

On the origin of spodumene-rich pegmatites in the Araçuaí Pegmatite District, Minas Gerais, Brazil: insights from mica and apatite mineral chemistry

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Spodumene rich pegmatites (SRP) represent a crucial source of lithium, a key critical raw material. In Brazil, the primary lithium deposits are represented by the SRPs in the Araçuaí Pegmatite District. These pegmatites have a high modal content (up to 30%) of coarse-grained spodumene crystals dispersed within a matrix of quartz, feldspars, and muscovite and form rather homogeneous ore bodies. In the Araçuaí Pegmatite District, SRPs have been interpreted as residual magmas derived from Cambrian two-mica granites associated with large barren pegmatitic bodies formed during the late tectonic stage of the Araçuaí Orogen. However, the relation between barren and mineralized pegmatites as well as the formation mechanism of SRPs in this region and elsewhere remains puzzling.

The analysed rock samples encompass the full range of inferred differentiation grades in the Araçuaí Pegmatite District, from two-mica granites to barren pegmatitic bodies, culminating in SRP. We present compositional data of muscovite and apatite, obtained with a combination of EMP and LA-ICP-MS analyses, to investigate the genetic relation among the different lithologies as well as the internal evolution of the SRP.

Except for minor biotite in the two-mica granite, all mica can be classified as muscovite. The muscovite from the SRP has higher Al and lower Fe contents than muscovite from other lithologies. In the spodumene zone of SRP muscovite exhibits the highest contents of incompatible elements Li, Cs, Mn, and Zn, negatively correlating with the K/Rb ratio. Li median contents varies from 783 µg/g in the border zone to 2619 µg/g in the spodumene zone of SRP, while median contents of 1204 and 1156 µg/g are measured in barren pegmatites and two-mica granite.

Apatite is a ubiquitous accessory phase in the samples, reaching up to 5-10 % of modal content in the border zone of the SRP. Similarly to muscovite, apatite shows the highest contents of

incompatible elements U, Th, Mn, and Zn in the spodumene zone of SRP, along with a drastic reduction in REE contents.

Both muscovite and apatite display an extreme evolution in composition within the SRP (one to two orders of magnitude for selected trace elements), suggesting the fundamental role of internal differentiation in the pegmatite to concentrate rare elements.

The presented data support the use of muscovite and apatite chemical compositions as indicators of the degree of differentiation and Li enrichment among various products of granitic magmatism. Even if the genetic relation between the different lithologies is still not defined, it is clear that they share the same fingerprinting enrichment in rare elements, supporting an origin from the same magmatic system. The association observed between muscovite and spodumene is recurrent worldwide in SRPs and highlights the system's relatively high alumina/low fluorine character, a crucial factor favouring spodumene as the primary Li bearing crystallizing phase over Li-F bearing mica.