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Innovative and multi-disciplinary approach for discussing the emplacement of Variscan LCT-pegmatite fields

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Abstract. In order to define new metallogenic guides for Li, Nb and Ta supply, three Variscan LCT pegmatite fields are investigated. A multi-disciplinary approach is favoured to discuss both the genetic and emplacement models for LCT-type deposits. The spatial statistical analyses demonstrate the high clustering rate of Li-rich pegmatites and the spatial relationships between Li-pegmatites location and surrounding damage fractured zone. The Li-isotopes analyses on micas suggest the absence of genetic continuum between surrounding granites and pegmatites since $\delta^7\text{Li}$ values (‰) are similar through pegmatite field and inside granite samples. To refine these previous results, preliminary numerical models are developed to constrain the ascent of these low viscosity pegmatite-forming melts and determine key criterion of their uncoupling from parental magma source. According to these results, the high permeability seems to play a crucial role in the pegmatite melts collection and during their ascent. The combination of geostatistical, geochemical and numerical tools allows suggesting an alternative model of direct crustal anatexis to the common parental granite model. These high permeability damage shear zones are considered to favour the low volume of pegmatite-forming melt pumping and their escape from their deeper crustal magma source.

Keywords. Variscan belt, LCT-type pegmatites, spatial statistical analysis, Li-isotopes analysis, numerical modelling

1 Introduction

The rare-element pegmatite fields mainly enriched in Li, Nb and Ta (LCT-type defined by Černý and Ercit 2005) are subject to increasing mining exploration in the last few years. These rare- metals are mainly employed in glass industry (Li) and used for superalloys and micro-alloyed steels fabrication (Ta, Nb). Indeed, Li from LCT pegmatite deposits is recognised to be less sensitive to supply disruptions than Li from brine deposits (Bolivia and China), Kesler et al. (2012). In contrast to Li, the main world production of Ta is derived from LCT pegmatite and/or granite deposits whereas a negligible part of Nb- mainly extracted from alkaline complex- is exploited as a by-product of Ta. In order to foster competitiveness on the rare-metals market, new

metallogenic guides need to be defined to meet the growing demand for Li, Nb and Ta, at the European scale. However, genesis and mineralisation processes of LCT pegmatites are still being debated. Despite most of LCT pegmatite fields are interpreted as the product of extreme granitic fractionation (i.e. increase of rare-element content with distance to the consolidating parental granitic source), the connection between a pegmatite and parental granite cannot always be established (Deveaud et al. 2013 and references therein). An alternative model of direct crustal anatexis as responsible for LCT pegmatite genesis has been proposed to explain the presence of these pegmatites without outcropping granite. In contrast to the parental granite model, different melting rates and tectonic features would favour the formation of pegmatite-forming magma with variable rare-element content and would trigger the propagation of pegmatitic magma through continental crust. The aim of this study is to present new data collected in 3 Variscan LCT-pegmatite fields, in order to discuss these two genetic models, and to discover more cost-effective LCT-deposits at European scale.

2 The case study sites

Three Variscan rare-element pegmatite fields have been selected to apply a multi-disciplinary approach to understand the genesis of pegmatite-forming melt, their apparent disconnection from their parental source and their clustered consolidation at the regional scale.

The Monts d'Ambazac (MAPF, France), the Forcarei-Lalin (FLPF, Galicia, Spain) and the Barroso-Alvão (BAPF, Portugal) pegmatite fields present a similar age (~ 305-315 Ma), a similar geodynamical context (syn- to post-collisional) and the same kind of mineralisation (LCT-type deposits). However, some discrepancies exist such as rare-element-bearing major phases (Figure 1) and distinct deformation degree affecting them and their hosting-rocks.

These differences are essential to determine the key criterion for the emplacement of LCT-pegmatite fields and to determine physico-chemical parameters at the origin of the more evolved pegmatites. In order to use efficiently the existing data base (mineralogy, rare-

elements grade, geometry of mineralised bodies), we develop a spatial statistical analysis to characterise the spatial distribution of rare-elements pegmatites from MAPF and BAPF. These results are briefly described below.

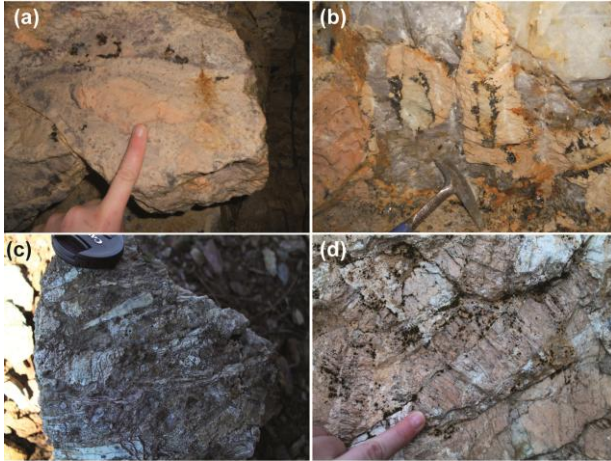


Figure 1. Rare-element-bearing major phases in the Variscan LCT pegmatite fields. (a) petalite and lepidolite (b) beryl crystals observed in the MAPF, (c) spodumene from FLPF, (d) and altered petalite observed in the BAPF.

3 The spatial distribution of the Variscan LCT pegmatites

In this study, all pegmatite occurrences from MAPF (n=118) and BAPF (n=1627) are compiled and stored in a Geographic Information System database. Several methods have been proposed for analysing the spatial pattern point sets such as the Distance to Nearest Neighbour (DNN) and the Ripley's K function to first, understand the pattern of pegmatite -2D- distribution on various scales and second, to highlight spatial relationships between mineralised pegmatites and surrounding lithology and/or tectonic features.

3.1 Spatial pattern of the MAPF

Euclidean distance is computed -in 2D map view- between each pegmatite and its closest neighbouring distinct one, giving an average DNN value equals to 528m. The ratio between the observed average DNN value and the expected DDN value for a purely random spatial distribution equals 0.36 thus highlighting the high clustering degree (Clark and Evans 1954) of the pegmatite occurrences throughout the MAPF. This high clustering rate (occurring on all scales up to 5 000m) defines a strong control of the structures on pegmatite locations. Indeed, pegmatite clusters present a similar orientation ($\sim N020^\circ$) than surrounding faults. However, the spatial statistical analysis based on the Euclidean distance calculation between pegmatites and the surrounding granites, demonstrates the absence of spatial relationships between granite and pegmatites. In addition, there are no spatial relationships between the degree of pegmatite differentiation and the considered parental granite, as defined by the classical LCT-pegmatite genesis model (London 2008).

3.2 Spatial pattern of the BAPF

Similar methods have been applied to the BAPF for analysing the spatial pattern of pegmatites. In addition to the MAPF, an interpolation of the regional schistosity (Gumiaux et al. 2003) has been used to determine spatial relationships between the mineralised pegmatites locations and the deformation of the hosting-micaschist. This interpolation allowed adding temporal indices about pegmatite emplacement within the regional deformation.

Our results show that the more evolved pegmatite dikes (spodumene- and petalite-bearing pegmatites) present a higher clustering rate ($R=0.26$) than barren pegmatites ($R=0.59$). Several statistical tests have been developed and we demonstrate that Li-bearing pegmatites (i) are not spatially related to the Cabeiceras de Basto granite considered as parental granite and (ii) are spatially related to surrounding oriented $\sim N160^\circ$ structures. According to the regional schistosity interpolation, the orientation of brittle structures is related to "virgations" affecting the regional schistosity, thus reinforcing the importance of structural controls on pegmatite emplacement (Figure 2).

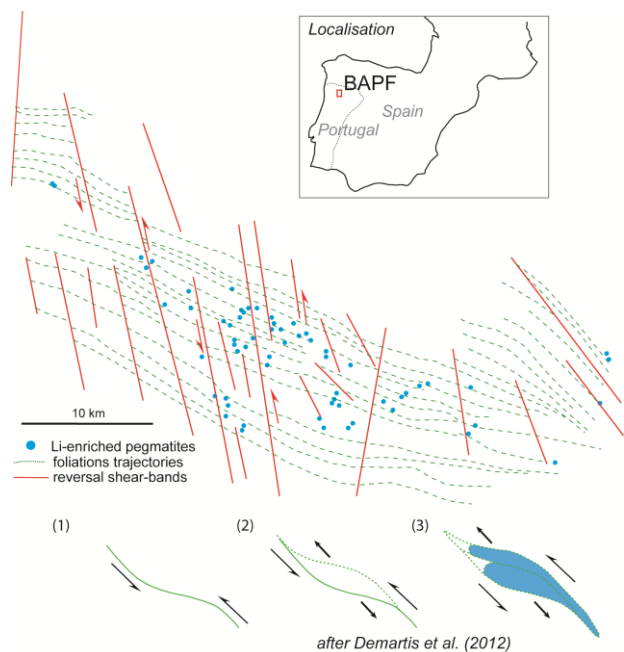


Figure 2. Schematic model of Li-bearing pegmatites emplacement, in the BAPF. (1) Activation of curved shear-zones, (2) opening of releasing bends during motion of shear-zones and (3) magma pumping and crystallisation of pegmatite-forming melt that fills the space.

Thus, we propose that the opening of releasing bands during motion of shear-bands favours magma pumping process and crystallisation of pegmatite-forming melt that replenishes the voids (Figure 2). The high clustering rate of Li-rich pegmatites is coherent with this model. A similar emplacement model has been proposed by Demartis et al. (2011) to explain the location of the LCT pegmatite field of Comechigones (Argentina). The spatial statistical analyses developed in the MAPF

and BAPF demonstrate the key role of structures for pegmatite-forming melt pumping and for the clustered distribution of LCT-pegmatites. However, Li-bearing pegmatites appear to be not issued from the surrounding granites. In order to confirm this result, we analyse the chemical composition of the different pegmatite types (from the less to the more evolved) and of the surrounding granites, which are commonly considered as parental magma for the pegmatite-granite systems.

4 Genesis of LCT-type pegmatites

Since LCT-pegmatites are assumed to be the product of extreme granitic fractionation (London 2008), we propose to investigate $\delta^7\text{Li}$ signatures of micas from the MAPF and from the surrounding Saint-Sylvestre granite (Figure 3). Micas are favoured since they constitute the main Li-bearing phase in pegmatite and they crystallised during all the pegmatite consolidation. A granitic origin for these pegmatites would be characterised by an increasing $\delta^7\text{Li}$ value from the granite to the more-evolved pegmatites (type VI), following Barnes et al. (2012).

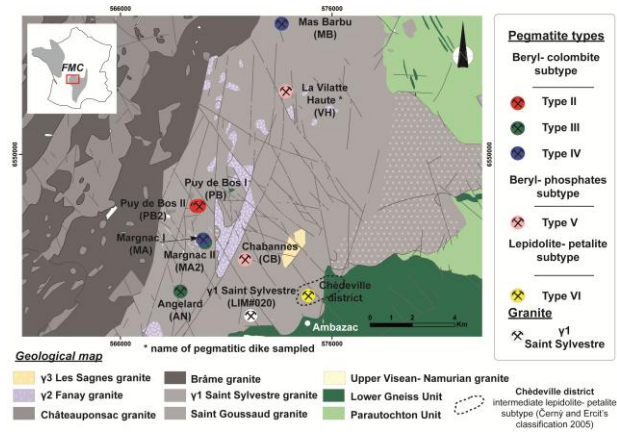


Figure 3. Geological map of the MAPF, modified from Deveaud et al. (2013). Each pegmatite sampled during this study is located on the map. Pegmatites are grouped according to their differentiation degree, from type II to type VI.

4.1 $\delta^7\text{Li}$ values for granite and pegmatite micas

After mica sample digestion, Li-isotopes analyses were performed on a double focusing Neptune MC-ICP-MS (ThermoFinnigan) using the standard-sample bracketing method on L-SVEC standard (NIST8545) as reference material.

Li-isotopes signatures obtained on granite and pegmatite mica range from -3.5 to +3.5 ‰ (Figure 4).

The Li-isotopes fractionation inside dark- and white mica is independent from the magmatic fractionation degree of mica sampled (Figure 5). Moreover, we do not observe distinct $\delta^7\text{Li}$ signatures between mica from Saint-Sylvestre granite and from the MAPF. Most of pegmatites present similar $\delta^7\text{Li}$ values which are independent from the pegmatite type.

Consequently, we suggest that (i) the MAPF is not issued from the surrounding Saint-Sylvestre granite and, (ii) the more evolved pegmatite (type V and type VI) are

not issued from the extreme fractionation of the less evolved pegmatite dikes (type II, III and IV).

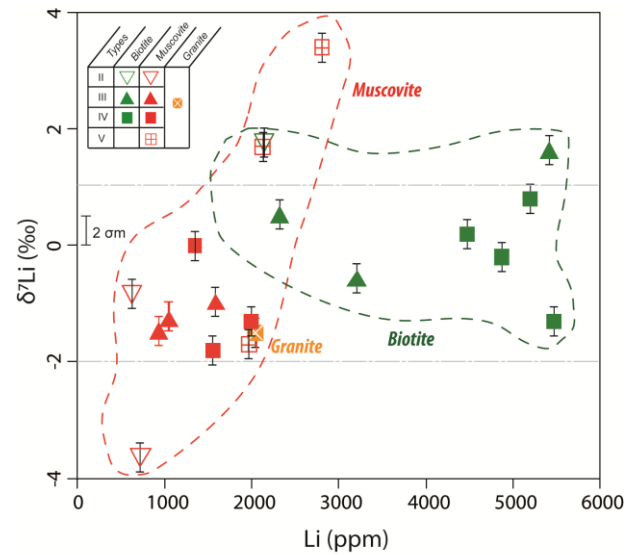


Figure 4. Evolution of $\delta^7\text{Li}$ (‰) with Li-content obtained on micas from the MAPF and Saint-Sylvestre granite. Each pegmatite type has been distinguished (from the type II to the more evolved type V).

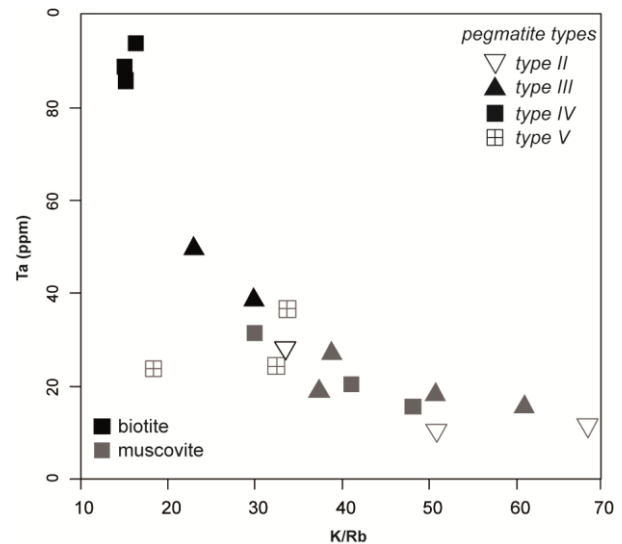


Figure 5. Rare-element content of dark- and white mica solutions from the MAPF. Ta concentration from distinct pegmatite types are plotted against K/Rb ratio. In contrast to Ta, this element ratio is known to decrease with the increase pegmatite fractionation degree.

These Li-isotopes results are coherent with the spatial statistical analyses previously obtained which suggest the absence of relationships between pegmatites and surrounding granite. In addition, the independence of pegmatite chemical composition from each other highlights the key role of parental magma source. The absence of relationship between magmatic fraction trend and Li-isotopes fractionation suggest that these $\delta^7\text{Li}$ signatures are inherited from a crustal source that is common to granite and LCT-pegmatites. However, mechanisms and physico-chemical processes allowing the genesis of peraluminous two-mica granite or the emplacement of LCT-pegmatites are still unknown. To investigate these processes, we used a numerical

approach.

5 The escape of pegmatite-forming melt: numerical investigation

Our previous results demonstrate the main role of tectonic context during pegmatite-forming melt genesis and their propagation through activated shear-bands, opening of tension gashes, magma pumping and space filling by pegmatite-melt. Based on field observations, the role of chemistry and rheology of hosting-rocks on pegmatite differentiation and on their morphology seem to play a crucial role. In particular, high permeability zone could be a key parameter on the ascent of these extremely low viscosity melts. Preliminary numerical models have been performed with Comsol Multiphysics™ by coupling Darcy's law and heat equation.

5.1 Model geometry and key parameters

The “exotic” physical properties such as large viscosity contrasts due to temperature- and water content-dependence have been accounted for in a model dedicated to the ascent of a pegmatite-forming melt through a permeable medium. After Thomas and Davidson (2012), the water-content in pegmatitic melt may reach 18 wt % at the end of crystallisation. In these conditions, viscosity of pegmatite-forming melt is comprised between $10^{1.5}$ and $10^{3.5}$ Pa.s⁻¹ (e.g. Bartels et al. 2011). The parental magma source is assumed to impose a fixed temperature of 800°C at the base of the permeable model (0.5 x 1.5 km rectangle at a depth of – 10 km). There is no heat flow on lateral boundaries. The physico-chemical parameters of the matrix are based on micaschist properties. A fixed pressure is imposed on top boundary while no flow is allowed on others. With time (several tens of kyr), the melt becomes more and more buoyant and convective regime (thermal and velocity fields) are recorded.

5.2 Preliminary results

Our preliminary results demonstrate that the role of matrix permeability appears as much important as the high viscosity contrast between pegmatite-forming melt and hosting rocks. Uncoupled “blobs” (the “apical pools” of McKenzie 1985) are observed with low viscosity ~ 100 Pa.s⁻¹ (Figure 6) and with extremely high permeability (greater than 10^{-11} m²). These high permeability values could reflect transient mechanisms affecting the surrounding crustal rocks during partial melting of a deeper parental source.

6 Conclusion

The combination of geostatistical, geochemical and numerical tools allows discussing the common model of extreme granitic fractionation to explain the emplacement of LCT-pegmatite deposits. Here, we demonstrate by the characterisation of Li isotopes ($\delta^7\text{Li}$) that the chemical evolution of pegmatites is independent

from each other and there is no continuum between granite and LCT-pegmatites. Moreover, spatial statistical analyses indicate that the distribution of Li-rich pegmatites is clustered. This high clustering rate encountered in the three Variscan pegmatite fields is explained by the spatial relationships with surrounding tectonic features. These high permeability damage-fractured zones are considered to favour the pegmatite melt pumping and their escape from their deeper crustal magma source. Our preliminary numerical models are consistent with these previous results since they highlight the role of high permeability zone added to the role of high viscosity contrasts between pegmatites and their hosting-rocks. The process of evolved magma pumping - in high permeability damage fractured zone - provides an alternative model of direct crustal anatexis of the parental granite source. Moreover, this model justifies the high clustering rate of Variscan LCT pegmatites during their emplacement. To refine our results, additional $\delta^7\text{Li}$ values from the FLPF and BAPF are necessary to verify the inherited model of the crustal source. Finally, numerical models must be enhancing to test the role of crustal melting scale, and to determine the lifecycle of LCT-pegmatites system.

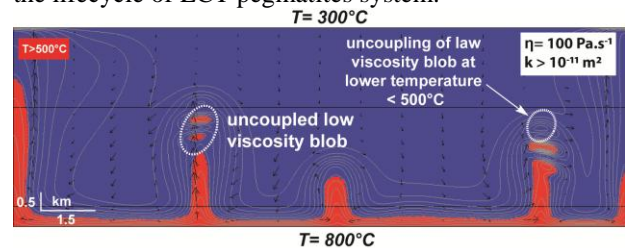


Figure 6. Example of pegmatite-forming melt escape - through high permeable medium - from a deep crustal hot source. Numerical solution at $t = 820$ years after the beginning of calculation

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