Characterization and Liberation Study of the Beauvoir Granite for Lithium Mica Recovery

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Abstract: A significant proportion of Europe’s lithium endowment is hosted by unconventional lithium resources such as rare-metal granites (RMG) of which the Beauvoir granite in France is a prime example. In such hard-rock deposits, where lithium is mostly hosted in micas (lepidolite, zinnwaldite), the ability to assess whether lithium can be extracted economically from the ore is essential and requires a comprehensive understanding of mineralogical properties and lithium deportment. Using three exploratory drill cores distributed along the North–South axis, a preliminary geometallurgical assessment of the granite has been conducted based on a combination of techniques including Inductively Coupled Plasma Atomic Emission Spectroscopy (ICP-AES), Atomic Adsorption Spectroscopy (AAS), Electron Probe Microanalysis (EPMA), Scanning Electron Microscope (SEM) coupled with automated mineralogy software, X-Ray Diffraction (XRD), optical microscope and sieving. Lithium distribution appears to be variable, reflecting the evolution of the granite, with higher mica content in the southern area and higher Li grade towards the center of the orebody. The size of micas in the assessed sample does not vary significantly. The grindability and liberation size of micas varies in the different zones investigated, PERC S being the most difficult to grind. There is always more than 50 wt% of the micas that are liberated in the samples when crushed to 1 mm. Indirect estimation of Li content based on EPMA and SEM analysis suggests that the content of lithium inside mica crystals could vary. If this estimation is confirmed by direct Li measurement, it for sure makes the calculations of the Li deportment more challenging.

Keywords: lithium; micas; rare metals granite; liberation; grindability

1. Introduction

The development of electric mobility to reduce fossil fuel consumption in the context of energy transition, but also the increased access to new portable technologies like computers and cell phones, create a high demand for battery materials [1–3]. Due to its special characteristics, e.g., light solid density, smallest ionic radius among alkali metals and highest electrochemical potential, lithium is extensively used in Li-ion batteries with the battery sector representing 65% of the lithium consumption in 2019 [4,5]. In this context, knowing that the lithium market is difficult to predict and that current recycling rates are only about 1%, it is expected that the global lithium consumption will be multiplied by up to a factor of seven by 2030, a rise that can be sustained at least until the end of the century [6–10].

In 2021, brines from Chile and Argentina produced 32% of the global lithium metal (34.3 kt Li) while the six western Australian pegmatite operations (mainly spodumene-based) produced about 52% (55.3 kt Li) [11–13]. To regain partial sovereignty on the supply of lithium considered as a critical raw material by the European Commission,
unconventional deposit types like rare-metal granites (RMGs) are now considered as potential resources for lithium extraction [14–20]. RMGs are hard rocks formed by the crystallization of highly fractionated magmas enriched in volatile elements and are known to potentially contain other metals like niobium, tantalum, beryllium, tin, or others [21–23].

RMGs have always been considered of poor economic value for metal extraction due to their low metal grades but, when altered, they are often exploited for their industrial minerals like kaolin or feldspars for the ceramics industry [24]. There are known examples of the successful production of lithium and other metals like tin (Sn), tantalum (Ta) or niobium (Nb) as a by-product of the exploitation of kaolinized RMG [25]. The kaolinized Yichun Li-mica granite in China has been mined for lithium and tantalum and the St Austell granite in the UK has been investigated for lithium recovery, as well as tin, niobium and rare earth element (REE) recovery [26–30]. The kaolinized Beauvoir granite is currently being exploited for kaolin production. Li-micas in residues from the kaolin process are currently recovered and stockpiled while tin in cassiterite (800 g/t Sn) and tantalum/niobium in columbite (190 g/t Ta and 120 g/t Nb) are currently recovered as by-products of kaolin production [31].

Most of the existing research and available information on lithium mineral processing is limited to spodumene, zinnwaldite and petalite as primary Li-bearing minerals [2]. Therefore, the study of lepidolite as a potential resource is of special relevance. In this contribution, a characterization study of lithium micas at the Beauvoir RMG is presented. The aim is to discuss the analytical challenge raised by lithium detection by using advanced analytical tools to understand the distribution of lithium in the orebody and study the Li-bearing mica characteristics. This paper brings insights into the mineralogy of lithium at the Beauvoir RMG through a multi-scale approach, from the macro (deposit) scale to the micro (mineral) scale. The results lay the foundation for a predictive geometallurgical model and the characterization data are used to discuss the valorization of lithium from the Beauvoir granite.

2. Background
2.1. Geological Setting and Granite Description

The Beauvoir RMG is a granitic intrusion in the NW French Massif Central. It formed at the last stage of the Variscan orogeny from the crystallization of acidic magmas [32–37]. This Li-bearing peraluminous intrusion of the high-phosphorous category outcrops at the contact of the Colettes granite and is surrounded by two-mica schists (Figure 1) [38]. Around 95% of this 310 Ma intrusion is composed of albite, quartz, K-feldspar and micas, plus other minor phases such as topaz, apatite, and zircon [39–41]. Hydrothermal alterations contribute to the formation of other mineral phases, mainly kaolinite, illite and some more muscovite and quartz [42]. According to a recent assessment of lithium resources in Europe, the Beauvoir granite has the highest lithium potential among French RMGs with 320,000 tons of Li₂O in total and 106 tons of Li at 1.3 wt% Li₂O in the upper 300 m [32,43]. Lithium is hosted mainly in micas like lepidolite and Li-muscovite, and to a lesser extent in amblygonite. Lepidolite has been extensively described in the literature as a mineral of potential economic interest for lithium extraction from hard-rock deposits [44–48].
2.2. Distribution of Lithium in the Granite

The Beauvoir granite is located below the current kaolin quarry and can be subdivided into three units from top to bottom, namely B1, B2, and B3 (Figure 2) [49–52]. This division corresponds to consecutive injections of magmatic pulses at different levels of the magmatic chamber [51]. These units coincide with a geochemical zonation, the lithium grade increasing from the bottom to the top of the granite with 2400 ppm Li at a 800 m depth to 7000 ppm Li at a 250 m depth until reaching a composition close to that of pure lepidolite, whereas iron follows an opposite trend with three clear zones corresponding to B1, B2 and B3 units [52,53]. The enrichment in lithium in felsic rocks is a direct consequence of the enrichment of residual melts at the extreme fractional crystallization stage due to the incompatibility of Li in the crystal structure of almost all minerals which crystalize from this kind of melt (quartz, albite and muscovite) [54,55]. In the case of the Beauvoir granite, last-stage mixing with meteoric water is thought to also have played a role in lithium concentration [56–58]. Therefore, it explains why the B1 unit, the closer to the surface and the last one to crystallize, is the most promising part of the deposit for lithium prospection. In terms of lithium host minerals, in addition to micas such as muscovite (KAl$_2$(Si$_3$Al)O$_{10}$(OH,F)$_2$) and lepidolite (K(Li,Al)$_3$(Si,Al)$_4$O$_{10}$(F,OH)$_2$), authors also reported the presence of lithium phosphates such as amblygonite ((Li,Na)AlPO$_4$(F,OH)) in the deposit [32]. However, neither the relative proportion of these minerals nor the corresponding lithium deportment and the variability of the later within the deposit have been investigated.
2.3. Distribution of Lithium in Micas

Mica of the Beauvoir granite has been described in detail by three published articles [52,59,60]. The authors underline differences at two different scales. Firstly, different Li-bearing micas exist. Indeed, the $\text{SiO}_2/\text{Al}_2\text{O}_3$ ratio versus the $\text{Fe}_2\text{O}_3$ diagram suggests that the micas encountered in the granite are from a continuous series between muscovite, lepidolite and zinnwaldite. Secondly, the authors showed that it is possible to find, within a single crystal, different generations of micas with theoretically different Li and Fe content reflecting the global magmatic evolution of the granite. Figure 3 shows the replacement of an early Fe-muscovite (MI) by a non-ferrous lepidolite (L), this last transformed at its borders into a late-stage muscovite (MII). These three zones observed in mica crystals first identified by Kosakevitch have been described by Monier and co-workers as follows (Li-content of each zone is given in Table 1) [52,59]:

- MI (named Zone 2 in the rest of the paper, see Section 3.2) is found at the core of the crystal and has a composition close to that of zinnwaldite. It was among the first minerals to crystalize in the melt when only a small part of quartz and feldspar already crystallized.
- L (named Zone 3′ in the rest of the paper, see Section 3.2) is in crystallographic continuity with MI. It represents the outer part of mica crystals, and its composition evolves from the one of zinnwaldite to the one of pure lepidolite. It corresponds to the conditions at which the granite crystallized, and it is the predominant stage in the B1 facies which is the least ferriferous. Often, recrystallization of this phase can be observed and could be classified as a subdivision of the L-type micas.
- MII is an early alteration phase observed in all granite that is in crystallographic continuity with L (Li-muscovite).
Figure 3. Thin section and drawing of Li-micas from Beauvoir granite with a central zone of zinnwaldite (Muscovite I) in a crystal of lepidolite with an external border of secondary Li-muscovite (Muscovite II), modified from [59]. Zones numbers in italic refers to the division made in Table 1.

Considering the theoretical structural formulas of these phases, MI is the poorest in lithium (Li$_2$O < 2%), L is the richest (Li$_2$O > 4%) and MII (2% < Li$_2$O < 4%) is intermediary. The reason for the variations in the nature of the micas is not clearly defined and could have either a metasomatic or magmatic origin [61]. To summarize, Li in the Beauvoir granite is contained in minerals from the phosphates and the mica family.

Table 1. Classification of the Beauvoir Li-bearing micas zonation based on SiO$_2$/Al$_2$O$_3$ ratio defined by Monier and co-workers [52].

<table>
<thead>
<tr>
<th>Zone</th>
<th>SiO$_2$/Al$_2$O$_3$</th>
<th>Characteristics</th>
<th>Li Content $^b$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>1.6</td>
<td>Close to Li-free muscovite</td>
<td>Low (&lt;2%Li$_2$O)</td>
</tr>
<tr>
<td>2</td>
<td>1.8</td>
<td>Intermediary from zinnwaldite to lepidolite</td>
<td>Intermediary (between 2 and 4%Li$_2$O)</td>
</tr>
<tr>
<td>3–3’</td>
<td>2.4–2.2</td>
<td>Close to pure lepidolite</td>
<td>High (&gt;4%Li$_2$O)</td>
</tr>
</tbody>
</table>

$^a$ Calculated after [52]. $^b$ Li$_2$O grades from [62].

2.4. Li-Mica Flotation

The article published by Korbel and co-workers compiles the available literature about Li-mica flotation [62]. As for the flotation of muscovite, the main collectors used to float lepidolite and zinnwaldite are cationic, from the amine family. These collectors provides good flotation of lepidolite over a wide range of pH from 2 to 11 [63]. The use of ether-amine-based collector may be an interesting alternative to primary amine because it requires lower dosages [64]. The main challenge to overcome when floating Li-micas from other silicates like quartz and albite is the lack of selectivity due to their very close surface properties [62]. However, the articles already demonstrated the feasibility of this separation on the Goncalo pegmatite ore from Portugal with ether-amine and primary fatty ammonium acetate salt (Aeroamine 3000C), at pH 3, or on the Yichun granite ore from China with an amine-based Gemini collector [64–66].
3. Methodology

3.1. The Challenge of Lithium Determination in Minerals

This study aims at characterizing the variability of lithium both vertically and laterally inside the B1 unit itself. However, lithium is a very light element, which makes the quantitative detection by non-destructive techniques a complicated and sometimes impossible task. Typically, all equipment that relies on X-ray (equipped with Electron Dispersive Spectrometers, EDS, or Wavelength Dispersive Spectrometer, WDS detectors) like X-ray Fluorescence (XRF), Scanning Electron Microscopy (SEM), Electron Probe Microanalysis (EPMA) and automated mineralogy systems like Mineral Liberation Analysis (MLA) and Quantitative Evaluation of Materials by Scanning Electron Microscopy (QEMSCAN) cannot be used to detect lithium. A practical way to deal with this issue is to find a correlation between lithium and another element that is easier to detect. In the case of the Beauvoir granite, a correlation between bulk Li$_2$O and Rb$_2$O content has been observed [59,67]. Rubidium has similar characteristics to those of lithium and can substitute it in mineral lattices which explain why their whole rock content sometimes correlates. The Inductively Coupled Plasma—Atomic Emission Spectroscopy (ICP-AES) and Atomic Absorption Spectroscopy (AAS) analyses performed all along the drill cores validate this observation as shown by the correlation observed in Figure 4. Thus, rubidium grade in rock samples appears as a good proxy to estimate lithium grade. However, previous studies have shown that this correlation is not always observed in mica concentrates [59]. It is especially true in the case of the Beauvoir granite in which lithium is hosted both in micas and amblygonite. It means that the lithium content has to be measured using direct detection tools such as AAS and ICP or other nondestructive spectrometric tools such as Laser Induced Breakdown Spectroscopy (LIBS) which is still under development [68].

![Graph showing Li$_2$O/Rb$_2$O correlation in samples along the drill cores. Rb measured by ICP-AES and Li by AAS.](image)

**Figure 4.** Bulk Li$_2$O/Rb$_2$O correlation in samples along the drill cores. Rb measured by ICP-AES and Li by AAS.

3.2. Discriminate Different Types of Micas with EPMA Analysis

Different attempts to find proxy elements to correlate Li in micas have been performed previously. Tischendorf and co-workers used various Li-mica analysis to determine useful correlations that reduce time and cost [69]. However, the problem of different generations of micas present in a same sample was identified. For the particular case of Beauvoir RMG, a work in separated micas from the three identified facies was performed [52]. By petrographic analysis, four different zones in mica crystals were identified: Zone 1,
Zone 2 (corresponding to MI on Figure 3), Zone 3 and Zone 3’ (corresponding to L on Figure 3). One meaningful conclusion is that B1 analyzed micas are all of the “Zone 3” category. The author also suggested that the Si/Al ratio in the micas is directly linked to their Li content. In Zone 1, the ratios are lower and so is the Li content, whereas for Zone 3, Si/Al ratios and Li are higher. Zone 2 has an intermediate composition. This elemental relation was used in recent work to compare elemental trends in lithium micas collected in Australia, finding that Li content is directly related with Si content [68,70]. Other authors have also used EPMA for the analysis of crystal chemistry of di-octahedral (Li-muscovite) and tri-octahedral (trilithionite–polyliithionite, i.e., lepidolite series) micas [69–72]. They found that the replacement of Li by Al in the octahedral sites induces a replacement of Si for Al in the tetrahedral sites of the micas, and that the mechanisms of these substitutions vary between mica types.

Based on these observations, we decided to calculate the SiO$_2$/Al$_2$O$_3$ ratio value of Zones 1, 2, 3 and 3’ to qualitatively compare theoretical micas families and estimate their Li contents in a SiO$_2$ vs. Al$_2$O$_3$ diagram. Values of Monier and co-workers were used [52].

3.3. Definition of Mineral Phases in MLA Analysis

MLA is a technique based on a scanning electron microscopy, where BSE images and X-ray analysis are treated to obtain mineralogical composition of each analyzed particle or grain of a sample [73]. Since the SEM itself does not allow the acquisition of data for light elements, the MLA cannot identify Li-bearing minerals (or Be-bearing). To treat the data obtained by this technique in a meaningful geometallurgical characterization, the identified mineral phases were grouped according to their relevance or performance in mineral processing. The grouping can be found in Table 2 and is the base for further analysis and produced graphs.

Table 2. Minerals grouping used for MLA analysis.

<table>
<thead>
<tr>
<th>Group Name</th>
<th>Minerals (Abbreviation)</th>
<th>Formula</th>
</tr>
</thead>
<tbody>
<tr>
<td>Quartz</td>
<td>Quartz (Qtz)</td>
<td>SiO$_2$</td>
</tr>
<tr>
<td>K-feldspar</td>
<td>K-feldspar—(Kfs)</td>
<td>KAlSi$_3$O$_8$</td>
</tr>
<tr>
<td>Albite</td>
<td>Albite (Ab)</td>
<td>NaAlSi$_3$O$_8$</td>
</tr>
<tr>
<td>Cassiterite</td>
<td>Cassiterite (Cst)</td>
<td>SnO$_2$</td>
</tr>
<tr>
<td>Muscovite</td>
<td>Muscovite (Ms)</td>
<td>KAl$_2$(AlSi$_3$O$_10$)(OH,F)$_2$</td>
</tr>
<tr>
<td>Kaolinite</td>
<td>Kaolinite (Kln)</td>
<td>Al$_2$Si$_3$O$_5$(OH)$_4$</td>
</tr>
<tr>
<td>Talc</td>
<td>Talc (Tlc)</td>
<td>Mg$_5$Si$_4$O$_10$(OH)$_2$</td>
</tr>
<tr>
<td>Biotite</td>
<td>Biotite (Bt)</td>
<td>K(Mg,Fe)$_2$(OH,F)$_2$(Si$_3$AI$_1$O$_10$)</td>
</tr>
<tr>
<td>Columbite</td>
<td>Mn-Columbite (Mn-Col)</td>
<td>(Mn$^{2+}$,Fe$^{2+}$)Nb$_2$O$_6$</td>
</tr>
<tr>
<td>Ixolite</td>
<td>Ixolite (IxI)</td>
<td>(Ta,Nb,Sn,Mn$^{2+}$,Fe$^{2+}$)O$_2$</td>
</tr>
<tr>
<td>Microlite</td>
<td>Microlite (Mlt)</td>
<td>(Na,Ca)$_2$Ta$_2$O$_6$(O,OH,F)</td>
</tr>
<tr>
<td>Apatite</td>
<td>Apatite (Ap)</td>
<td>Ca$_5$(PO$_4$)$_3$(F,Cl,OH)</td>
</tr>
<tr>
<td>Other</td>
<td>CaAl$_2$, NaCaAl$_2$, AlSrCa- or CaNaAlF-phosphate</td>
<td></td>
</tr>
<tr>
<td>phosphates</td>
<td>Topaz, zircon, cryptomelane, gorceixite, plumbogummite, fluorite, steel, unknown</td>
<td></td>
</tr>
<tr>
<td>Others</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

The term muscovite refers to micas in general, except biotite, which has its own classification due to the absence of Li in its composition. It should be noted that Be-bearing minerals in Beauvoir are still a matter of discussion. However, since some phosphates have been identified previously in relation with the granite, we assigned this element to phosphates [32].

MLA results were processed in the Dataview software, version 3.1.4. The classification of muscovite particles according to their percentage of free surface was performed by choosing four arbitrary thresholds, i.e., 20, 50, 80 and 90 wt% of free surface. Particles
between 0 and 20 wt% of free surface were classified as “locked”, particles between 20 and 50 wt% as “sub-middlings”, particles between 50 and 80 wt% as “middlings”, particles between 80 and 90 wt% as “liberated” and particles with more than 90 wt% of free surface as “free”. The selected size fractions for the filtering of the results were based on equivalent circle diameter (ECD) and are intended to match the elemental size by size analysis that is presented later in this paper.

4. Materials and Methods

4.1. Material

The materials used in this study come from three exploratory drill holes drilled in the actual kaolin quarry in 2018. They are the first samples of the Beauvoir granite taken since the French continental deep drilling program (GPF 1) in 1981 [59]. The three drill holes of approximately 150 m in length each, PERC-North (Exclusive Exploration Permit Core—North), PERC-Central and PERC-South (designed as PERC-N, C and S hereafter), are oriented to cross the altered (kaolinized) zone and reach the fresh underlaying granite. Since they were taken in the first 150 m, the samples correspond to the upper petrographic unit of the granite (called B1). The literature indicates that this unit is composed of fine albite laths (35%); anhedral and interstitial quartz with globular crystals (30%); abundant and colorless lepidolite (22%) with composition between polythionite and trilithionite with less than 5% zinnwaldite and anhedral and scarce K-feldspars (9%) [32,39,51].

Two samples were selected from each drill cores at an upper and a lower position (see cross-section of Figure 2): south upper at a 62 m depth (S62), south lower at a 102 m depth (S102), center upper at a 67 m depth (C67), center lower at a 108 m depth (C108), north upper at a 69 m depth (N69), north lower at a 118 m depth (N118). From the six samples, polished thin sections were prepared for petrographic and mineralogical analysis (details given in Section 4.2.2).

After the taking of the six samples for polished thin sections described above, the three drill cores were split in half by diamond saw, and one half was jaw-crushed to below 4 cm, homogenized and split by coning and quartering in order to obtain a test sample for each location. An analytical subsample was taken for ICP-AES analysis. Representative aliquots of 10 kg were crushed in another jaw crusher to reduce size to below 1 cm, homogenized and sampled before being fed into a roll crusher until a 100% passing 2 mm for liberation studies. Grinding was performed during 20 min in a laboratory stainless steel rod mill of 25 cm long by 15 cm in diameter filled with twelve stainless steel rods of 24 cm long by 1.9 cm diameter weighting 530 g each. The mill was run at 70 rotations per minute with a 60 wt% solid in batch configuration with 1 kg of ore.

4.2. Analytical Methods

4.2.1. Chemical Analyses

All chemical analyses were conducted at the GeoRessources laboratory (Nancy, France). The bulk chemical composition of the composite and metallurgical samples was determined with ICP-AES using an ICAP 7000 Series (Thermo Scientific, Waltham, MA, USA). Samples were subjected to an alkali fusion with lithium metaborate before dissolved by acidic attack and introduced in the device nebulizer. The lithium content of the samples, which cannot be measured accurately with the ICP-AES since the introduction of lithium metaborate artificially increases the lithium content of the sample, was quantified by AAS with a Perkin Elmer A800 (Perkin Elmer, Waltham, MA, USA) after it was solubilized as a cation.

4.2.2. Mineralogical Analyses

Particle size distribution analysis of crushed samples was carried out in a Rotap® sieve shaker, in dry mode for fractions above 100 μm and in wet mode below.

Qualitative mineralogical analysis of the major phases was carried out by XRD with a Bruker’s D2 Advance (Bruker, Billerica, MA, USA ), operating with a copper Ka ray
(1.789 Å), a tension of 35 kV and an intensity of 45 mA. Major peaks were interpreted through the ICDD database.

Mineral identification and description from polished and thin sections were performed with a Nikon OptiPhot-2 (Nikon, Shinjuku, Japan), in both polarized and cross-polarized light. Mineralogical and textural interpretations were performed based on the work of Rainbault and co-workers [39].

A Hitachi S4800 SEM (Hitachi Ltd., Tokyo, Japan) was used to make textural observations, mineral identifications and perform local X-ray microanalyses. It was equipped with an Energy Dispersive Spectrometer (EDS) using a Si (Li) semiconductor detector to perform semi-quantitative analysis. Imaging voltage was fixed at 15 kV.

Mineral liberation analyses were conducted on 3–5 g aliquots of the six samples (see Section 4.1) ground to −1 mm at the Helmholtz Institute Freiberg for Resource Technology (HZDR, Dresden, Germany). Each aliquot was mixed with graphite and epoxy resin to produce polished sections. To avoid any vertical segregation due to the difference in settling velocity of particles of different sizes, the polished sections were sliced and mounted again in another resin. They were carbon coated with a Leica (Baltec, Pfäffikon, Switzerland) MED 020 vacuum evaporator before being sent to MLA analysis. The MLA pairs a FEI Quanta 600F (FEI, Hillsboro, WA, USA) field emission SEM and two Bruker Quantax X-Flash 5010 EDX detectors. FEI’s MLA suite 3.1.4 (Bruker, Billerica, MA, USA) was used for data acquisition and processing. Mineral quantification was estimated using mineral surface areas and densities.

Additionally, a CAMECA SX100 (Cameca, Gennevières, France) electron probe microanalyzer (EPMA) was used to perform in situ elemental analysis of minerals, with column conditions fixed at 15 kV and 20 nA, and a detection limit of 0.1 wt% on oxides.

5. Results

5.1. Characterization Study

Mica-oriented characterization study was carried out on three bulk samples of the three drills and the six georeferenced samples shown on the cross-section of Figure 2. Firstly, combined ICP-AES/XRD characterization was performed on bulk samples of the three drills. Then, more in-depth techniques (EMPA, SEM and MLA) were used to discern mineralogical differences inside the granite on the six georeferenced samples.

5.1.1. Elemental Bulk Composition

Despite our work not being focused on metals other than lithium, determining the bulk geochemical differences on the three drill cores analyzed is a good starting point. The ICP-AES results of selected bulk samples for some major oxides and other metals are presented in Table 3. Li grade measured by AAS follows a trend of Center > South > North, whereas for the other metals (with exception of Sn), a trend of South > Center > North is observed.

5.1.2. X-ray Diffraction

The XRD diffraction patterns of the three different analyzed cores are presented in Figure 5. Major gangue minerals identified are feldspar (mainly albite), quartz and, to a lesser extent, kaolinite. No accessory minerals susceptible of hosting tin, tantalum or niobium was identified possibly due to their low abundance in the sample as the typical limit of detection for mineral identification XRD is around 1–3 wt%. The difference in the intensity and broadening of the peaks of lithium-bearing micas like muscovite, zinnwaldite and lepidolite is due to cation substitutions which make a slight difference in the interplanar distance in the crystal. This difference is so small that XRD patterns do not allow the differentiation of these different lithium micas in the three composite samples [70].
Table 3. Bulk elemental composition of the PERC N, C and S drill holes by ICP-AES and AAS (for Li). MA-N is a standard from the international working group provided by the Centre de recherche pétrographique et géochimique (Vandoeuvre-les-nancy, France). WV = working value. DL = detection limit.

<table>
<thead>
<tr>
<th>Samples</th>
<th>MA-N</th>
<th>MEASURED</th>
<th>WV</th>
</tr>
</thead>
<tbody>
<tr>
<td>PERC N</td>
<td>PERC C</td>
<td>PERC S</td>
<td></td>
</tr>
<tr>
<td>SiO₂ (wt%)</td>
<td>68.82</td>
<td>68.06</td>
<td>67.99</td>
</tr>
<tr>
<td>Al₂O₃</td>
<td>16.30</td>
<td>17.24</td>
<td>17.54</td>
</tr>
<tr>
<td>Na₂O</td>
<td>3.35</td>
<td>4.37</td>
<td>4.98</td>
</tr>
<tr>
<td>K₂O</td>
<td>3.83</td>
<td>3.46</td>
<td>3.14</td>
</tr>
<tr>
<td>CaO</td>
<td>0.88</td>
<td>0.59</td>
<td>0.52</td>
</tr>
<tr>
<td>P₂O₅</td>
<td>0.74</td>
<td>0.95</td>
<td>0.67</td>
</tr>
<tr>
<td>Fe₂O₃ tot</td>
<td>0.37</td>
<td>0.32</td>
<td>0.42</td>
</tr>
<tr>
<td>MgO</td>
<td>0.07</td>
<td>0.06</td>
<td>0.04</td>
</tr>
<tr>
<td>MnO</td>
<td>0.14</td>
<td>0.04</td>
<td>0.05</td>
</tr>
<tr>
<td>Li₂O (wt%)</td>
<td>0.71</td>
<td>0.89</td>
<td>0.86</td>
</tr>
<tr>
<td>Sn (ppm)</td>
<td>863</td>
<td>1073</td>
<td>781</td>
</tr>
<tr>
<td>Ta</td>
<td>150</td>
<td>155</td>
<td>270</td>
</tr>
<tr>
<td>Ti</td>
<td>101</td>
<td>87</td>
<td>72</td>
</tr>
<tr>
<td>Nb</td>
<td>90</td>
<td>106</td>
<td>167</td>
</tr>
<tr>
<td>Be</td>
<td>67</td>
<td>115</td>
<td>206</td>
</tr>
<tr>
<td>W</td>
<td>46</td>
<td>51</td>
<td>60</td>
</tr>
<tr>
<td>Total (wt%)</td>
<td>96.42</td>
<td>96.60</td>
<td>97.77</td>
</tr>
</tbody>
</table>

Figure 5. X-ray diffractograms of the bulk sample from PERC N, C and S drill holes.

5.1.3. Modal Mineralogy by MLA

The modal mineralogy of the six samples calculated by MLA and shown in Figure 6 are in agreement with the global mineralogy of the Beauvoir granite as described in the literature. Indeed, the granite is mainly composed of four mineral phases in order of
importance: albite, muscovite, quartz and K-feldspar, making up at least 90 wt% of all samples. Head proportions of those major minerals do not change drastically from north to south and vertically for center and south. However, the north upper sample has a greater K-feldspar content (20 wt% against 3–1 wt%). Generally, K-feldspar content increases from south to north at the expense of albite. Its content passes from around 3 wt% in the south lower sample to 20 wt% in the north upper one. Muscovite is always more abundant in the coarser size fractions and accounts for more than 60 wt% in the fraction > 1 mm of both south samples. Accessory mineral phases include kaolinite, apatite, biotite, cassiterite and other minor phases. There is a clear difference in the content of kaolinite between upper and lower levels, marking the most intense kaolinization close to the surface, especially in the case of north and center positions where kaolinite represents almost 5 wt% of the sample and 10% in the <36 µm. Accessory minerals such as cassiterite, microlite or apatite are more abundant in the upper part of the deposit. Peculiar mineral phases classified in the “others” group in Table 2 such as topaz, fluorite or zircon can overpass 5 wt% in the central zone of the deposit.

Figure 6. Head and size-by-size modal mineralogy of the upper and lower samples from the PERC N, C and S drill holes obtained by MLA filtering.

5.1.4. Characteristics of Micas Obtained by MLA

An illustration of individual particle classification by size fraction filtered from the MLA is displayed in Figure 7 for the south upper sample. It shows that a great proportion of muscovite is liberated especially in the case of the smallest grains. In the coarser grains, muscovite is still found in association with other mineral phases, even if some grains are already liberated. Non-liberated micas sometimes display very complex associations with multiple inclusions of main gangue minerals like quartz and albite and other accessory minerals such as apatite as shown in the zoom of Figure 7. It is important to keep in mind that samples were ground to 100% passing 1 mm prior to MLA analysis; therefore, the observed liberation size corresponds to the metallurgical liberation and not the geological liberation.
minerals such as apatite as shown in the zoom of Figure 7. It is important to keep in mind that samples were ground to 100% passing 1 mm prior to MLA analysis; therefore, the observed liberation size corresponds to the metallurgical liberation and not the geological liberation.

**Figure 7.** Illustration of the grain classification results obtained by MLA for the south upper sample.

MLA revealed that the mean \(D_{80}\) (the size below which 80% of the total mica population belongs to) of the micas is 260 µm, but a variability was observed between the samples. The \(D_{80}\) of muscovite in the three upper composite samples (Figure 8) varied from 190 µm for north to 320 µm for central samples. In the case of the three lower samples, values were almost similar and found to be between 278 and 320 µm. In the upper section, the north area is the one displaying coarser micas, whereas it is the one with finer micas in the lower section. Moreover, the size of the micas decreased from upper to lower section in the northern area, whereas it increased in the center and did not change in the south.

**Figure 8.** Muscovite size distribution in the three upper (left) and lower (right) composite samples of the PERC N, C and S drill holes obtained by MLA.

Figure 9 displays the major gangue phases associated with muscovite, which globally reflect the modal mineralogy of Figure 6. The percentage of free muscovites in the ground ore increases from 45 wt% in north upper to 60–65 wt% in north center and south. This
is in accordance with the results from Figure 8, which indicates an increase in muscovite size from north to south upper sample. Regardless of the position in the deposit, the major gangue minerals in association with muscovite are albite, quartz and K-feldspar. K-feldspar seems less associated with muscovite compared with quartz because sometimes it represents just a few percent of the total ore, but in reality, muscovite and K-feldspar are often associated. For instance, the K-feldspar content in the south upper sample is less than 3 wt% (see Figure 6) but represents 12% of muscovite associations, whereas quartz content is 18 wt% but represents 5% of muscovite associations. MLA results have shown that among the metal-bearing accessory minerals, cassiterite is the one most associated with muscovite, especially in the southern area of the deposit where up to 80% of cassiterite is associated with muscovite in the fraction above 710 $\mu$m. Nb-Ta minerals are also associated with muscovite. This confirms optical microscopic observations which have shown inclusions of Nb-Ta minerals and cassiterite inside the micas. In addition, it seems that the percentage of association with minerals containing Nb, Ta, Sn and Be increases in the southern part of the deposit.

![Figure 8](image_url)  
**Figure 8.** Muscovite size distribution in the three upper (left) and lower (right) composite samples of the PERC N, C and S drill holes obtained by MLA.

![Figure 9](image_url)  
**Figure 9.** Muscovite mineral associations with major gangue minerals per size fraction in the upper and lower samples from the PERC N, C and S drill holes, obtained by MLA filtering.

The liberation degree of muscovites of the six samples is shown in Figure 10. For all samples, muscovite is found to be mostly “free”, meaning that it has more than 90% of free surface. Proportions of free particles increase from upper to lower positions for both north and south positions, respectively, by 24 and 10 wt% (see dashed lines on the PERC N plot), whereas they vary by only 2% in the case of center samples. If this difference can easily be linked to the size reduction in muscovites in the upper north part of the deposit, it is less easy to explain it for south and center samples.
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easily be linked to the size reduction in muscovites in the upper north part of the deposit, it is less easy to explain it for south and center samples.

Figure 10. Muscovite mineral liberation in the upper and lower samples from the north, center and south drill holes obtained by MLA.

After grinding to only 1 mm, the percentage of muscovites liberated in the samples is at least 60% in the three lower samples. This suggests a good liberation of muscovites of the Beauvoir ore since the early size reduction steps. This is already suggested by Figure 8, where about 50 wt% of muscovites of south samples above 1 mm are free, and by the observation at the binocular of the fraction 355–500 µm of a laboratory sample, where 44% of micas are free.

5.1.5. SEM Images and Electron Probe Micro-Analyses

Figure 11a,b display micas showing great variations in grey-level values in their BSE images. Some zones with a lighter grey level, parallel to the cleavage plans, appear to have a higher average atomic number than the rest of the mineral. On central lower sample images (Figure 11b), this change in grey level is also observed, but it is more diffused and progressive in the crystals. For both locations, these respective variations are observed repeatedly on BSE images of the micas, implying that they are characteristics of the micas at these given locations.
5.2. Mineral Liberation Study

After Stage III, almost all of the products are below 2 mm in size; the P80 of PERC C, generalized to the entire particle size distribution curves after grinding with a roll crushe which itself yielded more fines than PERC S (Figure 13). The same trend is observed and the zones correspond to the classification defined by Aubert and co-workers [62].

Figure 11. BSE images of micas with SiO₂/Al₂O₃ ratio values from the north lower sample (a), from the center lower sample (b). Letters are reported in Figure 12.

Figure 12. SiO₂ vs. Al₂O₃ diagram of in situ EPMA white mica analysis of north lower and center lower samples. The zones correspond to the classification defined by Aubert and co-workers [62].

In the north lower sample (Figure 11a), two different ranges of Si/Al ratio are present in the same crystal corresponding to the two zones identified on the BSE images. The light grey areas have a higher Si/Al ratio than the rest of the crystal (points a, c and e). In the central lower sample (Figure 11b), the two points (g in a dark gray zone and h in a light gray zone) acquired display a high Si/Al ratio value in the same order of magnitude of the light grey zones of Figure 11a. This can be due to thickness of the grain or the association with the underneath mineral.

The optical microscope images are placed aside as a reference of the variation in interference colors in the zones analyzed by EPMA. In Figure 11a, it is shown that the
second-order polarization colors of zones which are closer to the lepidolite vary from yellow to orange, whereas they are blue/purple in the zones corresponding to Li-muscovite.

All the EPMA spot analyzes performed on the north lower and central lower composite samples were plotted in SiO$_2$ vs. Al$_2$O$_3$ diagrams (Figure 12). The scattering of points in this two-dimensional space reflects the previous observations: micas of north lower samples form two different clusters due to the presence of high and low Si/Al ratio in their structure, whereas micas from central lower samples form just one cluster because they are more homogenous. If we assume that the value of this ratio can be linked to the lithium content of micas (see Section 3.2), the higher this ratio, the higher the lithium content should be. Therefore, micas from north lower samples display low and high Li zones in their structure, corresponding to crystals close to Li-muscovite and lepidolite, respectively, whereas it is not the case with micas from the central lower zone, all of which have a high lithium grade. This observation is in accordance with the bulk analysis of the three cores (Table 3) where the lithium grade of PERC C is the higher compared to the two other locations.

5.2. Mineral Liberation Study

5.2.1. Grindability and Relation with Mica Content

After the two first stages of crushing, a slight difference in the particle size distributions appears below 2 mm, with more fine particles produced with PERC C than PERC N which itself yielded more fines than PERC S (Figure 13). The same trend is observed and generalized to the entire particle size distribution curves after grinding with a roll crusher (III). After Stage III, almost all of the products are below 2 mm in size; the $P_{90}$ of PERC C, N, and S was found to be 0.75, 1, and 1.5 mm, respectively. Thus, there is a noticeable variability in the ore comminution behavior with respect to the spatial distribution in the granite. It is interesting to notice that this difference is related to the muscovite content and size. Indeed, first, the south zone is the richest in muscovite, whereas the central zone is the poorest, with 25 and 17 wt%, respectively (Figure 5). Second, the south core is particularly rich in coarse micas compared to the other zones; it accounts for 65 wt% of the fraction above 1 mm. It is known that mica is one of the most difficult minerals to grind due to their platy shape and relative elasticity [74].

Figure 13. Cumulative particle size distribution of PERC N, C and S after three comminution stages.
5.2.2. Lithium Distribution

The evolution of lithium grade per size fraction after the roll crusher indicates that lithium grade follows the trend of PERC C > PERC S > PERC N, and this difference is accentuated in the coarser size fractions (Figure 14). For the three cores, the Li grade is maximum and rather constant above 355 µm with values reaching almost 4000 ppm for PERC N, 4500 ppm for PERC S and 5000 ppm for PERC C. Below 355 µm, the Li grade per size fraction decreases continuously until about 2000 ppm in the <36 µm fraction. The PERC C sample displays the higher Li grade but has a different distribution of the element in the different size fractions. For example, in PERC N and S, respectively, 33% and 42% of lithium reports to the 1–2 mm size fraction, whereas it is only 24% for PERC C. This is directly linked to the difference in grindability of the three samples observed in Figure 13. Indeed, the PERC S sample is the most difficult to grind, whereas PERC C is the easiest one.

Figure 14. Lithium grade (AAS measurement) and weight distribution per size fractions within PERC N, C and S after crushing (Stage III).

6. Discussion

6.1. Geology, Mineralogy and Lithium Content

Previous work based on samples coming from drill holes of the upper part of the Beauvoir granite (i.e., essentially from the B1 facies) suggested that the southern area of the deposit is more homogenous and richer in lithium [59]. This study confirms that the ore has less variability in the southern part compared to the north part, which has undergone more intense kaolinization and which creates a sharper vertical and lateral contrast. Also, the lithium content of granite is higher in the southern part of the deposit compared to the north, but the central part, represented by PERC C in this study, displays even higher lithium values. This highlights that the lithium mineralization seems to be unevenly spatially distributed. Thus, further investigations into the variability of the lithium content within the micas as well as lithium deportment in other minerals and its relationship with the host units will give more insight in this matter.

The results obtained in this study on only six samples from three exploratory drill holes do not allow to identify a clear trend in the spatial variability of the micas characteristics within the deposit. However, several authors have observed mica zonation patterns as well as variable distribution across different lithium-containing pegmatites: Karibib pegmatites, Namibia [75] Pakeagama Lake (Canada) [76], Agamela Stock (Portugal) [77] and Yichun Granite (China) [78]. Therefore, there is still an open discussion on whether or not such a trend can be identified in a rare metal granite like Beauvoir and what is the influence of magmatic processes on the distribution of Li in the deposit. PERC N displayed the lowest lithium grades; however, modal mineralogy shows the highest K-feldspar content of all the
analyzed samples. This is an important observation, as industrial minerals can still be a potential valued by-product of lithium extraction from the Beauvoir granite.

At deposit scale, following the rubidium content of the ore may be a good way to follow the spatial evolution of the lithium content as proposed by several authors [59,67]. However, at mineral scale, this correlation might lead to erroneous results due to different mica generations (Figure 11). This issue has already been raised, and the use of the mica octahedral site occupancy has been proposed instead to classify different types of micas [69].

The estimation of Li content inside mica crystals from the Si/Al ratio does not replace direct analysis of Li. Indeed, it provides information only on the occupancy of the octahedral sites of the micas, which does not necessarily mean that the Li content is different. Therefore, the variations in Si/Al ratio suggest a variation in the nature of the micas more than a variation in Li content between the different generations in the same crystal. In order to validate the last hypothesis, a direct Li analysis using LA-ICP-MS or LIBS must be carried out.

Automated mineralogy is sometimes considered as a fully integrated solution to characterize elemental and mineral composition, morphology and liberation degree of particles. This study highlights the limitation of this technique when dealing with Li-bearing minerals, especially in the case of Li-bearing micas which have variable Li content and no proxy element to provide a robust estimation. Studies with more samples are necessary in order to assess the acceptable standard deviation of the results and uncertainties of the automated mineralogy, as suggested in recent works [79].

Further characterization work in the deposit must tackle the challenge of Lithium characterization by a combination of techniques. At macro scale, µXRF may be used to describe textures and mineral associations. At micro scale, EPMA can be used to estimate Li content of micas using the Si/Al ratio. Direct in situ measurements with LA-ICP-MS promise a reliable Li grade measurement but remain a very punctual technique [70]. LIBS may be a solution to detect and quantify lithium but is still under development.

6.2. Implications for Mineral Processing

There are only a few articles dealing with the processing of rare metal granites, and deposits of this kind currently in operation are rare. The Yichun albite–topaz–lepidolite granite in China [80], currently exploited for lithium from micas, can serve as a benchmark for the development of a flowsheet for similar deposit types like Beauvoir or granites from the Dong Ram area in Vietnam [81]. The flowsheet of the Yichun granite comprises several stages to recover not only lepidolite by flotation, but also feldspar, and niobium/tantalum with spiral and shaking tables. Even after 30 years of continuous mining operation, there is not much literature providing insights on the main challenges related to the processing of this ore, and if it exists, it is of very poor quality. Nevertheless, regrinding of the above 500 µm fraction and the use of a shaking table is presented as a great opportunity to reduce Li and Nb/Ta losses [82]. The later was reduced by around 40%.

6.2.1. Comminution, Size Fractions and Liberation

The present study, even if limited to three drill cores, shows that the Beauvoir ore grindability may vary significantly, which may be directly linked to the mica content of the ore. In a grinding circuit, it is expected that hard-to-grind minerals like micas accumulate in the circulating loads [74]. In addition, the MLA results suggest that micas present a significantly coarser size distribution than cassiterite and Nb-Ta minerals, respectively, around 200–300 µm and <100 µm. Therefore, mineral liberation will be a critical factor for the performance of the future process. If grinding is not designed regarding the ore characteristics in the case of the Beauvoir granite, the attempt to reduce the size of the resistant lithium micas can lead to disadvantages such as increased energy expenditure, overgrinding of metallic by-product minerals and increased general fine production.

Based on the above-mentioned issues, it may be appropriate to propose a step-by-step comminution/flotation scheme (coarse, medium and fine flotation) because there is a real
opportunity in the case of the Beauvoir granite to recover Li-bearing micas from the early stage of grinding, 500 µm being the size at which we consider that the separation of micas that are partially liberated would be possible and worth to perform. However, this may not be appropriate for an industrial project.

The inclusions of gangue minerals in the micas will lead to difficulties for concentrate purity, but also the presence of phosphorous in apatite and other phosphates are expected to create problems for the downstream hydrometallurgical processes.

6.2.2. Flotation Design

The fact that muscovite is mainly associated with albite across the whole profile might be a problem for the selective recovery of Li-bearing micas from the gangue. As highlighted in previous studies, silicates like quartz and albite have similar point-of-zero charge (PZC) with muscovite [82,83]. In addition, the flotation of coarse (usually above 250 µm) and platy particles is challenging since it is commonly assumed that optimal particle size range for flotation is below 150 µm. This point could be addressed by using the sorting at coarse size fraction or by playing on key flotation parameters that avoid particle detachment from bubble surface like the modification of hydrodynamic regimes [64]. The recent study reported by Filippov and co-workers on the lepidolite recovery from the pegmatite ore of the Gonçalo deposit (Portugal) showed that the coarse size fractions (−2 + 0.84 mm and −0.84 + 0.24 mm) responded well to electrostatic separation resulting in a Li concentrate at >3.5% Li2O [64]. The finer size fraction (−0.240 + 0.063 mm) was processed by flotation using monoetheramine as a powerful collector at 65 g/t only to obtain a lepidolite concentrate at 4.5% Li2O.

Mixing of collectors can play an important role in the mineral separation by flotation [84]. Indeed, investigation of the effect of combined cationic and non-ionic collectors in the flotation of mica minerals has been performed [84,85]. It was shown that non-ionic reagent enhanced the flotation of biotite and muscovite by primary amine collector [84]. Moreover, the concentration of amine required to achieve a 95% recovery of biotite, and muscovite minerals decreased dramatically in the presence of a fatty-alcohol-based non-ionic reagent [84]. The mechanisms of adsorption of a mixture of cationic and non-ionic reagent onto the phyllosilicates are not well understood. However, the mechanism can be similar to the amine-alcohol co-adsorption on the Fe-bearing amphiboles, leading to a denser adsorption layer formation and thus to enhanced surface hydrophobicity [86]. Further studies have to be performed to confirm this mechanism and the possibility of the enhancement of the coarse particle flotation using the ionic–non-ionic reagent mixture.

With the use of dodecylamine (DDA) and octanol at a ratio of 2:1, an increase in recovery of muscovite over a wider range of pH compared to that in the case of the use of DDA alone (marked decrease in recovery in values of pH between 4 and 10) was determined [85]. At pH 6, the recovery of muscovite changed from 48.90% for DDA alone, whereas the collector mixture showed a value of recovery of 85.36%. Furthermore, the mixture of cationic collectors at the Yichun mine in China increased the recovery of lepidolite from 40% to 70% prior to roasting and sulfuric acid treatment [87,88]. Froth flotation with a cleaner stage and a DDA collector allow to reach recovery as high as 77.0% with a 4.7% Li2O [88].

6.2.3. The Mica Content: A Key Ore Property

This work highlighted that the feasibility of lithium exploitation depends on the particle scale, but of course the long-term tactical–strategic geometallurgical planning must play an essential role [89,90]. For this planning, the mica content appears to be a key ore property. Indeed, it is directly linked to the lithium potential of a given zone because it conditions both the Li grade and the grindability of the ore. Therefore, it is of the uppermost importance to measure the mica content in the ore along exploratory drill holes and report it in a geometallurgical model. This can be systemically performed by either XRD or MLA.
analysis on punctual core samples or, for a better estimation, on ground core samples on a given core length.

7. Conclusions

This study of the Beauvoir granite based on the three exploratory drill cores shows that the ore is mostly composed of micas, albite and quartz. Other mineral phases include K-feldspar, kaolinite, phosphates, cassiterite, columbo-tantalite and microlite. Lithium is hosted in muscovite, which belongs to a continuous series that may be classified according to their Si/Al ratio between zinnwaldites and lepidolite poles that have different lithium contents, typically 3.4 and 7.7% Li₂O, respectively. There is also a low, but not quantified, amount of lithium in phosphates, which makes the assessment of the full lithium deportment very complex. The central zone has a higher percentage lithium content (4148 ppm Li) but shows a lower content of muscovite (Li-micas). The liberation size (Dₘₙ) of micas at Beauvoir seems to be relatively variable, with values between 190 and 320 µm, but with a clear spatial trend. For all analyzed samples, it was noted that an important proportion of partially liberated to fully liberated micas (>50% muscovite surface) is present. Muscovite is mostly found in association with albite and K-feldspar, especially in the upper part of the deposit where between 25 and 40 wt% of muscovites are associated with feldspars. In the center lower and south lower samples, feldspars and quartz both account for 15 wt% of muscovite associations.

The detailed characterization of the comminution products highlights the importance of mica content in the ore. Indeed, it is linked to both lithium potential and grindability of a given zone of the deposit. The difficulty to grind micas leads to the proposal of a multi-stage liberation route where coarse lepidolite flotation would allow to minimize energy consumption, metal-bearing phase overgrinding and production of slimes.


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References
17. Lenharo, S.L.R.; Pollard, P.J.; Born, H. Petrology and textural evolution of granites associated with tin and rare-metals mineralization at the Pitinga mine, Amazonas, Brazil. Lithos 2003, 66, 37–61. [CrossRef]
24. Dehaine, Q.; Filippov, L.O. Rare earth (La, Ce, Nd) and rare metals (Sn, Nb, W) as by-product of kaolin production, Cornwall: Part 1: Selection and characterisation of the valuable stream. Miner. Eng. 2015, 76, 141–153. [CrossRef]
26. Schwartz, M.O. Geochemical criteria for distinguishing magmatic and metasomatic albite-enrichment in granitoids? Examples from the Ta-Li granite Yichun (China) and the Sn-W deposit Tikus (Indonesia). Miner. Depos. 1992, 27, 101–108. [CrossRef]
29. Dehaine, Q.; Filippov, L.O.; Joussemet, R. Rare earths (La, Ce, Nd) and rare metals (Sn, Nb, W) as by-products of kaolin production—Part 2: Gravity processing of micaceous residues. Miner. Eng. 2017, 100, 200–210. [CrossRef]
30. Filippov, L.O.; Dehaine, Q.; Filippova, I.V. Rare earths (La, Ce, Nd) and rare metals (Sn, Nb, W) as by-products of kaolin production—Part 3: Processing of fines using gravity and flotation. Miner. Eng. 2016, 95, 96–106. [CrossRef]
32. Cuney, M.; Marignac, C.; Weisbrod, A. The Beauvoir topaz-lepidolite albite granite (Massif Central, France); the disseminated magmatic Sn-Li-Ta-Nb-Be mineralization. Econ. Geol. 1992, 87, 1766–1794. [CrossRef]
35. Gébelin, A.; Brunel, M.; Moniè, P.; Faure, M.; Arnaud, N. Rare-elements (Li-Be-Ta-Sn-Nb) magmatism in the European Variscan belt, a review. Tectonics 2007, 26. [CrossRef]


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