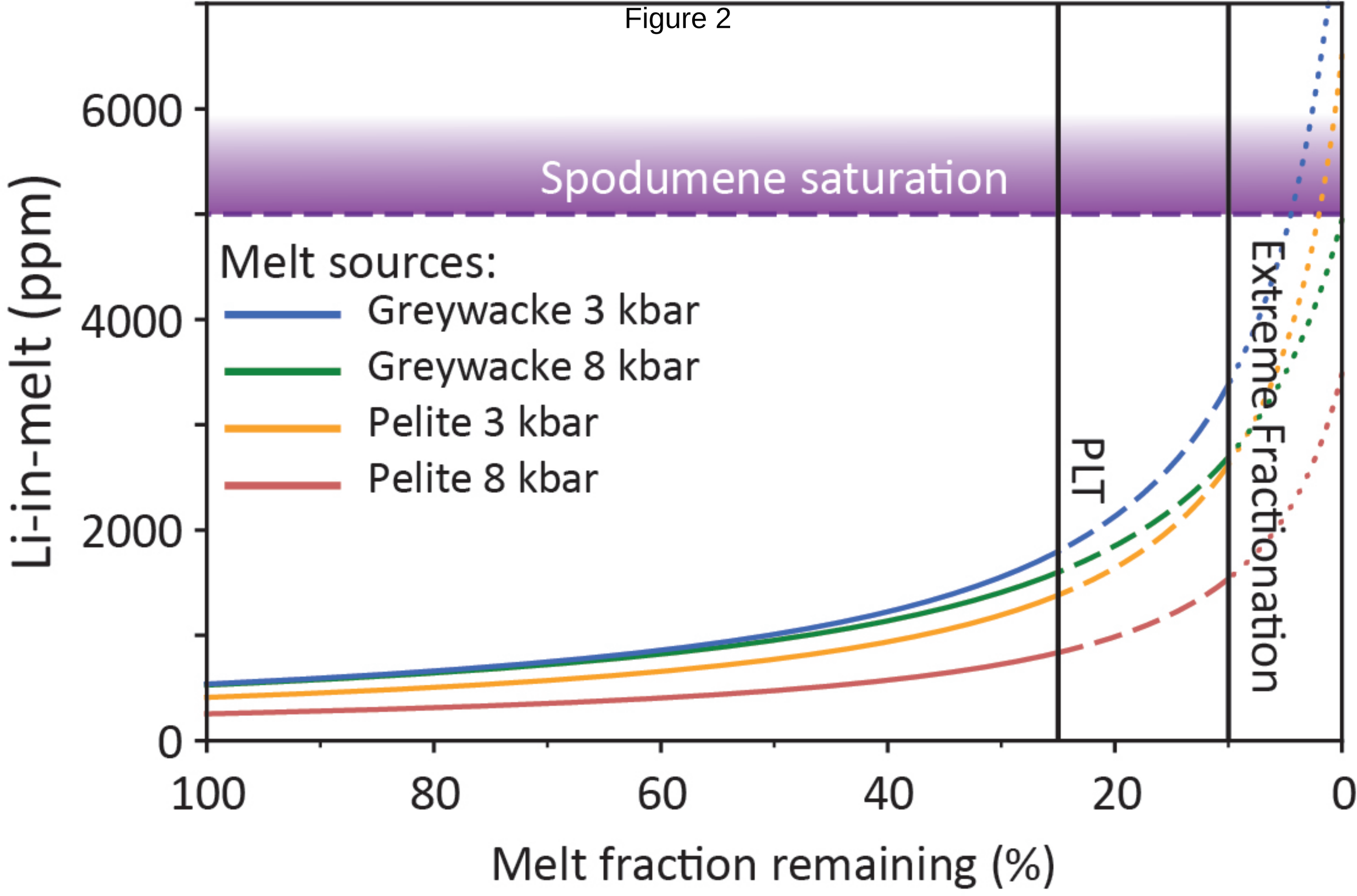

Spodumene saturation
Melt sources:
Greywacke 3 kbar
Greywacke 8 kbar
Pelite 3 kbar
Pelite 8 kbar
PLT
Extreme Fractionation
Li-in-melt (ppm)
6000
4000
2000
0
100
80
60
40
20
0
Melt fraction remaining (%)

Figure 3

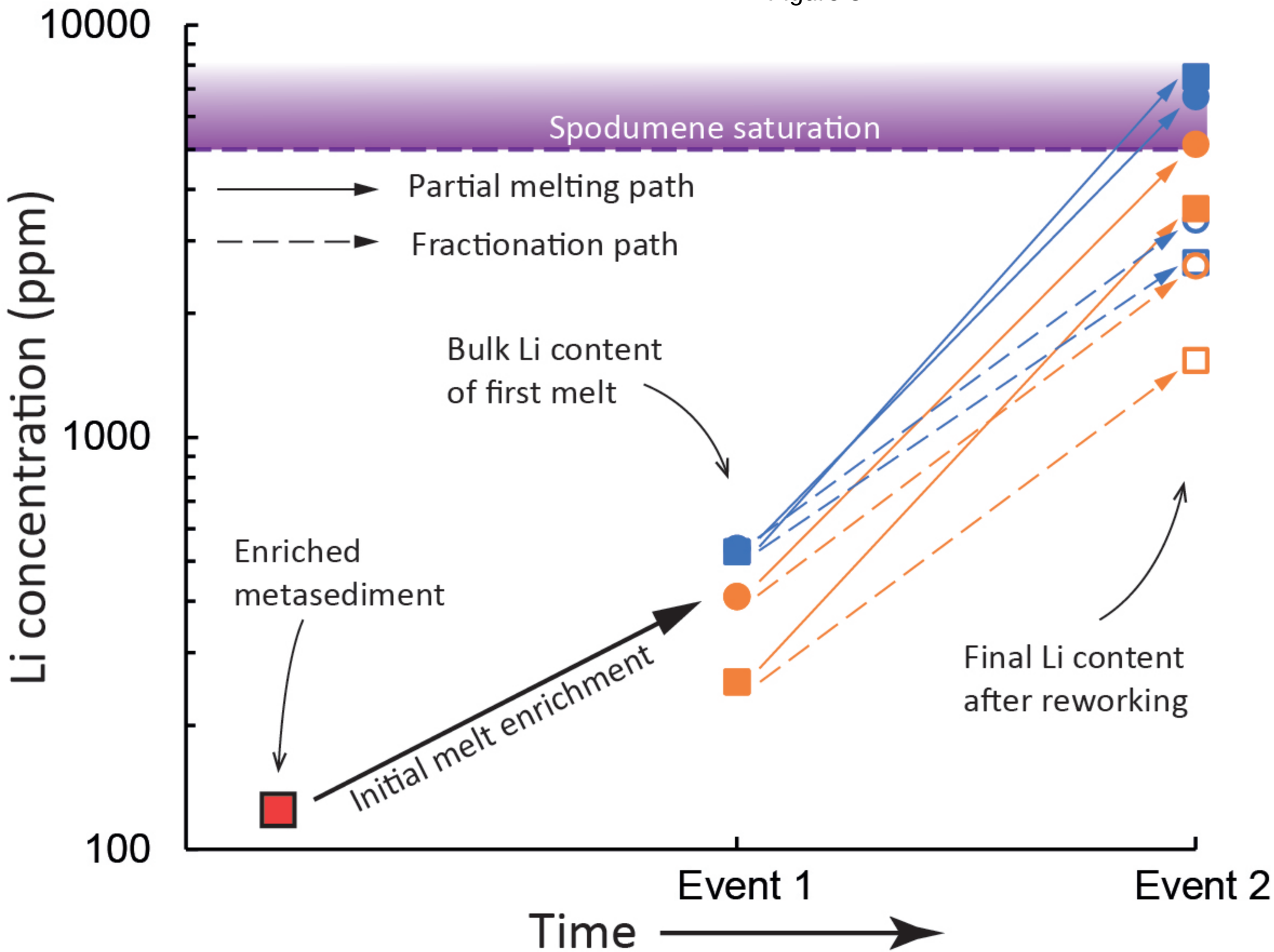

10000
1000
100
Li concentration (ppm)
Spodumene saturation
Partial melting path
Fractionation path
Bulk Li content
of first melt
Enriched
metasediment
Initial melt enrichment
Final Li content
after reworking
Event 1
Event 2
Time
Greywacke
3 kbar
8 kbar
Pelite
3 kbar
8 kbar

Figure 4

Step 1 - Anatexis of metasedimentary rocks

Li in metasedimentary rocks = 125 ppm

Li in melt = 255-538 ppm

A

C

B

Melt
K-feldspar
Plagioclase
Quartz
Biotite

Step 2 - Anatexis of granites

D

Li in pegmatites = 3590-7496 ppm

Li-poor leucosomes

Li-rich pegmatites