Rare-metal granites as a potential source of critical metals: a geometallurgical case study

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Abstract

Because of their low grades in critical metals such as Light Rare Earth Elements (LREE) or Sn, rare-metal granites are not considered as economic for metal recovery but, when altered, they are often exploited for their industrial minerals. The St Austell rare-metal granite is well known for its world-class kaolin deposits which formed as a result of the extensive weathering and alteration of the underlying granite. The St Austell granite body is composed of several granite components, each having its own accessory minerals assemblage. As a result of the kaolinisation process, some metal-bearing accessory minerals of the granite, such as monazite (LREE) or cassiterite (Sn), are partially liberated from the gangue which allow their pre-concentration in the micaceous residue which is considered as a potential source for critical metals recovery. A geometallurgical approach is fundamental for this unconventional 'resource', not only because of its nature but also because the target metals must be considered as by-products. Similarities with other similar rare-metal granites suggest that topaz granite is the most prospective for disseminated magmatic Sn-Nb-Ta-REE mineralization. However, comparison of the potentiality of 3 granite types *i.e.* biotite, topaz and tourmaline granites suggest that biotite granites is actually the most prospective due to higher degree of kaolinisation of the biotite granite which favour pre-concentration of its accessory mineral in the micaceous residue. In order to develop a geometallurgical framework for extraction of kaolin and metals from the selected granite component, a field sampling campaign is performed. Core samples are processed in the laboratory using a characterisation program that mimics the full-scale kaolin refining route. Two main products are recovered through this program, *viz.* ~~GT53~~MR180 (-180+53 μm) and ~~R5~~P5 (-5 μm), which correspond to a fine micaceous residue and a fine kaolin product respectively. These products are both analysed routinely for major and minor trace elements by XRF and yields are recorded to indicate process performance. A selected number of ~~GT53~~MR180 samples are also being characterised in terms of particle size by laser light scattering, geochemistry by ICP-MS, and mineralogy by QEMSCAN®. Comparison of characterisation results of ~~GT53~~MR180 samples and corresponding industrial residue samples shows a good correlation, suggesting that sample analyses are representative for the in-situ deposit and the processing behaviour. Monazite is found to be either fully liberated or fully locked from one sample to the other. Next, pilot-scale gravity concentration tests are performed on micaceous residue samples. Characterisation of the processing products shows that monazite lost in the tailings is mostly locked within tourmaline or micas and is fine grained. Then, predictive regression models for spiral separation performance in terms of recovery, product grade and enrichment as a function of the feed grade are developed for ~~GT53~~MR180 LREE grade data. Finally, kaolin resources can be classified using quantitative indicators such as yield of the ~~R5~~P5 product and the iron oxides content which provides insight into the kaolin quality in terms of whiteness. This geometallurgical classification can be used to delineate zones of interest within the deposit. Although kaolin quality and recovery primarily inform extraction planning, zones which are also of interest for metal recovery can be identified. The proposed model predicts whether the expected LREE grade and recovery satisfy the by-product requirements.

Key words: kaolin residue, Light Rare Earths, critical metals, rare-metal granites, geometallurgy, QEMSCAN®.

1. INTRODUCTION

Ensuring sustainability all along the raw materials value chain has been a growing concern for the

extractive industry in recent years. While more efficient recycling of materials will be achieved in the

foreseeable future, as depicted by the concept of circular economy, recycling of some materials suffers

practical limitations which are not exempt from natural resources extraction with regards to our growing

societal needs. This is particularly true when it comes to critical metals which have a low recycling rate

and for which there is no substitute. These critical metals have been highlighted by a recent initiative by

the European Commission defining the criticality of a material based on three main indicators, *i.e.* the

supply risk, the economic importance and the environmental country risk. A list of 14 economically

important Critical Raw Materials (CRMs) was published in 2010, recently extended to 20 (European

Commission, 2014) which include Niobium (Nb) and the Light Rare Earth Elements (LREEs). For some

of these metals, mined as by-product, the availability is also conditioned by the major metal of the ore

in which they occurred usually in low concentration (Mudd et al., 2016). Rare Earth Elements (REEs)

for instance, are mainly recovered as co-products/by-products of certain other minerals in many mining

operations (Kumari et al., 2015). As of 2018, there are no more active mines in the western world

operated exclusively for the recovery of Rare Earths. REE projects outside China, which re-opened or

opened after the 2011 spike in REE prices, such as the Mountain Pass mine and the Bear Lodge deposit

(United States), or the Mount Weld mine (Australia), have all sustained or suspended their production

or development due to relatively low REE prices (Imholte et al., 2018). This situation has stimulated

other countries to look for alternative REE resources and to develop their own REE industry, in

particular by looking into recycling of REE from End-of-Life products. However, much less attention

has been paid to historical landfilled stocks and freshly produced flows of REE-containing industrial

process residues which usually present much lower REE concentrations but are present in very large

volumes (Binnemans et al., 2013). Total amounts of REE locked in these residues are also very large

and may secure an independent source of REE as well as shielding resource-poor countries from export

quotas and price fluctuations (Binnemans et al., 2015). Industrial mineral wastes, such as those from the

kaolin industry in Cornwall, UK, have proven to be a potential source for REE, Sn and Nb recovery

(Dehaine et al., 2017; Dehaine and Filippov, 2015a; Filippov et al., 2016; Scott et al., 1998). The kaolin

83 deposits are formed by weathering and alteration of granite rocks. As a consequence of the kaolinisation

84 process, accessory minerals of the granite are partially liberated from the gangue. Hence, during refining

85 of kaolin ore, spontaneous pre-concentration of the metal-bearing accessory minerals, such as monazite

86 hosting LREE (*i.e.,* cerium (Ce), lanthanum (La) and neodymium (Nd)), cassiterite hosting Sn or rutile

87 hosting Nb, occurs in the micaceous residue (Dehaine and Filippov, 2015a).

88 In order to convert such an unconventional resource into a potential REE resource, development of

89 ~~innovative technologies and geometallurgical approaches~~an integrated approach to mine value chain

90 optimisation ~~are~~is required on a strategic and tactical level. Geometallurgy is a discipline that seeks to

91 reduce technical risks through advanced ore characterisation which allows maximisation of resource

92 efficiency and minimisation of energy consumption and waste generation. This could be expressed in

93 the creation of a spatial predictive model which informs optimal extraction, processing and waste

94 disposal. ~~combines geological and metallurgical information to create a spatially-based predictive model~~

95 ~~for mineral processing plants in order to reduce technical risk~~ (Lamberg, 2011)~~. This aims to~~

96 ~~document~~The predictive model accounts for variability within the orebody ~~within the orebody and~~

97 ~~quantify the impact~~ of ~~some~~ geological, mineralogical, chemical and geotechnical factors that impact~~on~~

98 metallurgical processes (grinding, recovery) through ~~in order to classify the resources in~~

99 geometallurgical domains (Cabri et al., 2017). ~~The database thus generated can be readily integrated~~

100 ~~into a 3D model (Williams and Richardson, 2004).~~ Thus, it is necessary to develop simple and effective

101 characterisation methods of the distinct processing products/residues. This is particularly true when it

102 comes to industrial minerals such as kaolin for which the final product is a mineral concentrate rather

103 than a mono-element product (Glass, 2016). Geometallurgy can be broadly split into two key levels:

104 *strategic geometallurgy*, which focuses on the whole orebody and long-term life-of-mine scale and

105 *tactical geometallurgy* which relates to the short- to medium-term scale during daily operation (McKay

106 et al., 2016).

107 Such a strategic~~n~~ approach is fundamental for the unconventional 'resource' investigated in this

108 feasibility work, not only because of its nature but also because the target metals must be considered as

109 *by-products*. Applying a geometallurgical approach to this case study is rather challenging for several

110 reasons. First, there are no a priori certainties on the economic feasibility of metal by-product recovery

111 and there are no known specifications for ~~a~~ marketable product consisting of REE. Next, if the operation

112 is already in production, there is no processing on site for metal recovery from micaceous residues. This

113 influences the development of a hypothetical geometallurgical model: ~~most geometallurgical programs~~

114 ~~are applied to more conventional deposits already in operation for which a significant amount~~there is no

115 historical process ~~of~~ data ~~are~~ available, ~~with large numbers of~~nor a geometallurgical ~~variables~~ database

116 allowing the use of multivariate modelling (Boisvert et al., 2013). Finally, given the complexity of the

117 investigated deposits, with multiple granite-types each one having its own accessory mineral

118 assemblage, this approach could not consider the whole St Austell granite but will have to focus on a

119 specific granite type, ore-type or location. ~~The representativeness of the results that will be obtained is~~

120 ~~also a key feature for a potential application .~~ Hence, there is a need to develop performance prediction

121 models to evaluate process efficiency and product characteristics (metal grades and recovery) therefore

122 assessing the critical metals by-product potentiality. The proposed approach applied to a typical

123 kaolinised rare-metal granite seek to feed the initial strategic/feasibility work on similar deposit with the

124 objective of supporting tactical geometallurgy during kaolin production.

125

2. GEOLOGICAL BACKGROUND

127 The Cornubian granite province is well-known for its historically important deposits of tin and base

128 metals (Manning and Hill, 1990), and for its current international importance as a major kaolin producer.

129

2.1. The Cornubian Sn-W province

131 Tin is distributed on the continents in relatively narrow continent-sized belts (Schuiling, 1967). For

132 instance, most of tin deposits across Europe are related to Variscan granites. Figure 1~~Figure 1~~ illustrates

133 the multi-scale spatial relationship between Variscan granites and Sn-W deposits from the Variscan belt

134 to the St Austell district. The major tin belt passes from the Cornubian massif (Redruth, Camborne, etc.)

135 in UK through the French Massif Central (Echassières) and continues towards Italy with tertiary tin

136 deposits (Elbe, Tuscany and Scilly). Other tin belts are also related to Variscan granites, like the one

137 which pass from Northern Portugal (Panasqueira) through Spain or the one around the Bohemian massif

138 (Cinovec-Altenberg). Most of these tin deposits are associated with Sn-W cupolas located at the top of

139   tardi- to post-orogenic plutons emplace at shallow depths (Jébrak and Marcoux, 2008).

140   The batholith of SW England was formed during the late Variscan orogeny in the Late Carboniferous

141   to Early Permian (270-300 Ma) intruding deformed Devonian-Carboniferous terrestrial to marine

142   sediments. The batholith has a 200-km-long extension (Willis-Richards and Jackson, 1989), comprising

143   six major and several minors granite bodies. The major bodies, from east to west, are Dartmoor, Bodmin

144   Moor, St Austell, Carnmenellis, Land's End and the Scilly Isles (Figure 1Figure 1b).

145   The outcrops are dominated by biotite-bearing monzogranite, with minor intrusions of tourmaline-

146   bearing aplites, pegmatites and local fine-grained facies (Manning et al., 1996). In addition, there are

147   some occurrences of other granite types, resulting from multiple intrusive episodes, such as the lithium-

148   mica granites or the topaz-bearing granite in the Tregonning-Godolphin Granite, and the St Austell

149   Granite (Manning et al., 1996; Manning and Exley, 1984; Manning and Hill, 1990). The batholith was

150   affected by several episodes of alteration including quartz-tourmaline veining associated with

151   greisening, intrusion of rhyolite dykes, quartz-hematite veining. Kaolinisation is the last alteration event,

152   which is believed to have a meteoric origin (Psyrillos et al., 1998; Sheppard, 1977), and was relatively

153   extensive in the western part of the St Austell pluton where the majority of the active kaolin pits are

154   located (Figure 1Figure 1b).

155   According to Willis-Richards & Jackson (1989) there is a close spatial association between the

156   batholith and the rich Sn, Cu mineralisation of the Cornubian Peninsula. Most of the ores of Sn, Cu, W,

157   Zn and As came from vein deposit type mineralisation, along or parallel to the axis of the batholith

158   (Moon, 2010). Within the St Austell granite, zones of intense kaolinisation are spatially related to

159   swarms of steeply dipping, quartz-tourmaline±cassiterite±wolframite sheeted veins with greisen

160   (quartz-muscovite-tourmaline±topaz±wolframite) alteration selvages, observable at Goonbarrow pit

161   (Bray and Spooner, 1983). These mineral lodes are located South, North, or to a lesser extent within the

162   St Austell granite kaolinised area (Figure 1Figure 1c and Figure 2Figure 2).

163

164   **2.2.   The St Austell rare-metal granite**

165   Rare-metal granites are so-called because of their mineralisations being disseminated through the

166   granites rather than concentrated in layers, lodes, etc. The St Austell rare-metal granite share numerous

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characteristics with other granites of SW England, summarised in many reference papers and textbooks (Floyd et al., 1993; Manning, 1996; Manning and Hill, 1990). These granites were originally described by four different granite types, all observed in distinct SW England granites: biotite granite, lithium mica granite, the tourmaline granites and topaz granite (Hill and Manning, 1987). Additional granite type were added later based on field and textural observations within the St Austell granite, indicating complex late-stage magmatic and hydrothermal processes (Manning et al., 1996). The St Austell rare-metal granite is composed of six major granite types, each unit being characterised by a specific mineral assemblage and textures (Manning et al., 1996), see Figure 2Figure 2.

The *biotite granite* (BG) is the main lithology, which represent up to 70% of the outcrop of the St Austell granite, exposed in the Western Area, at the eastern part of the pluton and intermittently within the Central Area. It corresponds to the megacrystic biotite granite observed at Land's End and Dartmoor granites (Manning et al., 1996; Manning and Exley, 1984), see Figure 1Figure 1b. It is coarse grained, mainly composed of quartz, K-feldspar and micas with tourmaline and topaz as minor phases. Biotite is the main mica, but muscovite is also present. The main accessory phases have been reported as rutile, topaz, apatite, monazite, cassiterite, zircon and uraninite (Manning et al., 1996). The main host for the LREE differs depending on the authors. Works of Jefferies, (1985) on Carnmenellis biotite granite describe monazite as the main LREE-bearing mineral accounting for approximately 75 % of the total LREE content. In contrast microprobe analyses on samples from Cornubian granites by Alderton et al. (1980) show significant concentrations (up to 0.5%) of La, Ce, Nd and Sm in other phases, particularly apatite, but also zircon and titanite.

The *lithium mica granite* (LMG) has a similar grain size and texture to the biotite granite (Manning et al., 1996). It is characterised by the presence of lithium mica (zinnwaldite) and plagioclase (albite component). The accessory minerals are present as inclusion in zinnwaldite (apatite, monazite, zircon and rutile) or in plagioclase (apatite, fluorite and secondary micas).

The tourmaline granites were subdivided by Manning et al. (1996) in a *Tourmaline granite* (TG), it-self regrouping a globular quartz and medium coarse-grained equigranular facies, and in a *fine-grained tourmaline granite* (FGTG) with fine-grained (<0.5 mm) equigranular facies. The globular quartz facies is characterised by a considerable textural variation. It contains large quartz grains, phenocrysts of

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micropethite, zinnwaldite with a fine-grained groundmass of quartz, K-feldspar, plagioclase, zinnwaldite, tourmaline and topaz. The accessory phases are limited to apatite and rutile. On the other hand, the fine-grained facies is an equigranular rock with euhedral albite and zinnwaldite. Accessory minerals include monazite, apatite, zircon and rutile with some occurrences of arsenopyrite and cassiterite.

The *topaz granite* (TZG) is medium-grained, characterised by euhedral-subhedral fluorine-rich topaz, interstitial lithium mica, albite, plagioclase feldspar with perthitic orthoclase, and subhedral quartz (Manning and Hill, 1990). It contains a smaller proportion of accessory phase but a wider variety including sub-economic minerals (Manning et al., 1996). The accessory minerals are apatite, amblygonite ($LiAlPO_4F$), zircon, Nb-Ta oxides (columbite-tantalite and ilmenorutile) and uraninite. The mineralogy of the six granites is summarised in Table 1~~Table 1~~.

Apart from these magmatic disseminated mineralisations, Sn-W mineralisations associated with quartz-tourmaline sheeted veins previously described, are also present within the St Austell granite (Bray and Spooner, 1983). Whereas two different types of mineralisations, a Sn breccia-stockwork in the Treliver area, generally unenriched in base metals, and a E-W veins complex enriched in base metals have been identified north of the St Austell pluton (Camm and Dominy, 1999; Camm and Moon, 2001). In addition, Müller and Halls (2005) describe an intrusive tourmaline breccia in biotite granite at Wheal Remfry where zoned rutile is the main host mineral for Sn (up to 1.88%) and exhibits high W (up to 1.95%) and Nb (up to 2.05%) domains.

2.3.   Similarities with rare-metal granites worldwide

The spatial relationship between critical metals mineralisations and Variscan granites previously discussed is confirmed by the numerous similarities between the St Austell rare-metal granite and several rare-metal granites worldwide. From all the magmatic units of the St Austell pluton, the topaz granite is the youngest, and the most geochemically different, relatively enriched in Li, Rb and with a much higher Nb/Zr ratio (Manning and Hill, 1990). Nonetheless, there are close similarities between the mineralogical assemblages described above and the accessory minerals assemblage of evolved granite elsewhere described as peraluminous high to intermediate phosphorus granites (Linnen and Cuney,

223 2005; Simons et al., 2017). All the St Austell granite types are felsic, weakly peraluminous granites

224 (Manning et al., 1996). The comparison of the P_2O_5/SiO_2 ratio of the St Austell granites with other rare-

225 metal granites show that the St Austell granites belong to the intermediate phosphorous granite (IHP)

226 type (Figure 3Figure 3). The relatively low Zr and Th contents of St Austell granites, visible in Figure

227 3Figure 3b are characteristic of non-peralkaline rare-metal granites (Linnen and Cuney, 2005).

228 These granites are formed in a late-orogenic context and associated with orogenic belts (Figure

229 1Figure 1b) and are usually characterised by highly differentiated leucocratic two-micas granites,

230 corresponding to the S-type, rich in hydroxylated minerals (muscovite) and enriched in lithophile

231 elements such as Nb, Ta, Li, Be, P , F (Jébrak and Marcoux, 2008). Among these granites some display

232 similarities with the St Austell granite and are associated with Sn, W, and Ta-Nb disseminated

233 mineralisations (Černý et al., 2005). Some of this granites are associated to kaolin deposits which are

234 also known to display similarities with Cornish kaolin deposits (Wilson et al., 1997; Wilson and Jiranek,

235 1995).

236 According to Scott et al., (1998), the Yichun granite in China is one of the closest rare-metal granite,

237 in terms of lithology. It comprises also several magmatic units including a biotite granite and a Li-mica

238 (lepidolite) granite such as the St Austell granite. The Li-micas granite is the most evolved magmatic

239 unit and display a similar accessory assemblage of amblygonite/montrebasite, apatite, fluorite, Mn-

240 tantalite, microlite, Ta-cassiterite, ilmenite, monazite, zircon, sphalerite and topaz (Belkasmi et al.,

241 2000; Yin et al., 1995). The Beauvoir topaz-lepidolite albite granite (Massif Central, France) is also very

242 similar to the St Austell topaz granite and display a Sn-Li-Ta-Nb-Be disseminated mineralisation with

243 an accessory minerals assemblage composed of topaz, apatite, amblygonite, columbite-tantalite,

244 microlite, zircon, uraninite and sphalerite (Cuney et al., 1992; Raimbault et al., 1995). Further

245 similarities exist with the Podlesí granite in Czech Republic which is composed of three magmatic units,

246 including biotite granite, and two Li-mica granites. All granite type contain disseminated Ta-Nb-Ti-W-

247 Sn minerals including rutile, cassiterite, Fe-columbite, ixiolite and ferberite (Breiter et al., 2007). The

248 Younger Granites province of Nigeria displays also some similarities with the above-cited rare-metal

249 granite complexes, especially the subalkaline granites which are mineralised with rich deposits of

250 cassiterite, columbite, sphalerite, wolframite and galena and are the primary source of alluvial cassiterite

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251 and columbite mineralisation (Ogunleye et al., 2006).

252   By analogy with the Beauvoir granite it is possible to suggest the most prospective granite type within

253 the St Austell pluton for disseminated magmatic mineralisation. Rare-element chemistry of the Beauvoir

254 granite shows that the concentration of most metallic elements including Nb, Ta, Sn and W increases

255 with the Li contents from the bottom to the upper part of the granite body (Cuney et al., 1992). As

256 discussed above, the St Austell topaz granite exhibits the greatest enrichment in Li and is

257 petrogenetically distinct from other granite types (Manning and Hill, 1990) with mineralogical and

258 geochemical characteristics matching closely those of other rare metal granite worldwide. Thus, the St

259 Austell topaz granite is considered to be the most prospective for REE and Sn-Nb-Ta mineralisation.

260 The top of the Beauvoir Granite cupola is the most enriched part of the orebody in terms of rare-metals

261 due to a combination of magmatic process and interaction with meteoric water (Raimbault et al., 1995).

262 It is therefore the upper parts of the topaz granite that should have the greatest potential for disseminated

263 magmatic mineralization. The exact morphology and the intern organisation of the topaz granite is

264 poorly constrained but Manning and Hill (1990) suggested that the individual outcrops of topaz granite

265 may be connected at depth. The field relations indicate that the Nanpean stock represents a position well

266 below the roof of the granite body and that the Hensbarrow stock represents the roof. Thus, from a pure

267 metallogenic perspective, the Hensbarrow stock, which represents the upper part of the St Austell topaz

268 granite, is the most prospective for REE and Sn-Nb-Ta magmatic mineralisations.

269

3. ST AUSTELL KAOLIN OPERATIONS

271   China clay, also called kaolin, is a commercial clay material mainly composed of kaolinite

272 ($Al_2Si_2O_5(OH)_4$), a hydrated aluminosilicate clay mineral. Its main uses are for coating and filling in

273 paper industry (75% of world's production), the ceramics industry but it has also a wide variety of

274 speciality uses, *i.e.* for plastics, ink, paint, rubber, and pharmaceutical industry. The composition of the

275 granites has a significant influence on the quality of the kaolin ore (Scott et al., 2002). The commercial

276 value of a kaolin product is based on its whiteness and its fine particle size. Particle size affects fluidity,

277 strength, plasticity, colour, abrasiveness and ease of dispersion. Other important properties include its

278 flat particle shape, which increases opacity or hiding power, its soft and non-abrasive texture, due to the

279 absence of coarser impurities, and its chemical inertness (Highley et al., 2009).

280   The UK is one of the world's largest producers and exporters of kaolin, after the USA, Brazil and

281 China, supplying up to 1.36 million dry tonnes of clay per annum, in 2008 for the paper, ceramics,

282 paints, plastics and rubber industries (Highley et al., 2009). Production is supplied from a large number

283 of open pits scattered throughout the western part of the granite outcrop within a complex landscape of

284 spoil heaps and mica lagoons as shown in Figure 2Figure 2. Kaolinite is a secondary mineral formed by

285 weathering and hydrothermal alteration of the alumina silicates of host rock, mostly feldspar minerals,

286 by acid aqueous fluids to form an aggregate of kaolinite and secondary micas.

287   Kaolin extraction has traditionally been done by hydraulic mining in which high-pressure jets of

288 water are used to disaggregate the weak, kaolinised granite and disperse the kaolinite particles, together

289 with the other components of the granite, into a slurry, leaving behind the less well decomposed material

290 in the form of coarse sand and gravel, locally called "stent". The resulting "clay stream" flows into the

291 pit sink and is then pumped to the in-pit processing loop More recently, 'dry' mining has been

292 introduced, allowing more selective extraction and improving yields (Highley et al., 2009). The plant is

293 fed with clay matrix transported directly from the pit by cut and carry operations without any washing.

294 Hence, the material fed into the plant is much coarser and more classification stages are required to

295 remove the coarsest materials (Ano, 2008). The first stages of kaolin ore processing consist of removing

296 the coarsest fractions of the clay matrix which are not valuable for kaolin recovery. Thus, the kaolin

297 refining plant can be seen as a classification process producing different size classified wastes and a fine

298 kaolin product (Figure 4Figure 4). The first sized-classified wastes *i.e.* crushed stones (-21+11 cm),

299 gravels (-110+8 mm) and sands (-8+0.5 mm), are removed by screening and are sent to stockpiles. The

300 last classification stage is realised by series of hydrocyclones. The finer particles (overflow) go to the

301 product stream for further refining whereas the coarser particles (underflow) are reported to the

302 micaceous residue where they are held in suspension and pumped to a tailing dam. Additional techniques

303 are used to improve the brightness (whiteness) and particle size of specific grades of clay. These include

304 blending, fine grinding, chemical reductive bleaching and/or the removal of iron-bearing impurities

305 using magnetic separation. Some kaolin products are also calcined at specific temperatures to give

306 different products.

307   The gravity processing route presented in Figure 4Figure 4 corresponds to an hypothetical critical

308 metal by-product recovery route which have been tested on pilot-scale and described in Dehaine (2016).

309 It starts by a screening of the micaceous residue to remove the coarser fraction (+180 µm), which is not

310 treated, and preparation of the feed for the gravity concentration (-180+53 µm) and flotation routes (-53

311 µm). The proposed gravity concentration route comprise one spiral concentration step followed by a

312 refining of the spiral concentrate by shaking table concentration. For details about the flotation route see

313 Dehaine (2016) and Filippov et al. (2016).

314

315   **4. MATERIALS AND METHODS**

316 **4.1. Core samples**

317   All the core sample data used in this work are from a sonic drilling campaign undertaken in 2011 by

318 Imerys Minerals Ltd, UK. The dataset selected for this study covers only the Melbur-Virginia and Wheal

319 Remfry pits (referred as pit A and pit B hereafter) located at the western part of the St Austell granite

320 (see Figure 2Figure 2) and comprise around 1200 core sample collected every 3 meters over 100 drill

321 holes.

322 *4.1.1. Core samples processing protocol*

323   The main objective of this sampling campaign was to assess kaolin resources and alternatively

324 critical metals resources, in terms of yields and chemical composition of the potential end-products

325 through a laboratory-scale sample processing program that mimics the full-scale kaolin refining route

326 (Figure 5Figure 5). Samples The whole core samples were crushed to 15 mm if required (50 mm top

327 size) in a jaw-crusher and then stirred/mixed with water to liberate any kaolin from the kaolinised

328 granite. At this stage, the obtained material is equivalent to the output of the washing barrel of the plant

329 (stream 5, Figure 4Figure 4). The slurry is then classified on a vibrating 180 µm screen as with the

330 classification stages realised by the bucket-wheel de-sander/ 660 mm cyclones to obtain a -180 µm

331 material (stream 12, Figure 4Figure 4). This is followed by a screening of the -180 µm material on a

332 vibrating 53 µm screen as performed in the full-scale plant by primary cyclones. The -53 µm material

is then treated to remove the -53+15 μm material by sedimentation. If required, a magnetic separation

was then applied to the -15 μm material to improve brightness/colour as applied in the refining stages

of the industrial process. Finally, the -15+5 μm fractions are removed by sedimentation and a -5 μm

high quality kaolin product is recovered. In the end 3 products are recovered, *viz.* ~~GT53~~ MR180 (-

180+53 μm), ~~R15~~ P15 (-15+5 μm) and ~~R5~~ P5 (-5 μm) corresponding to the -180+53 μm fraction of the

micaceous residue fed to the gravity concentration route (stream 23, Figure 4~~Figure 4~~), a rough (-15 μm)

kaolin product and a fine (-5 μm) kaolin product (stream 16, Figure 4~~Figure 4~~), respectively. Each

product is weighed at each step of the process, so that the yield of each product is known. All the final

products were then analysed by X-Ray fluorescence (XRF) for major elements whereas ProTrace XRF

analyses (analyses for trace elements) were only carried out on the -180+53 μm (~~GT53~~MR180) and the

-5 μm (~~R5~~P5) fractions.

4.1.2. Core sub-sampling

In order to control the accuracy of the XRF data for the LREE and other metals a set of 30

~~GT53~~MR180 samples from the set of core samples covering the western part of the biotite granite, *i.e.*

kaolin pits A and B, have been selected to be analysed by ICP-MS for calibration and by QEMSCAN®

for mineral characterisation. All these samples correspond to relatively-high to high grade kaolin ore,

i.e. completely altered granite. The linear regression models for Nb and LREE prediction, obtained with

process samples in Dehaine and Filippov (2015), suggest that TiO_2 can be used as a "tracer" for Nb and

LREE. Moreover, TiO_2 is generally well quantified by XRF analysis. Hence, sub-samples have been

selected among all the available core samples following a procedure based on their TiO_2 content ~~to have~~

~~a~~to ensure that the obtained sub-set ~~representative~~ ~~sub-set~~ displays the same variability as the original

dataset (Dehaine, 2016). In the end, only 26 usable samples were recovered due to technical issues

(contamination, losses).

4.2. Process samples for metal accounting and for gravity concentration testwork

The samples used for metal accounting were collected and aggregated from daily sampling over one

week from every output stream of three distinct kaolin refining plants processing ore from Virginia-

360 Melbur, Wheal- Martyn or Treviscoe pit. These samples are considered to be typical material from the

361 biotite, tourmaline and topaz granite respectively. For each location, the batch of samples were

362 composed of waste samples and a kaolin product, *i.e.* 5 to 6 representative samples each, depending of

363 the mining technique (dry or hydraulic mining). Each sample is a composite, aggregated from

364 increments collected on-site from moving streams or belts, for details about the sampling procedure see

365 (Dehaine, (2016). To ensure an optimised sampling procedure in terms of frequency, sampling mode

366 and number of increments for all the critical metals, a multivariate variographic approach has been

367 developed to determine the required number of increments (Dehaine et al., 2016; Dehaine and Filippov,

368 2015b).

369 Process samples used for gravity concentration are screened (-180+53 μm) micaceous residues

370 corresponding to the spiral feed described in Figure 4Figure 4. Micaceous residue samples were

371 collected from the hydrocyclone underflow tank of the full-scale plant while treating material from the

372 same origin as the core samples, *i.e.* kaolin ore from pits A and B. Residue samples were collected using

373 a by-pass to divert the whole stream into the sample collector, following the protocol described in

374 Dehaine et al. (2016). The large micaceous residue samples collected were then screened in two

375 successive steps, using a vibrating Roto-Sieve apparatus model RS60 (Chauvin SA, France) in order to

376 recover the -180+53 μm size fraction of the residue for the pilot scale gravity concentration testwork.

377 The sieves opening have been selected based on the washability of the materials (Dehaine et al., 2017),

378 in order to improve the efficiency of the gravity concentration, but also to be able to use data from the

379 GT53MR180 core sample products s-to characterise the feed properties of the gravity route with the

380 objective of establishing process performance prediction models for core sample data.

381

382 **4.3. Pilot-scale gravity concentration testwork**

383 The influence of the LREE grade in the feed on spiral concentration efficiency has been tested with

384 a 5-turn MKIIA Reichert spiral (Mineral Deposit Limited, Australia) using the setup described in

385 Dehaine et al. (2017) in scavenging configuration. Operating condition are set at 0.12 $m^3.h^{-1}$ wash water

386 flowrate and 15% solid pulp density to favour recovery (Dehaine et al., 2017), concentrate splitters

387 positions are adjusted visually for the first test but remains fixed for all the others. Spiral concentrates

388 are then processed using a No.13 Wilfley laboratory shaking table (Holman-Wilfley, United-Kingdom)

389 following the flowsheet presented in Figure 4Figure 4. The characterisation results for the shaking table

390 presented in this paper have been obtained using the shaking table products obtained from test No.4 in

391 Dehaine et al. (2017), which display the best separation performance.

392

393 **4.4. Particle size analysis**

394   Particle size analysis was performed by laser light scattering using a Helium-Neon Laser Optical

395 System Mastersizer 3000 (Malvern instruments Ltd.) coupled with a Hydro Extended Volume (EV)

396 sample dispersion unit. Required amounts of sample were introduced in a beaker inside the dispersion

397 unit, equipped with a dip-in pump and a stirrer, to reach a 20% obscuration level. The particle size

398 distributions obtained are the average of five duplicate analyses.

399

400 **4.5. Chemical analysis**

401   Samples were crushed and riffled alternatively in accordance with the theory of sampling to obtain

402 representative subsamples suitable for whole rock analysis. Powdered samples (300 mg) are fused in Pt

403 crucibles along with ultra-pure $LiBO_2$ at 980 °C and dissolved in nitric acid. The chemical analysis of

404 the obtained solutions were carried out by Inductively Coupled Plasma Optical-Emission Spectrometry

405 (ICP-OES, Thermo Fischer ICap 6500) for major elements (Si, Al, Fe, Mn, Mg, Na, Ca, K, Ti, P) and

406 Mass Spectrometry (ICP-MS, Thermo Elemental X7) for 42 trace elements, following the routine

407 procedure of liquid chromatography ICP-MS described in Carignan et al. (2001), at the *Service*

408 *d'Analyses des Roches et des Minéraux* (SARM-CNRS, Nancy, France).

409

410 **4.6. Mineral analysis**

411   For mineralogical data samples were analysed using QEMSCAN®. Samples were ~~randomly sub-~~

412 ~~sampled~~riffled, mixed with graphite powder to promote particle dispersion and reduce settling bias

413 (Pascoe et al., 2007) and then prepared into 30 mm diameter epoxy resin blocks. The sample face of

414 each block was then polished to a 1 micron finish using diamond solutions and carbon coated with 25

415 nm layer, prior to analysis. Samples were analysed using a QEMSCAN 4300 system at the Camborne

School of mines, University of Exeter, UK (Andersen et al., 2009; Anderson et al., 2014; Rollinson et al., 2011) using PMA mode at a resolution of 5 microns, and a target of 5000 particles per sample (see Goodall et al., 2005; Gottlieb et al., 2000; Pirrie and Rollinson, 2011, for detail on the PMA measurement mode). ~~As the samples were well mixed and carefully prepared, the 5000 particles provided~~ a representative ~~sub-population of each sample.~~ Data measurement used iMeasure 4.2 SR1 and data processing used iDiscover software 4.2SR1 and 4.3. Data processing involved checking the data, customizing the database to the match samples, adding and improving SIP (database) categories as required, whilst considering the details of the technique outlined in Rollinson et al. (2011). Modal mineralogy, mineral association, average grain size, liberation data and images were output for the samples.

Additional textural observations were performed on a Hitachi FEG S4800 scanning electron microscope at the *Service Commun de Microscopie Electronique et Microanalyses* (SCMEM-UL, GeoRessources, Nancy, France) with an imaging voltage of 20 kV.

4.7. Methodology

The purpose of this work is to investigate and develop preliminary geometallurgical tools for by-product resource estimation of critical metals within the St Austell kaolin deposits with a focus on LREE. The methodology used in this study follows three main steps which aim to populate the core sample database: (1) to characterise the ~~GT53~~MR180 core samples through QEMSCAN analysis, to control, and calibrate if necessary, the trace element grades using duplicate ICP analyses (not presented here, see Dehaine (2016) for details), then (2) to integrate metallurgical parameters through regression models (*e.g.* spiral performance prediction models) applied to the ~~GT53~~MR180 trace element data. Finally (3), to use the core samples data along with the regression models to propose a simple preliminary geometallurgical classification scheme to help by-product resources estimation of critical metals.

5. RESULTS

5.1. Comparison of the granite types

Given the range and diversity of accessory minerals within the different granite types present at St Austell, it is likely that each granite type will have a ~~the same~~different potential as for by-product recovery of critical metal. In order to define the most prospective granite type for by-product metal recovery, one must compare the metal content of all the output streams of the kaolin processing route and the associated metal accounting. This is done for three distinct kaolin refining plants processing ore from weathered biotite, tourmaline or topaz granite.

Previous studies on kaolin ore originating from biotite granite have shown that metals are pre-concentrated in the micaceous residue, as a consequence of the preferred separation of the heavy minerals in the hydrocyclone underflow (Dehaine and Filippov, 2015a). Comparison of the critical metal content of the output streams from the 3 investigated locations is presented in Table 2. Most of the metals, with the exception of LREE, display a similar trend than observed for the biotite granite, with an increasing grade from the coarsest waste to the micaceous residue which display the highest overall grades. The LREE grade however is slightly higher in the product stream displaying the highest LREE grade for topaz and tourmaline which means that LREE are more concentrated in the finer fractions of the clay matrix.

In terms of distribution within the output~~s~~ streams, most of the metals are distributed in the coarsest streams (stent and sand) for topaz and tourmaline granite, or the micaceous residue for biotite granite (Figure 6~~Figure 6~~). Most of the metals display a less contrasted distribution for tourmaline and topaz granite. Indeed Sn, Nb and Ta are slightly more distributed in the sands then equally distributed between stent and the micaceous residue whereas the LREE show a more important proportion in the stent and product stream. This could be a consequence of the hydraulic mining processes as the stent fraction also contains fine material which sticks to the boulders or gravels.

The overall LREE and Sn content are significantly higher for the biotite granite than for the topaz and tourmaline granite. Nb and Ta content in the micaceous residue are of the same order of magnitude for the 3 locations, whereas the W content is three times lower for the topaz granite. ~~This highlights~~This highlight strong differences in the metal content of the kaolin ore originated from different granite types. This is confirmed by the recalculated feed (clay matrix) contents which clearly shows higher LREE, Sn

16

472 and W grades in the biotite granite clay matrix while Nb-Ta grades are slightly higher for topaz granite

473 clay matrix (Table 2). This is somehow surprising as the St Austell topaz granite, covered by the

474 Treviscoe kaolin operations, was considered as one of the most prospective source of disseminated

475 magmatic mineralisations from a metallogenic perspective. In addition, the high Nb-Ta grades obtained

476 after a two-stage gravity concentration on hydrocyclone underflow samples presented in Scott et al.

477 (1998), suggested that micaceous residue from these areas may have a potential interest for metal

478 recovery. This could be explained by the variations in the intensity of kaolinisation within the St Austell

479 kaolin deposit. The kaolin ore from the topaz granite is lower grade (i.e. less weathered) than the kaolin

480 ore from biotite granite. A less kaolinised ore (and thus less liberated metal-bearing minerals) means

481 lower yields and larger proportions of coarse wastes.

482   It appears that, as a consequence the variations in the intensity of kaolinisation within the St Austell

483 and differences in the mining technique, the micaceous residue from biotite granite, is the most

484 prospective stream both in terms of metal content and distribution. In addition, the corresponding kaolin

485 pits are among the largest pits located in the area associated with the more reliable extraction and

486 processing method and the granularity of the micaceous residue doesn't require additional grinding prior

487 to further processing.

488

489 **5.2. Core samples characterisation and validation of the geometallurgical program**

490   One of the primary ~~objective~~objectives of this study is to validate the core sample processing program

491 to ensure its data is reliable and can be used for geometallurgical modelling purpose. Hence, it is

492 necessary to compare the properties of the products obtained from the core samples treatment in the

493 laboratory with actual process samples collected from the plant. Since kaolin processing is mainly a size

494 classification process, the particle size distribution will be investigated, followed by the metal grades

495 and mineralogy of the samples which are both critical for the recovery of critical metals.

496 *5.2.1. Particle size distribution*

497   Particle size distribution of the spiral feed may significantly impact the performance of the separation

498 with the spiral concentrator. The influence of particle size on spiral performance in terms of heavy

499 mineral recovery has been investigated by Dehaine and Filippov (2016) who observed that for the

17

500 operating conditions at which the test have been performed, the variations of heavy minerals recovery

501 with particle size is limited and only decreases for particle sizes below 50 μm.

502 Particle size analysis of the ~~GT53~~MR180 samples shows that the mean particle size distribution is very

503 close to the one of the spiral feeds (Figure 7~~Figure 6~~). This suggests that ~~GT53~~MR180 samples are on

504 average representative, in terms of particle size, of the -180+53 μm size fraction of the micaceous

505 residue which is processed by the gravity concentration route. It can be observed that the variation in

506 particle size distribution of the ~~GT53~~MR180 (-180+53 μm) fraction is relatively limited and is more

507 important in the -50+10 μm size range. These variations are still limited compared to the high variability

508 of the size distribution properties of the raw micaceous residue stream observed in Dehaine et al. (2016).

509 However, these results were obtained from the raw micaceous residue, and these variations may be less

510 visible on the screened spiral feed. In addition, the overall limited variations displayed by the core

511 samples may also be explained by the fact that the samples set is only sourced from highly kaolinised

512 ore and that more variations (toward the coarser fractions) should be observed with lower grade, *i.e.* less

513 altered, material. Variations in particle size distribution for the spiral feed are more limited and are

514 almost always included into the variation intervals of the ~~GT53~~MR180 particle size distribution. These

515 variations, however, do not seem to affect the performance of the spiral as illustrated in Figure 7~~Figure~~

516 ~~6~~ by the rather small variations of spiral concentrate particle size distribution. In addition, it can be seen

517 that the finer size ranges below 30-40 μm, for which the spiral feed particle size variations are more

518 important, are poorly recovered in the spiral concentrate. This result is in agreement with the results

519 from size-recovery curves analyses presented in Dehaine and Filippov (2016), and suggests that

520 variations in particle sizes, at least for the finer size range is unlikely to influence the performance of

521 the separation.

522

523 *5.2.2. Whole rock geochemistry*

524 The critical metals and REEs composition spectra for all ~~GT53~~MR180 core samples products are

525 presented in Figure 8~~Figure 8~~ab along ~~with the average raw micaceous residue~~a high grade spiral feed

526 sample and corresponding fresh granite trace element composition for comparison. These spectrum

527 display clear enrichments in certain critical metals, decreasing in the order Sn, Nb, Zr and to a lesser

18

528   extent Ga (~~Figure 8~~Figure 7a) as well as an enrichment in LREE, decreasing in the order Ce, La, Nd

529 (~~Figure 8~~Figure 7b).

530   The ~~average~~ geochemical composition of the high-grade ~~micaceous residue~~spiral feed sample shows

531 a very similar pattern with however overall grades ~~above~~ around the maximum of those of the

532 ~~GT53~~MR180 samples for Nb, Ta, Zr, Y and REE. This correspondance suggests that MR180 samples

533 are on average representative, in terms of critical metal content, of the -180+53 µm size fraction of the

534 micaceous residue which is processed by the gravity concentration route. ~~This is due to the fact that these~~

535 ~~elements are known to be distributed in the finer size ranges of the micaceous residue (Dehaine and~~

536 ~~Filippov, 2015a) which are not recovered in the GT53MR180 samples (-180+53 µm).~~ One sample (ref.

537 11069860) is highlighted in ~~Figure 8~~Figure 8 due to its significantly different REE pattern, displaying

538 an enrichment in Y and in Heavy Rare Earths Elements (HREEs), probably due to the presence of

539 xenotime in that sample (Jefferies, 1985).

540   Comparison with geochemical data from fresh St Austell biotite granite (Simons et al., 2017, 2016)

541 display a strong enrichment in Sn, Nb, Ta and W in the ~~GT53~~MR180 core samples. This suggests an

542 enrichment of these metals in the kaolinised granite whereas indium is on the contrary strongly depleted.

543 The chondrite-normalised plot (~~Figure 8~~Figure 7c) displays a LREE enriched pattern and a strong

544 anomaly in Eu in agreement with trace element geochemistry of the St Austell biotite granite established

545 in previous studies (Chappell and Hine, 2006; Simons et al., 2017, 2016).

546   ~~Figure 9~~Figure 8a shows a positive linear trends line between Nb and TiO_2 (R^2=0.70). This

547 proportional relationship, also observed at microscopic scale and macroscopic scale in Dehaine and

548 Filippov (2015) suggest that Nb is most likely associated with a TiO_2-bearing mineral, *i.e.* rutile,

549 ilmenorutile or ilmenite. These results may appear in contradiction with the observations of Simons et

550 al. (2017) which suggest biotite (siderophyllite) is the main host for Nb in G3 (biotite) granites and

551 rutile/ilmenite in G4 (tourmaline) granites. While this may indicate a change in granite type at depth, as

552 biotite and tourmaline granites are known to be spatially associated, for example, crosscutting as sheets

553 or forming gradational contacts (Simons et al., 2017), it may also point to the alteration of most brown

554 micas in highly kaolinised material which are almost completely weathered by the kaolinisation process

555 (see Mueller et al. (1999), and mineralogy section hereafter).

556   Another interesting correlation is the strong correlation ($R^2 > 0.99$) between each LREE suggesting

557   constant relative proportions of LREE with a Ce/La ratio of 2.27, a Ce/Nd ratio of 2.41, a Ce/Pr ratio of

558   8.67 and a Ce/Sm ratio of 12.85 in all ~~GT53~~MR180 samples (Figure 9~~Figure 8~~b). These correlations

559   and the corresponding ratios are close from the one determined by whole-rock analysis on process

560   samples and by Energy-Dispersive X-ray Spectroscopy (EDS) analyses on monazite grains in Dehaine

561   and Filippov (2015). This other multi-scale correspondence of element-to-element ratios from the

562   macroscopic-deposit scale (core samples) to the microscopic scale (monazite grains) through the

563   mesoscopic scale (process samples) is a strong evidence that monazite is the only LREE-bearing mineral

564   in the St Austell biotite granite.

565   *5.2.3. Mineral characteristics*

566   The average modal mineralogy of the ~~GT53~~MR180 core samples obtained by QEMSCAN® analysis

567   is presented in Table 1~~Table 1~~. The mineralogy is largely dominated by quartz (48.7 wt %), followed by

568   fine secondary white-micas (muscovite) and tourmaline (schorl-dravite). Other minerals accounting for

569   less than 5 wt % each include remaining feldspars (orthoclase, microcline) and biotite (siderophyllite-

570   annite), topaz, Ti-oxides (mostly rutile, ilmenorutile and ilmenite) and kaolinite. This is consistent with

571   the mineralogy of the biotite granite described in Manning et al. (1996) and in agreement with the

572   mineralogical compositions of altered biotite granite (Mueller et al., 1999). The overall modal

573   mineralogy is very similar to the one of raw micaceous residues previously investigated, even if this size

574   fraction seem to contain more quartz and less biotite and feldspar than typical raw residues from biotite

575   granite (Dehaine et al., 2017; Dehaine and Filippov, 2015a).

576   In terms of accessory minerals hosting the critical metals, QEMSCAN® results confirmed cassiterite

577   as the main host for Sn, rutile-ilmenorutile as the main host for Nb and monazite as the host mineral for

578   LREE. These results confirm the observations of Dehaine and Filippov (2015), even if very little

579   wolframite is observed in the samples due the overall low W grades.

580   Mineral association data obtained by QEMSCAN® quantifies which mineral grain is adjacent to or

581   touches another mineral grain in a particle. Figure 10~~Figure 9~~ displays the monazite association data for

582   the ~~GT53~~MR180 samples based on QEMSCAN® results. On average, monazite grains mostly have no

583   association (36.9%), *i.e.* monazite boundary pixel occurs adjacent to a pixel of the background, which

20

584   means that most of the monazite grains surface seem to be free. The next major associations of monazite

585   is with tourmaline (17.3%) and micas (14.1%) and then by decreasing order Ti-oxides, zircon and quartz.

586   The average monazite liberation histogram suggests it is either locked (<10% free) or liberated

587   (>90% free) in the ~~GT53~~MR180 samples (Figure 11~~Figure 10~~). However, the large error bars suggest a

588   large variability of liberation data for all core samples. This is because each sample contains a distinct

589   population of monazite. Indeed, samples can be roughly split into two categories, the ones in which

590   monazite is almost completely locked, representing 50% of the core samples, and the ones in which

591   monazite is almost completely liberated (see, for example, Figure 12~~Figure 11~~a), representing 47% of

592   the core samples. When locked, monazite is often included in tourmaline (Figure 12~~Figure 11~~b),

593   secondary muscovite (Figure 12~~Figure 11~~c) or rutile (Figure 12~~Figure 11~~d). This relates to the

594   observations made on fresh biotite granite in which monazite is mostly included, along with zircon, in

595   biotite and to a lesser extent in tourmaline resulting in metamictisation (Simons et al., 2016). The results

596   obtained for the ~~GT53~~MR180 samples could be linked to the kaolinisation process, which is known to

597   affect brown micas from moderated de-ferruginisation in slightly altered granite to complete

598   decomposition in the completely altered biotite granite whereas tourmaline remains poorly altered even

599   for the highest degree of kaolinisation (Mueller et al., 1999). Given the highly kaolinised ore considered

600   in this study, it is likely that monazite grains which were previously included in biotite have been

601   progressively liberated during the decomposition of biotite whereas those included in tourmaline

602   remained locked resulting in the above-mentioned liberation characteristics of monazite in the

603   ~~GT53~~MR180 samples. It also worth mentioning that monazite has been found to be sometimes

604   associated with reticulated rutile (Figure 12~~Figure 11~~e). These ultrafine monazite grains display a needle

605   shape which has the same orientation that the rutile structure. It is not clear whether these needle-shaped

606   monazites just lay on the rutile surface or if they are actually associated with the rutile, but the presence

607   of intergrowths and the orientation of the monazite needles seems to suggest that there is an association

608   (Figure 12~~Figure 11~~f).

609   Characterisation results influence the recovery of monazite through the gravity concentration process

610   presented in Figure 4~~Figure 4~~. The high proportions of tourmaline and micas in the ~~GT53~~MR180 (-

611   180+53 µm) fraction may be troublesome for the gravity concentration techniques investigated. Indeed,

21

612 tourmaline has an average density close to monazite (Table 3Table 3) whereas fine phyllosilicates are

613 known to behave erratically in gravity operations. Phyllosilicates are known to cause gravity processing

614 problems, especially by increasing the slurry viscosity (Filippov et al., 2016; Sivamohan and Forssberg,

615 1985a). Monazite is mostly associated or included in tourmaline or muscovite and half of the samples

616 only contain locked monazite. Therefore only half of the LREE content of the -180+53 µm fraction of

617 the micaceous residue is potentially recoverable, which could explain some of the low recoveries

618 obtained in previous attempts to recover monazite from micaceous residues (Dehaine et al., 2017).

619

620 **5.3. Gravity concentration testwork on micaceous residue**

621 Large process samples were collected on-site from the micaceous residue stream of the kaolin

622 processing plant and prepared to correspond to the -180+53 µm feed of the pilot-scale gravity

623 concentration route described in Figure 4Figure 4. The samples were fed in a two-stage gravity

624 concentration route, starting by a spiral concentrator in scavenging configuration to maximise the

625 recovery. Spiral concentrates are then fed to a shaking table to increase the grade. Results from these

626 tests work are discussed from a mineral processing perspective process modelling and optimisation

627 perspective in Dehaine et al. (2017) and from process mineralogy and geometallurgical perspective in

628 the following sections.

629 *5.3.1. Characterisation of monazite in gravity products*

630 Monazite characterisation data in the different gravity concentration products obtained by

631 QEMSCAN® are presented in terms of mineral association and mineral liberation in Figure 13Figure 12

632 and Figure 14Figure 13 respectively. It can be seen that most of the monazite recovered in the gravity

633 concentrates is liberated even if some monazite is, to a lesser extent, locked in tourmaline or Ti-oxides.

634 The proportion of liberated monazite decreases in the middlings whereas the proportion of locked

635 monazite increases, in particular for the shaking table tailings. Most of the locked monazite in the

636 middlings is associated with tourmaline which has an intermediate density (3.12 g.cm^{-3}) between those

637 of the gangue minerals (2.60-2.82 g.cm^{-3}) and the one of monazite or the other critical metal-bearing

638 minerals ($\geq$4.25 g.cm^{-3}). Other significant monazite mineral associations in the middlings include zircon,

639 micas and Ti-oxides. The monazite lost in the gravity tailings is mostly locked, even if a non-negligible

640 proportion is partially (≤80%) or fully (≤100%) liberated. These locked monazite grains appear to be

641 mostly locked within tourmaline in gravity tailings but results for the shaking table suggest that a non-

642 negligible proportion of the monazite lost in the tailings is also associated with micas and kaolinite.

643   These results should be considered in parallel to the average monazite grain size in the various gravity

644 concentration products (Figure 15Figure 14). It can be seen that coarse monazite tends to report to the

645 gravity concentrates whereas ultra-fine (-10 μm) monazite, either liberated or locked, reports to the

646 tailings. This is consistent as recovery in the ultrafine particle size range is poor with most gravity

647 processing techniques (Burt and Mills, 1984).

648

649 *5.3.2. Spiral performance prediction models*

650   Spiral concentration is a gravity separation technique which differentiate minerals according to

651 particle size, density, and shape (Burt and Mills, 1984). In addition, some other important ore

652 characteristics such as feed grade, mineral associations or liberation degree may play an important role

653 on the separation performance (Sivamohan and Forssberg, 1985b). Most of these parameters can only

654 be assessed by systematic mineralogical and textural analysis, for instance via QEMSCAN®. This type

655 of analysis is time consuming and expensive and, in this study, only a few samples were characterised

656 using this technique. However, the grade of the spiral feed is known as the feed material of the spiral

657 concentration tests consist in the -180+53 μm fraction of the micaceous residue which corresponds to

658 the GT53MR180 samples analysed for trace elements, including LREE. Indeed, LREE content of the

659 GT53MR180 samples, calibrated using the ICP-MS results on duplicate samples, can be used as an

660 estimate of the LREE feed grade for the gravity concentration circuit. A multivariate analysis applied to

661 this LREE content and other important mineralogical properties of monazite (i.e., monazite liberation,

662 associations and grain size) allows the identification of the most important correlations between these

663 properties (Table 4Table 4). There is a strong positive correlation (R^2=0.83) between the average

664 monazite grain size and degree of liberation suggesting that the coarser is the monazite the more likely

665 it is liberated. In addition, monazite grain size and degree of liberation are both correlated to the LREE

666 content with correlation coefficient of 0.73 and 0.65 respectively. All these properties are known to have

667 a positive influence on gravity concentration performance, *i.e.*, coarse and liberated monazite

23

668   preferentially report to the concentrates (Figure 14Figure 13 and Figure 15Figure 14). The fact that there

669   are all positively correlated is an indication that the LREE feed grade could potentially be used to predict

670   spiral concentration performance.

671   The influence of the LREE grade in the feed material on the spiral concentration efficiency has been

672   tested using the spiral setup in scavenging configuration. To reflect the representativeness of the spiral

673   feeds in terms of grade, the LREE feed grade (G_F) is normalised using the minimum and maximum

674   LREE contents found in the core sample database as follows:

$$G_F^* = \frac{G_F - min}{max - min} \tag{1}$$

675

676   G_F^* can therefore vary between 0 and 1, these two extreme values corresponding to the case where

677   G_F^* equals respectively to the minimum or the maximum LREE content in the core sample database.

678   Figure 16Figure 15 shows the influence of the normalised LREE grade (G_F^*) in the feed material on the

679   performance of spiral concentration of LREE. The results show that the normalised feed grades are well

680   distributed over the [0;1] interval which suggests that they cover almost the whole range of LREE values

681   displayed in the GT53MR180 core samples data. As expected, spiral performance is found to improve

682   with increasing feed grade. Indeed, LREE recovery and concentrate grade increase with LREE content

683   in the feed, with a maximum recovery and concentrate grade of 55% and 1000 ppm respectively.

684   However, LREE enrichment degree reaches a plateau at 4.5 for LREE normalised feed grade higher

685   than 0.6-0.7 (~ 150 ppm LREE). The relationship between spiral performance indexes (*y*) and

686   normalised feed grade shown in Figure 16Figure 15 exhibits clear trends that can be fitted by an

687   empirical model, using the equation:

$$y = a + b\ exp(c\ G^*{}_F) \tag{2}$$

    with *a: Asymptote*
           b: Scale
           c: Growth rate

688

689   The parameters *a*, *b* and *c* are fitted, and have no physical meaning. However, it is expected that they

690   would be linked to changes in mineralogy or liberation degree which consequently affects spiral

691   performance. The values of the fitted parameters are given in Table 5Table 5. Comparison between the

692 observed and predicted values shows a good agreement with high regression coefficient values (R²>0.9),

693 suggesting that the models can reasonably be used to estimate spiral performance (Table 5~~Table 5~~). All

694 performance indexes trend lines suggest that below 0.2 LREE normalised feed grade (around 60 ppm

695 LREE), no separation should be expected (Figure 16~~Figure 15~~). Hence, this value could be considered

696 as the minimum grade of the ~~GT53~~MR180 fraction that should be considered for LREE recovery.

697   The aforementioned results suggest that the regression models proposed to predict spiral performance

698 for LREE can reasonably been applied to ~~GT53~~MR180 core sample data. In addition, the range of LREE

699 feed grade used to calibrate the models, as mentioned before, covers almost the whole range of

700 ~~GT53~~MR180 LREE grades (from 0.25 to 0.95 normalised feed grade) therefore suggesting a good

701 representativeness of the results. Hence, prediction of spiral performance using a core sample database

702 can be realised using the following equation (with coefficient values given in Table 5~~Table 5~~):

$$y = \begin{cases} a + b\ exp(c\ G_F), & for\ G^*{}_F \geq 0.2 \quad\quad (3) \\ 0 & for\ G^*{}_F < 0.2 \quad\quad (4) \end{cases}$$

703

704

6. DISCUSSION AND CONCLUSION

6.1. Geometallurgical classification proposal

707   This study represents the first step in the development of an integrated geometallurgical model for

708 ~~the~~ optimised extraction ~~St Austell kaolin deposits that takes into account~~ of ~~both~~ kaolin and critical

709 metals ~~from the~~ St Austell deposits ~~rare metal granite~~resources. ~~Generally~~ For extraction

710 purposes~~speaking~~, two distinct approaches can~~ould either~~ be used to create a spatially-based predictive

711 geometallurgical model. The ~~first one is based on geometallurgical testing which~~traditional approach

712 involves small scale laboratory geometallurgical tests which aim to measure the metallurgical response

713 of ~~the~~ samples in order to build predictive performance ~~prediction~~ models through simulation (Bulled

714 and Mcinnes, 2005). ~~The~~ Because the metallurgical response is ~~then~~ reported on an elemental basis,

715 ~~therefore the~~ geometallurgical variation~~s~~ in the deposit ~~orebody~~ ~~are~~is only based on elemental grades

716 ~~only but not on~~rather than on mineralogical information. ~~The second~~ A more recent approach~~, based on~~

717 ~~particles,~~ uses quantitative characterisation of particles and the minerals ~~contained in the~~and particles

25

to develop the geometallurgical program (Lamberg, 2011). Process models can use ore characteristics, including mineral abundance, grain size, associations, degree of liberation, grindability, recovery potential, etc., as parameters in studies and simulations.

The geometallurgical approach described in this paper is unconventional, and borrows aspects from both aforementioned approaches. Firstly, this study is based on systematic lab-scale core sample processing program which mimics the full-scale kaolin processing plant. This is an unusual approach which generates sub-samples out of core samples. Rather than performing specific metallurgical tests referring to one particular unit operation, processing of these samples yields similar products and residues as produced by the full-scale process. In this case, if the final spiral performance prediction models are based on LREE grade and consider the mineralogy of the orebody indirectly. Indeed, LREE is only hosted by monazite, and a positive correlation is observed between the LREE grade and mineralogical factors such as liberation or monazite grain size (Table 4), which influence the recovery. This explains why the process performance can be predicted using only the LREE feed grade.

6.1.1.1. Geometallurgical classification proposal

A geometallurgical ore type classification for critical metals recovery must consider the kaolin resources in which metals are regarded as potential

746 by-products, . In other words, ~~*i.e.* to be considered for metal recovery,~~ the ore is considered for metal

747 recovery when it is extracted for the primary ~~must primarily valuable for the main~~ commodity, *i.e.*

748 kaolin~~, to be considered for metal recovery~~. When developing an ore-type classification for

749 geometallurgical purposes it is also important to capture essential elements which are relevant in terms

750 of metallurgical and commercial significance~~meaning~~.

751   A geometallurgical ore -type classification scheme proposal for ~~the~~ kaolin resources is presented in

752 Figure 17~~Figure 16~~a. It is based on the core sample data related to the R5P5 sub-samples which is an

753 indicator of the fine kaolin product. The first classification criterion for the kaolin resources is the yield

754 of the R5P5 product, *i.e.* the weight ratio of the R5P5 product over the feed. The R5P5 product is almost

755 entirely composed of kaolinite plus ancillary minerals. Hence, the yield of the R5P5 product gives an

756 indication of the future kaolin recovery, which is one of the main metallurgical response for the kaolin

757 refining operation considered. It is closely related to the classical geological ore classification based on

758 the degree of kaolinisation of the granite rock using visual inspection of texture preservation and

759 feldspars alteration, as the more kaolinised is the granite the higher will be the kaolin yield once it is

760 processed. The next classification criterion is the FeO content of the R5P5 products, which reflect the

761 brightness/whiteness of the kaolin product (Bertolino et al., 2010). This is a key property of the kaolin

762 product as it drives the~~its~~ commercial end-use ~~and will define to which industry it will be directed~~. In

763 the end, five different ore grades are defined from ~~the~~ Grade I, nearly fresh to partly kaolinised granite

764 (R5P5 yield<10%), to ~~the~~ Grade V, fully kaolinised granite (R5P5 yield>15%) and low iron content

765 (FeO<0.4%), material.

766   Classifying the kaolin ore in terms of its potential for critical metals recovery could be performed

767 using the process performance prediction models. Figure 17~~Figure 16~~b shows an example of the

768 distribution of the predicted LREE recovery after the spiral concentration step for two kaolin pits by

769 applying the empirical models presented in equations (3)~~(3)~~ and (4)~~(4)~~ to the corresponding

770 ~~GT53~~MR180 core sample data. One could use this chart to classify the resources in distinct recovery

771 classes but also to delimitate the potential resources itself using the minimum feed grade suggested by

772 the models (equations (4)~~(4)~~).

773

6.2. Application to core sample

The application of the aforementioned geometallurgical classification schemes to an actual drill core is presented in ~~Figure 18~~Figure 17. Applying the spiral performance prediction models to ~~GT53~~MR180 data allows evaluation of the potential separation performance in terms of concentrate grade or recovery. This data, along with data pertaining to the ~~R5~~P5 product (*i.e.*, yield and FeO grade), could then be used to classify both kaolin and critical metals resources using the corresponding geometallurgical classification schemes. These domains could be used to first define the zones of interest for kaolin and then to establish if these zones are also of interest for metal recovery, therefore targeting the zones of interest for metals recovery as a by-product of kaolin.

While this ~~approach~~ preliminary work is promising ~~and inexpensive~~, it would benefit from being validated by comparing the predicted performances with those obtained after processing micaceous residue samples collected when refining a block of ore for which the ~~GT53~~MR180 grades are known. This may be very difficult for several technical reasons including ore traceability, contamination, large sample collection, etc. In most geometallurgical approaches, process performance prediction models are built using production data comprising a large dataset collected after months of production. Hence implementation on site of a primary concentration process (spiral concentrators for instance) seems to be the next logical step. Indeed, it would give the opportunity to collect enough data for developing similar process performance prediction models and could also allow assessing the effect of the ore variability in terms of metal grades, alteration grade and lithology (granite type) which is fundamental for the ~~development~~ implementation of a ~~proper~~ tactical geometallurgical ~~model~~approach. In addition, it would allow recovering directly some primary concentrate samples for metallurgical testing, therefore removing the need for large residue samples collection. In the meantime, while no actual critical metal recovery is applied on site, this ~~inexpensive~~ approach could still be used to store the residues properly according to their potential for metals recovery.

6.3. On the commercial potential of kaolinised granites for metal recovery

Assessing the commercial potential of kaolin residues for metal recovery is not an easy task even

802   after all the work performed on this case study (Dehaine et al., 2017; Dehaine and Filippov, 2015a;

803   Filippov et al., 2016; Scott et al., 1998). Some of the kaolinised granites mentioned in section 2.3 have

804   been considered as potential sources for Nb, Ta, Sn or W and some of them are actually mined for these

805   metals. For instance, the Yichun granite is the largest Ta, Nb and Li raw material producer in China

806   (Schwartz, 1992; Yichun Tantalum Co., 2005) and the Beauvoir granite is known to produce cassiterite

807   (800 g/t Sn) and columbite (190 g/t Ta plus 120 g/t Nb) as by-products of kaolin production (Negroni,

808   2015; Pohl, 2011). There are also some mining operations such as the Greenbushes pegmatite complex,

809   (Australia) or the Manono-Kitotolo deposit (DRC) for which the situation is reversed and where Sn, Ta,

810   Nb or Li are primary products but kaolin is regarded as a by-product (Laznicka, 2006).

811   From a resources point of view, the metal grades within the kaolinised rare-metal granites are low,

812   most of them are actually well below the usual cut-off grades, but the volumes are large and the

813   mineralisation is disseminated within the ore which is already exploited for the kaolin. The potentiality

814   is also dependent on the granite type from which the kaolin ore originate and the variability in terms of

815   intensity of kaolinisation as well as the extraction method. Therefore, the St Austell metal granite which

816   is considered as one of the most prospective sources of disseminated magmatic mineralisations from a

817   metallogenic perspective is not necessarily the most prospective in terms of by-product recovery of

818   metals from kaolin production.

819   Whilst previous work on St Austell kaolin residues has demonstrated that heavy minerals can be

820   recovered and concentrated from kaolin residues using a simple processing route with a limited number

821   of operations, the metal grades obtained are still relatively low and further processing steps will be

822   required before reaching an acceptable concentrate grade (Dehaine et al., 2017). The effectiveness of

823   the heavy mineral recovery for this case study is accounted to the complexity of the considered material

824   which is low grade, fine grained and contains high proportions of medium density minerals and clay

825   minerals. However, high enrichment ratios (10-60) and reasonable recoveries, at least for LREE (~60%),

826   have been obtained in a few separation steps without any additional grinding required.

827   From an environmental point of view, only benefits could arise from a potential processing of

828   micaceous residue as the proposed processing route has a low environmental footprint with low energy

829   consumption (gravity concentration) and environment friendly chemical reagents (dispersion,

830 flotation). In addition, processing the micaceous residue will result in a reduced waste production (and

831 even more if historical tailing dams are considered) and a better waste management strategy (wastes

832 sorted by size fractions).

833 From an economic point of view, the arguments are more balanced. Indeed, the mineral concentrates

834 obtained by beneficiating the kaolin residues is rather atypical, with no equivalent on the market and

835 thus there is no reference for the terms of a potential contract (Negroni, 2015). Another limitation is the

836 application of penalties due to the presence of several metal-bearing minerals that would impair the

837 value of the concentrate. The more restrictive issue facing LREE production is associated with the

838 potential radioactive activity of the concentrate due to the presence of Th in monazite (Zhu et al., 2015).

839 This means that a limitation in terms of LREE concentration should be accepted above which some

840 special dispositions controls must be taken (dry processing, dust control, etc.). The annual income

841 cannot be estimated but an assessment of the annual production, considering only the grade V ore from

842 the sole kaolin pits investigated in this study, suggests that around 15.5 t/y LREE could be recovered in

843 metal pre-concentrates (Dehaine, 2016). Although the current concentrate grade is low, the annual

844 tonnage is high and total capital investment for the metal recovery process is low (Dehaine, 2016). With

845 demand for Nd and Ce expected to grow in the future due to their use in permanent magnets (Kingsnorth,

846 2014b), metal recovery from kaolin residue is worth investigating for other kaolinised rare-metal

847 granites worldwide.

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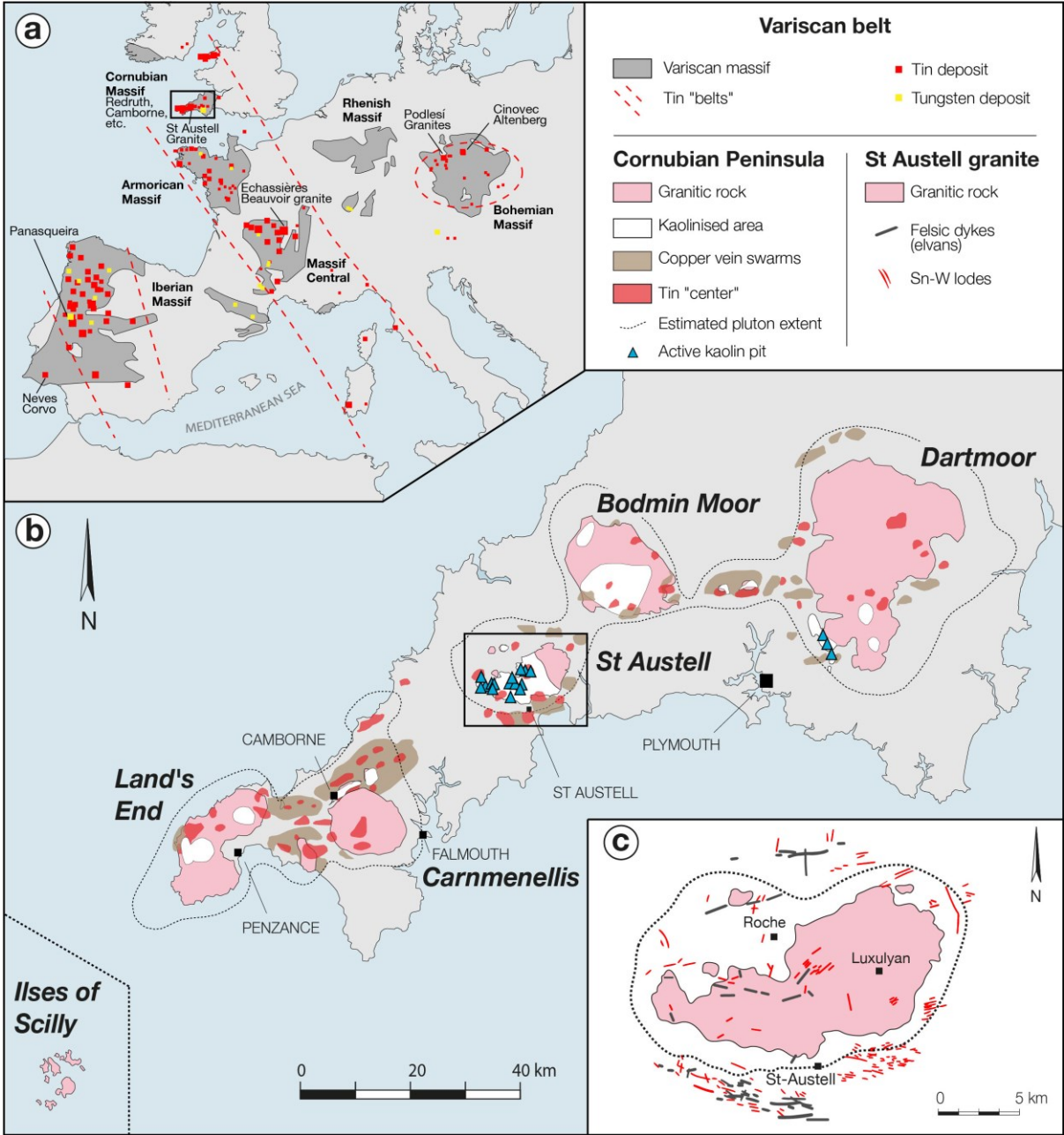
FIGURES

Figure 1. Spatial relationship at distinct geological scales between Variscan granites and tin or tungsten deposits.
(a) Tin belts in Europe and their spatial connection to the Variscan belt modified from Schuiling (1967). Main tin
and tungsten deposits are from the PROMINE project (BRGM, 2011). (b) Outline geological map of South-West
England peninsula showing the location of the major plutons of the Cornubian batholith, the kaolinised areas and
the active kaolin pits, modified from Černý et al. (2005). (c) Distribution of the felsite dykes (elvans) and Sn-W
mineral lodes around and within the St Austell rare-metal granite after Bray and Spooner (1983).
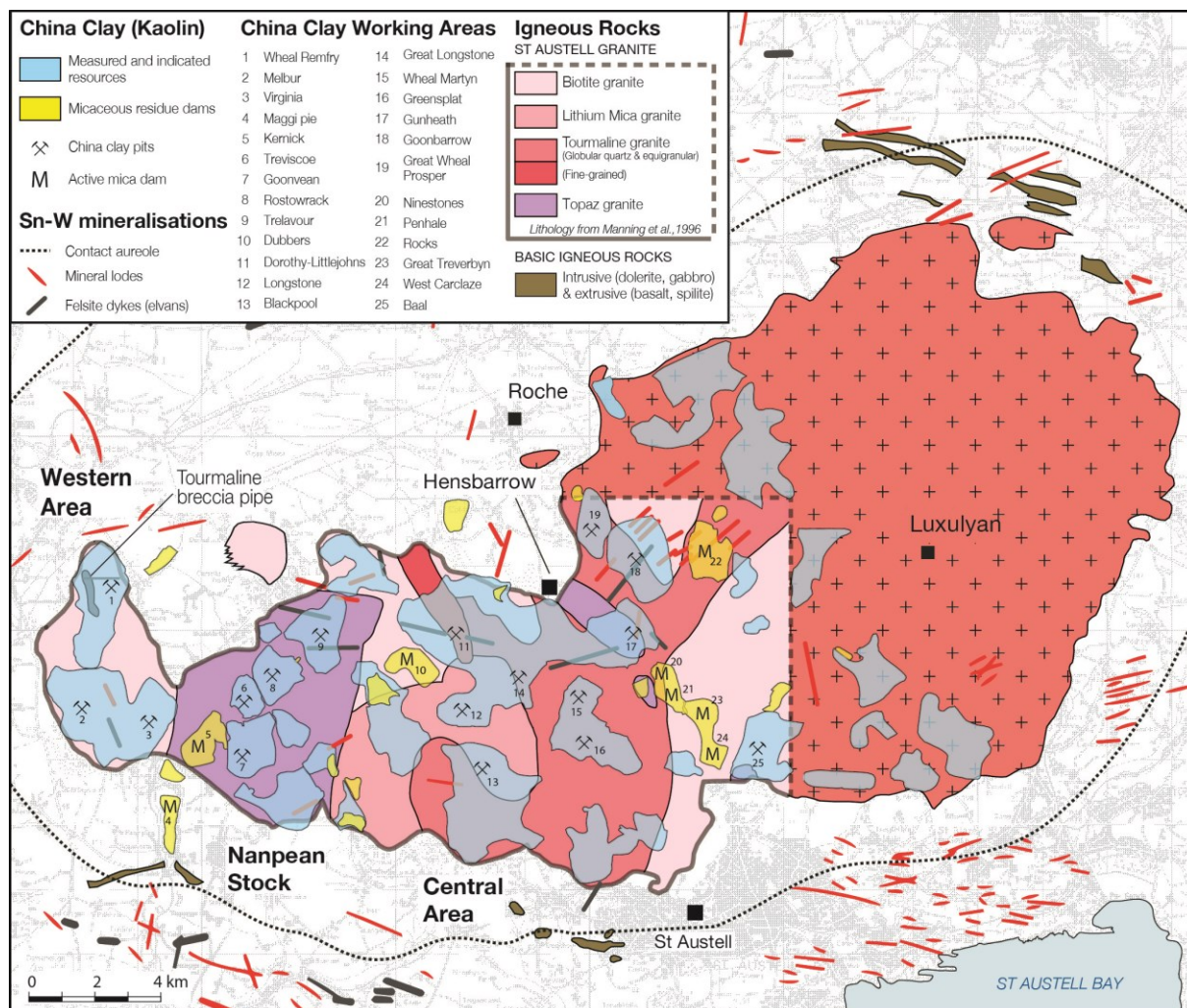
Figure 2. Geological map of the St Austell granite showing the distribution of the various granite types from Manning et al. (1996), the major kaolin resources and the micaceous residue dams after British Geological Survey Geological Map Data (British Geological Survey, 1997), as well as the main Sn-W mineralisation after Bray and Spooner (1983).

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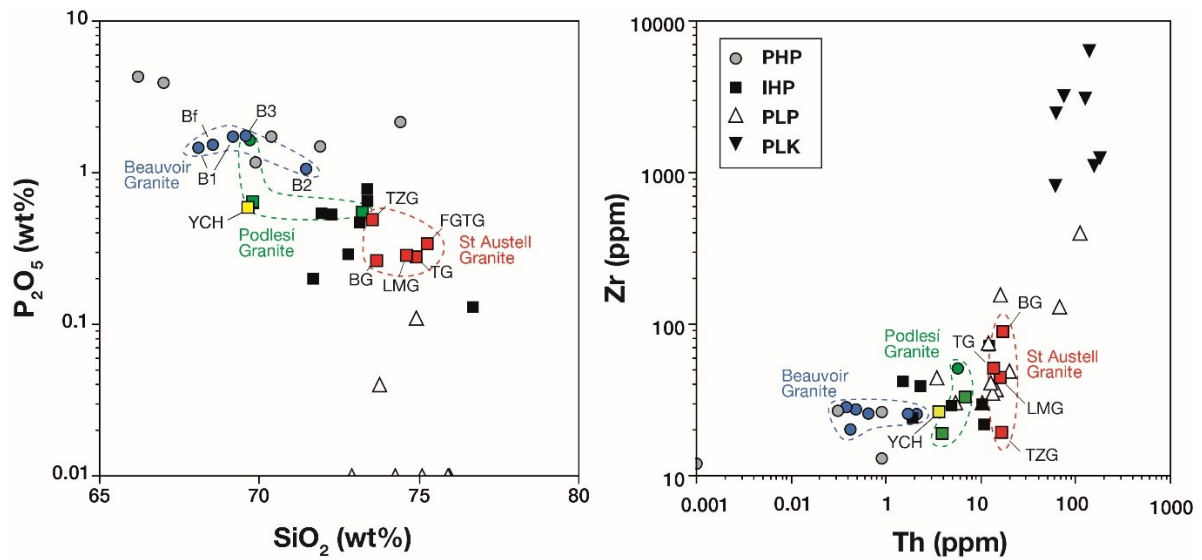
Figure 3. Trace element characteristics of peraluminous high phosphorous (PHP), peraluminous intermediate phosphorous (IHP), peraluminous low phosphorous (PLP) and peralkaline granites (PLK). (a) Silica-phosphorus diagram. (b) Zirconium-thorium diagram. B series: Beauvoir granite, Massif Central; YCH: Yichun granite, Jiangxi, China. Abbreviations for St Austell granites are given in the text. Based on data from Manning et al., (1996) and the review of Raimbault et al. (1995).

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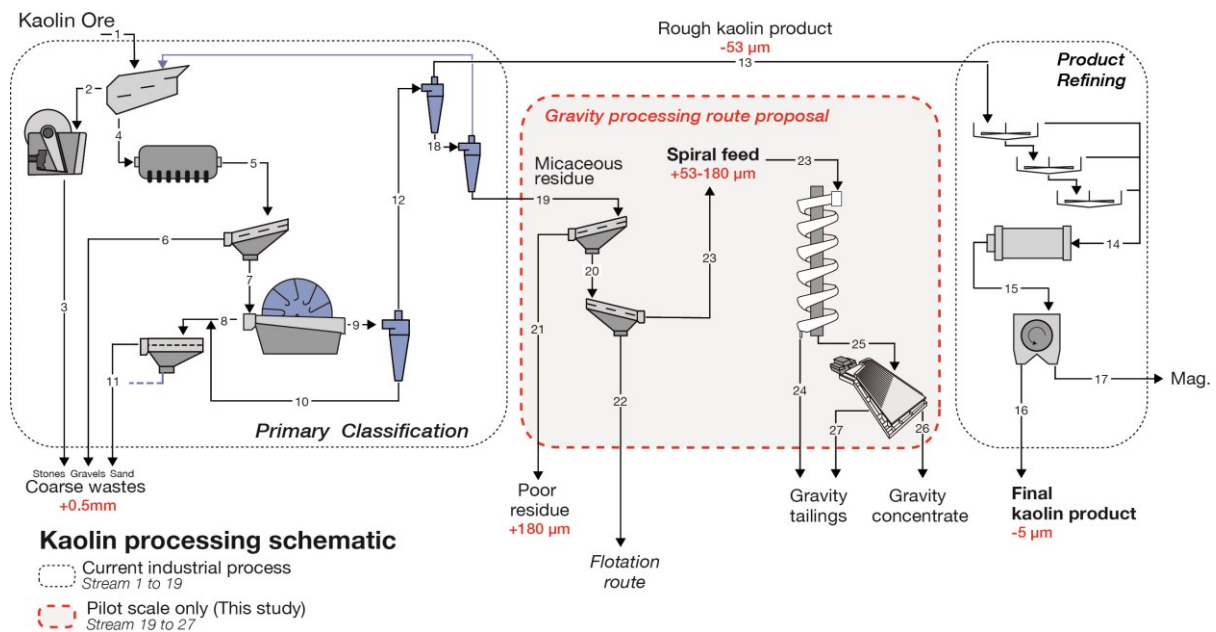
Figure 4. Simplified schematic of the kaolin ore processing and the by-product gravity processing route proposal.

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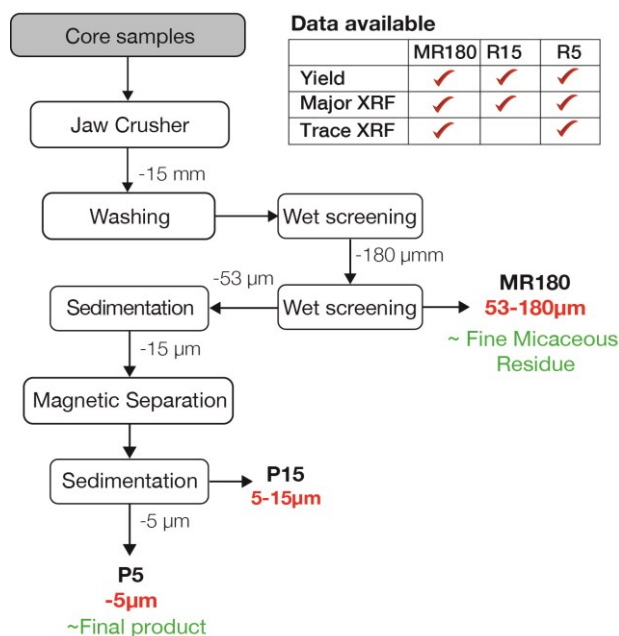
Figure 5. Sample treatment protocol of the geometallurgical program. ~~R5~~P5 and ~~GT53~~MR180 products

correspond to stream 16 and 23 in Figure 4~~Figure 4~~ respectively.

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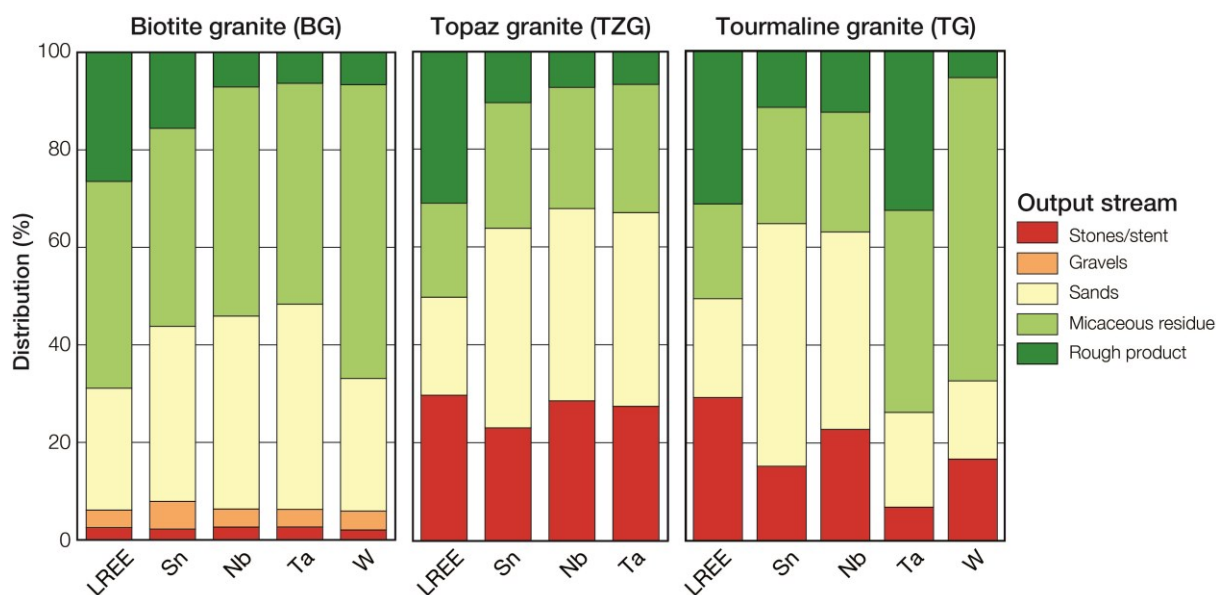
Figure 6. Metallurgical balances showing the distribution of the LREE (La, Ce, Nd) and rare metals (Sn, Nb, Ta, W) within the output streams of kaolin refining process for biotite, topaz and tourmaline granites.

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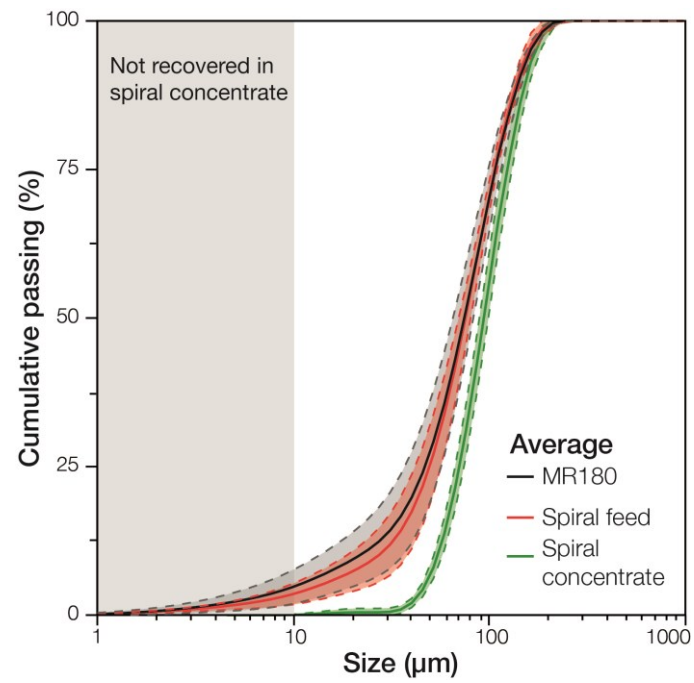
Figure 76. Comparison of particle size distribution between ~~GT53~~MR180 samples and corresponding

1080   process samples (spiral feed). Solid lines represent the average and dashed lines the lower and upper fence.

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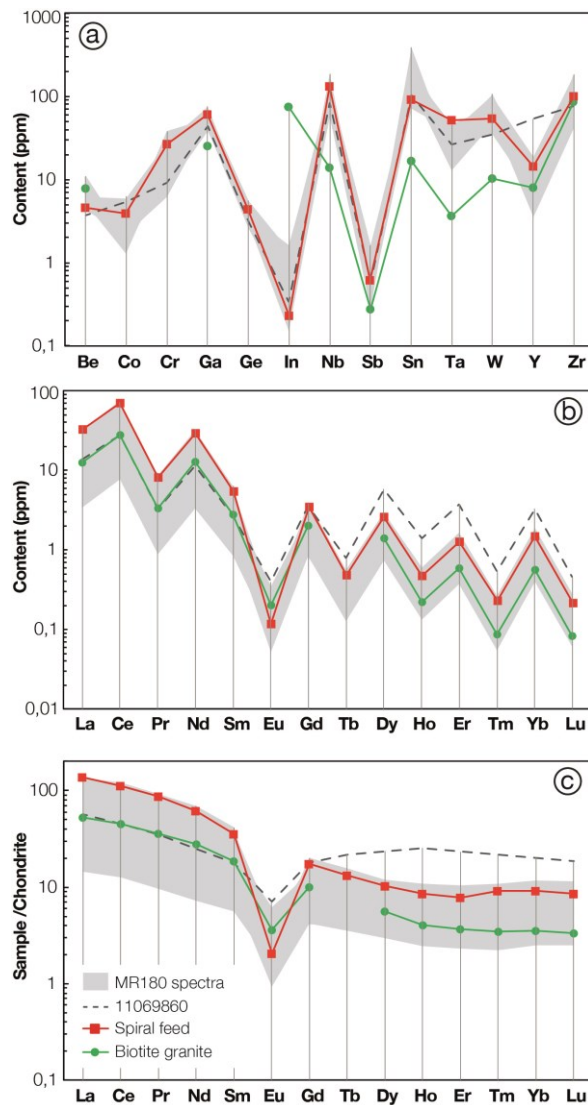
Figure 87. Geochemical composition spectra of the ~~GT53~~MR180 core samples products compared to the a ~~raw~~ ~~micaceous residue~~spiral feed sample, obtained by ICP-MS. (a) Critical metals contents, plus Sn, Ta, Y and Zr. (b) REEs content. (c) Chondrite normalised REEs content using the values of McDonough and Sun (1995). Sample 11069860 is highlighted due to its significantly different REE pattern, "~~micaceous residue~~Spiral feed" refers to the ~~average of 5 micaceous reside~~ highest grade process samples fed to the spiral and "biotite granite" corresponds to the average REE data of 3 fresh St Austell G3 (biotite) granite from Simons et al. (2016).

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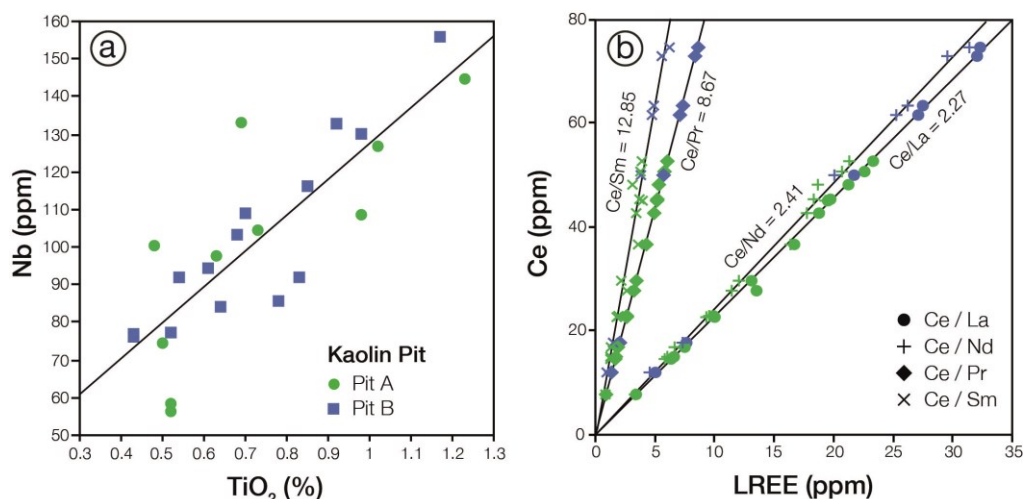
1090 **Figure 98.** Binary linear correlation plots and trend lines For Nb/TiO₂ and LREE using ~~GT53~~MR180 samples

1091 ICP-MS data. (a) Good correlation (R²=0.70) suggesting a proportional relationship between TiO₂ and Nb. (b)

1092 Strong correlations (R²>0.99) suggesting a proportional relationship between each LREE, similar to the one

1093 observed on process samples and at the microscopic scale on monazite grains in Dehaine and Filippov (2015).

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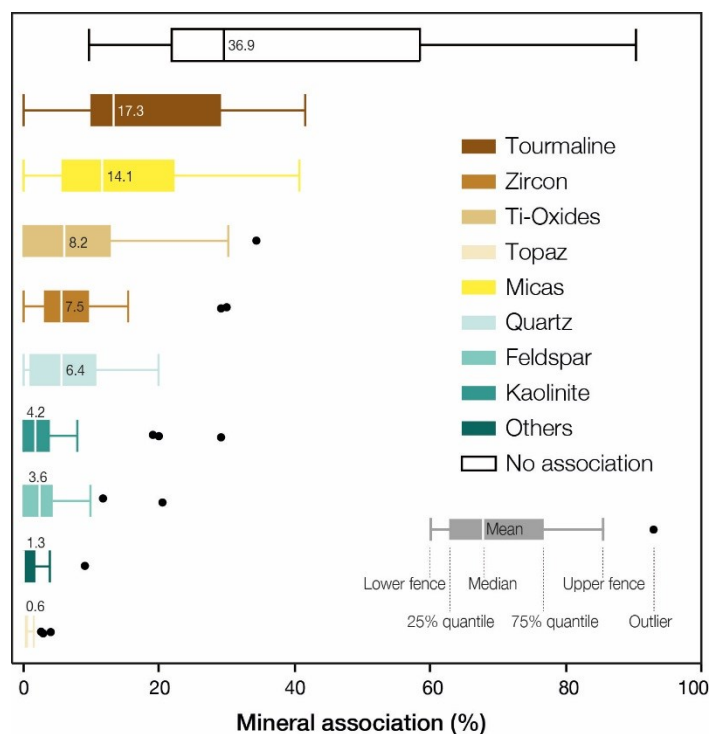
1096 **Figure 109.** Box-plots summarising monazite mineral association data in decreasing order for the

1097 ~~GT53~~MR180 samples.

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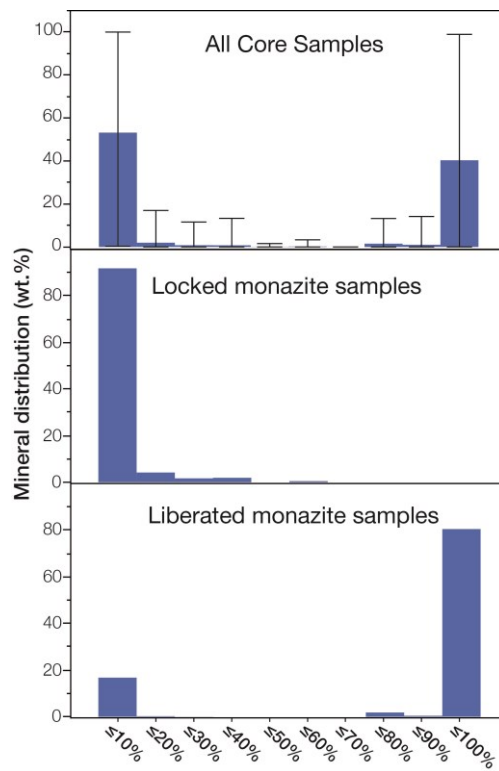
1099 **Figure 11~~10~~.** Average monazite liberation for all ~~GT53~~MR180 samples (top), for locked monazite only samples

1100 (middle) and for liberated monazite only samples (bottom).
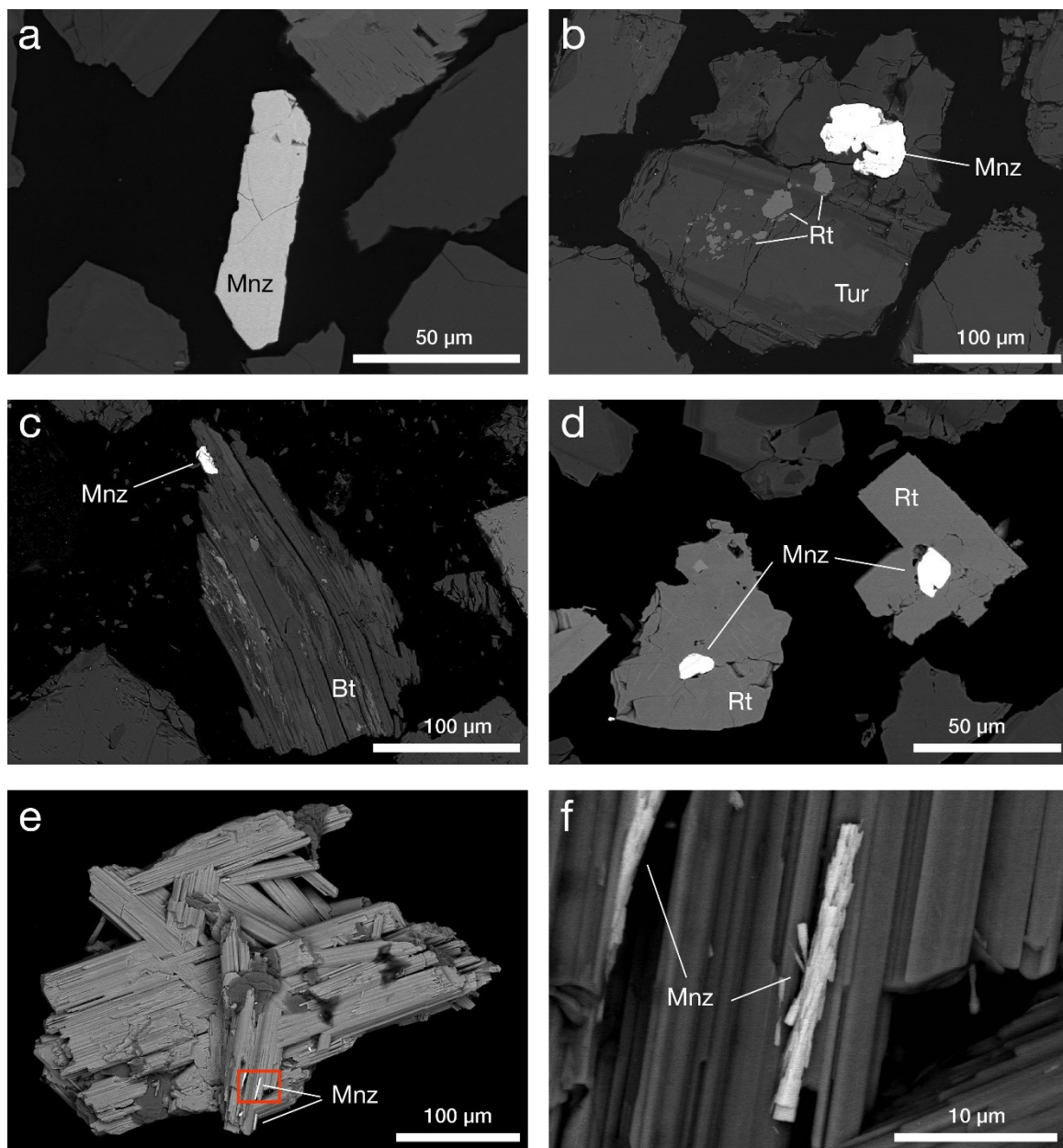
Figure 1211. Scanning electron micrograph in back-scattered electron mode illustrating the various habitus of monazite in the micaceous residue. (a) Liberated monazite (Mnz) grain, (b) Monazite included in a zoned tourmaline (Tur) with rutile (Rt) inclusions, (c) Weathered biotite (Bt) with inclusion of monazite, (d) Monazite included in Nb-rich rutiles, (e) Monazite associated with reticulated rutile and (f) Zoomed image showing a needle-shaped monazite oriented along the rutile structure.
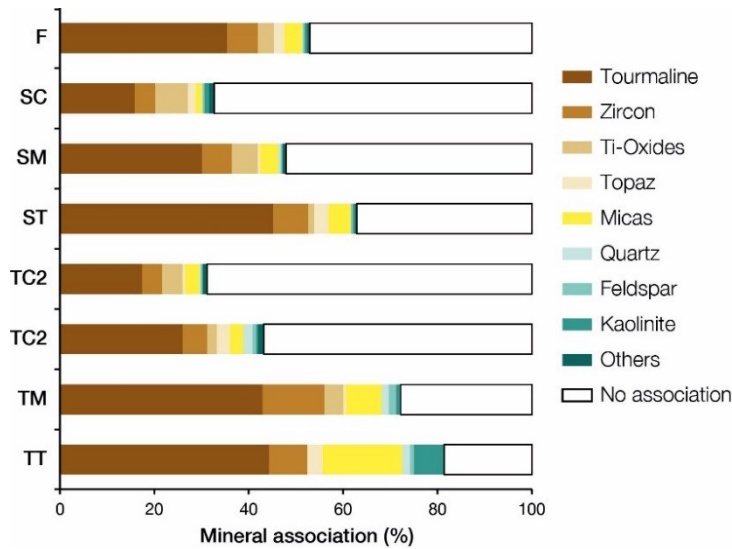
Figure 13~~12~~. Average mineral association for monazite in the gravity concentration products. F: Feed, SC: Spiral Concentrate, SM: Spiral Middlings, ST: Spiral Tailings, TC1: Table Concentrate 1, TC2: Table Concentrate 2, TM: Table Middlings, TT: Table Tailings.
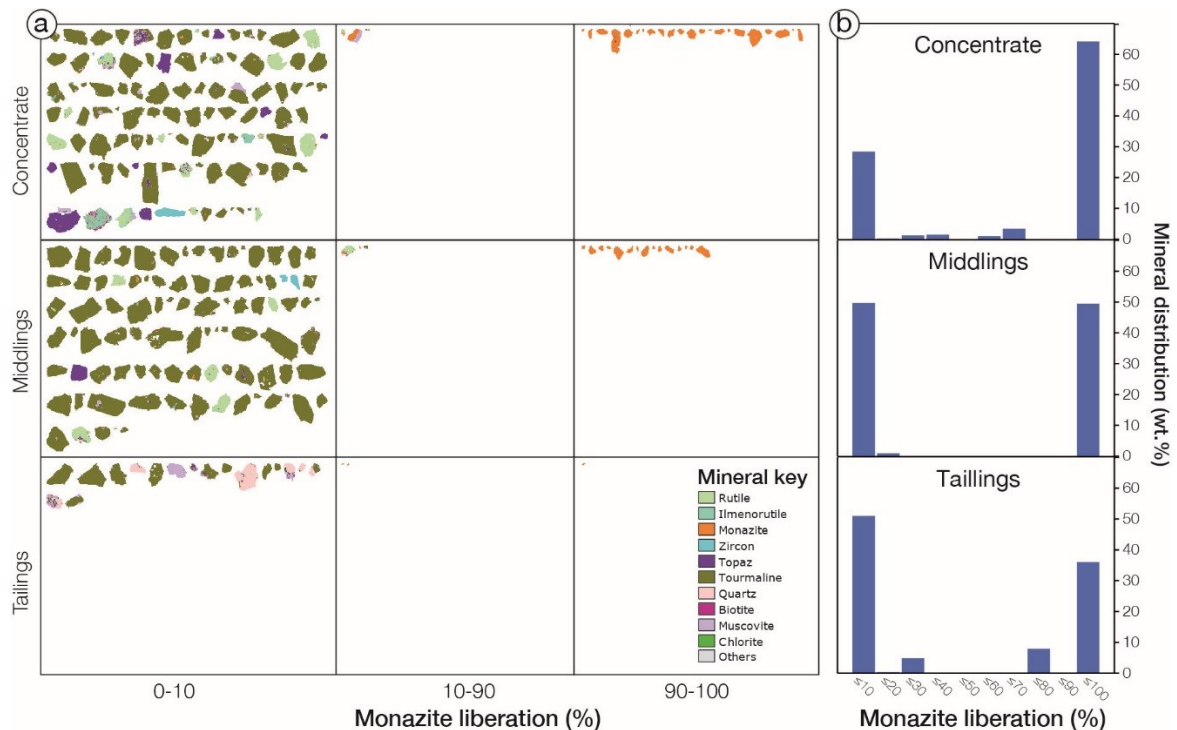

Figure 14~~13~~. Monazite liberation data for the gravity concentration products. (a) False colour QEMSCAN® monazite liberation grid showing typical examples of spiral concentrates (top), spiral middlings (middle) or spiral tailings (bottom), only the particles containing monazite are shown. (b) Average monazite liberation data for all gravity concentration products (spiral and shaking table) including concentrates (top), middlings (middle) and tailings (bottom).

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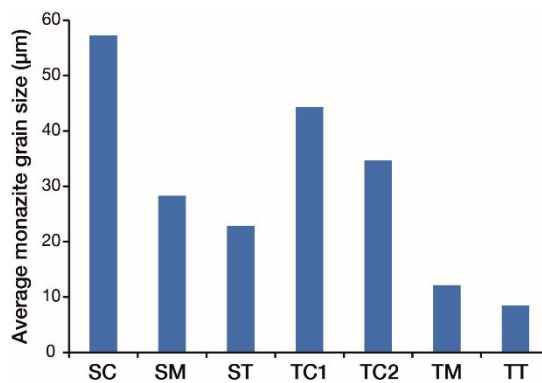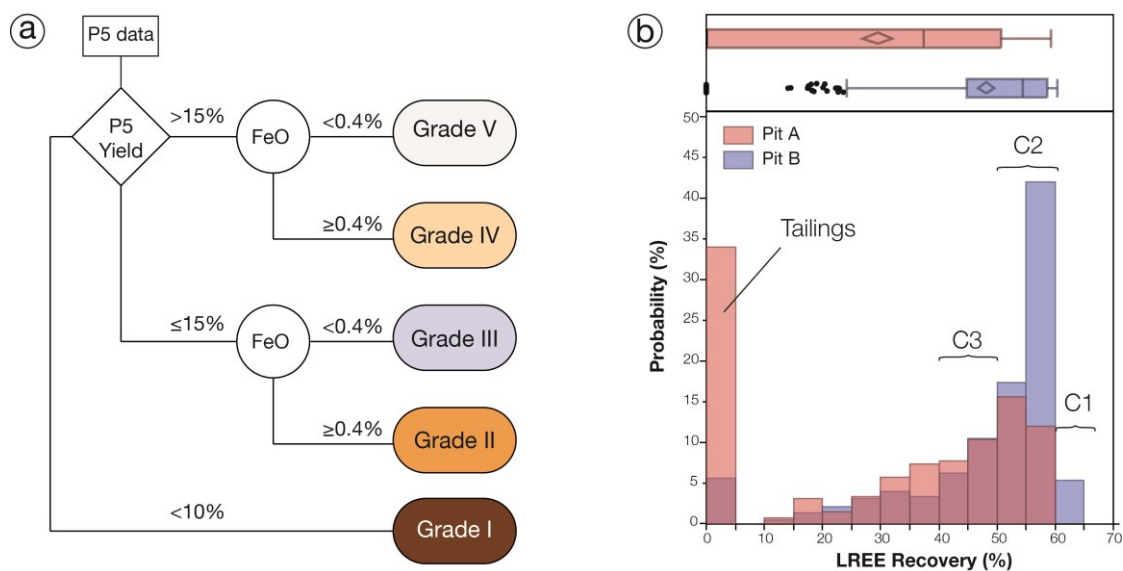
Figure 15~~14~~. Average monazite grain size in the gravity concentration products. Abbreviations are given in Figure 13~~Figure 12~~.
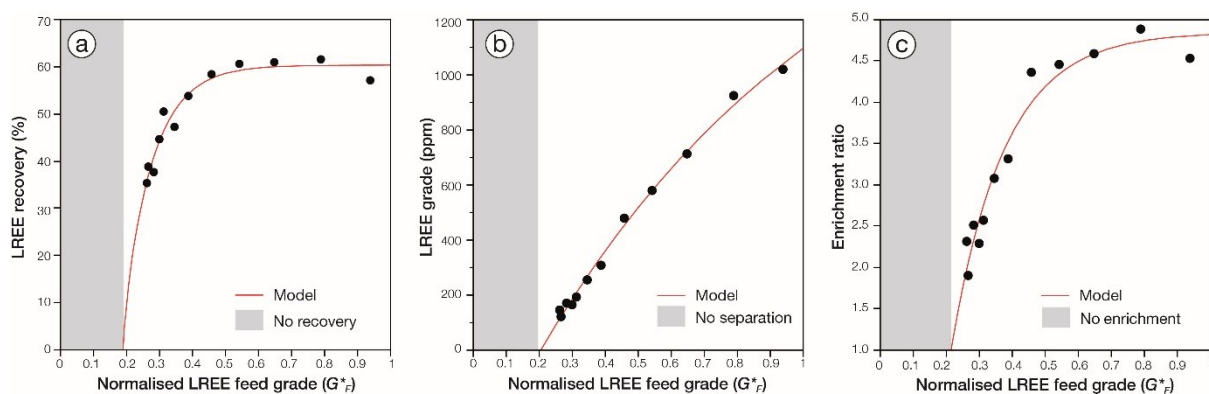
Figure 16~~15~~. Prediction of the performance of the rougher spiral concentration process for LREE as a function of normalised LREE feed grade. (a) LREE recovery, (b) LREE grade and (c) LREE enrichment ratio.

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1130 **Figure 17~~16~~.** Proposal for geometallurgical classification schemes. (a) Geometallurgical kaolin ore-type

1131 classification scheme using ~~R5~~P5 product yield and FeO content, threshold values are based on Negroni (2015).

1132 (b) Histograms of LREE recovery (R) for two kaolin deposits and corresponding box-plots that could be used to

1133 classify the ore into distinct categories (*i.e.*, C1: R$\geq$60%, C2: 60%>R$\geq$50%, etc.).

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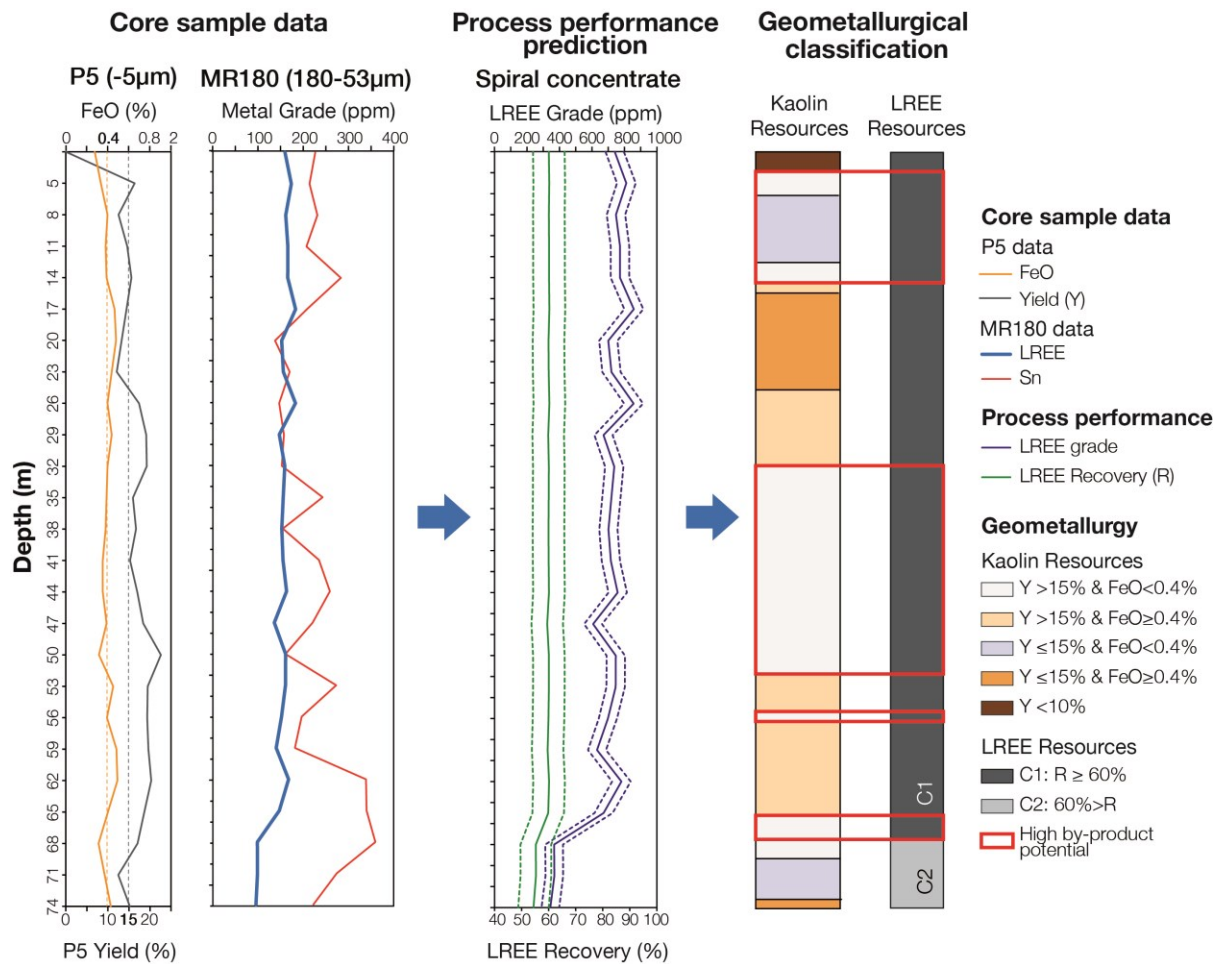
Figure 18~~17~~. LREE by-product potential evaluation using process performance prediction models and geometallurgical classifications schemes applied to drill cores. (Left) Core sample data related to the final kaolin product (~~R5~~P5) and to the spiral feed (~~GT53~~MR180), (middle) Application of spiral performance prediction models for LREE recovery, dashed lines represent 95% confidence intervals and (right) Geometallurgical classification of kaolin and LREE resources allowing to target the area of high potential for the recovery of LREE as by-product of kaolin.

TABLES

1144   **Table 1.** Mineralogy of the six major granite types of the St Austell rare-metal granite (Manning et al., 1996;

1145   Simons et al., 2016).

Granite Type	Major minerals	Minor minerals	Accessory minerals
Biotite Granite (BG)	Quartz, K-feldspar, plagioclase, biotite, muscovite	Tourmaline	Rutile, topaz, apatite, monazite, uraninite, zircon, cassiterite, cordierite
Lithium-mica Granite (LMG)	Quartz, K-feldspar, plagioclase (albite), lithium-mica (zinnwaldite, lepidolite)	Tourmaline, topaz	Apatite, monazite, zircon, rutile, fluorite
Tourmaline Granite (TG)	Quartz, K-feldspar, plagioclase, lithium-mica (zinnwaldite)	Tourmaline, topaz	Apatite, rutile
Fine-grained Tourmaline Granite (FGTG)	Quartz, K-feldspar, plagioclase (albite), lithium-mica (zinnwaldite)	Tourmaline, topaz	Monazite, apatite, zircon, rutile, arsenopyrite, cassiterite
Topaz Granite (TZG)	Quartz, K-feldspar (orthoclase), plagioclase, lithium-mica (zinnwaldite, lepidolite), muscovite	Topaz, fluorite, tourmaline	Apatite, amblygonite, zircon, columbite-tantalite, ilmenorutile, fluorite, uraninite

1146

1147   Table 2. Mean LREE, Sn, Nb, Ta and W contents in the output streams of the kaolin plants and the estimated clay

1148   matrix content obtained by metallurgical accounting for biotite, topaz and tourmaline granite.

Material	Mass Balance wt.%	LREE ppm	Sn ppm	Nb ppm	Ta ppm	W ppm
Biotite granite						
Matrix	100	79.1	68.7	39.9	11.4	30.8
Stone	4	50	38.3	26.2	7.6	15.7
Gravel	8	35.4	48.5	18.4	5.2	14.9
Sand	48	41.2	51.4	32.9	10	17.5
Micaceous residue	20	168.7	139.4	93.7	25.9	92.9
Product	20	103.7	53.5	14.2	3.6	10.3
Topaz granite						
Matrix	100	11.1	20.6	53.1	23.6	-
Stent	36	9	13.1	42	17.9	18
Sand	40.5	5.5	20.6	32.9	23	-
Micaceous residue	13.5	15.9	38.9	97.21	45.6	30.3
Product	10	34.3	21.4	38.84	15.8	15.5
Tourmaline granite						
Matrix	100	49	49.6	42.3	10.5	20.27
Stent	36	55	21	28	2	9.4
Sand	40.5	29.6	60.7	44.1	5	8
Micaceous residue	13.5	46.5	87.2	80.2	32	93
Product	10	109.9	56.8	55.1	34	10.9

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Table 3. Average mineralogical composition of the 26 ~~GT53~~MR180 core samples obtained by QEMSCAN® along with density data (Anthony et al., 2001).

Mineral	Average (wt %)	Std Dev.	Density (g.cm^{-3})
Quartz	48.47	12.16	2.60
Feldspar (orthoclase, microcline)	3.18	7.41	2.60
Kaolinite	1.12	1.03	2.60
Muscovite	21.99	10.15	2.82
Biotite (siderophyllite-annite)	1.72	1.28	3.09
Tourmaline (schorl-dravite)	20.97	10.69	3.12
Topaz	1.28	1.27	3.55
Ti-Oxides	1.07	0.48	4.25
Zircon	0.04	0.05	4.65
Monazite	0.04	0.03	5.15
Cassiterite	0.06	0.08	6.90
Others	0.03	0.21	-

Table 4. Multivariate analysis highlighting the main correlations between LREE grade and some mineralogical properties of the ~~GT53~~MR180 samples (2 outliers deleted). Significant correlations (≥ 0.65 or ($-0.65\geq$)) are highlighted in italic. Abbreviations: LREE: Calibrated LREE content/feed grade, NA: No association, A-Tour.: association with tourmaline, A-Micas: association with micas, A-Ti: association with Ti-oxides, Size: average grain size, %Locked: liberation $\leq 10\%$ and %Liberated: liberation $\geq 90\%$.

	LREE	NA	A-Tour.	A-Micas	A-Ti	Size	%Locked	%Liberated
LREE	**1.00**	0.07	-0.08	-0.38	0.02	*0.73*	-0.60	*0.65*
NA	0.07	**1.00**	-0.62	-0.33	-0.15	0.04	-0.08	0.05
A-Tour.	-0.08	-0.62	**1.00**	-0.17	-0.41	0.08	0.04	0.05
A-Micas	-0.38	-0.33	-0.17	**1.00**	*0.69*	-0.32	0.52	-0.56
A-Ti	0.02	-0.15	-0.41	*0.69*	**1.00**	0.09	0.09	-0.20
Size	*0.73*	0.04	0.08	-0.32	0.09	**1.00**	*-0.80*	*0.83*
%Locked	-0.60	-0.08	0.04	0.52	0.09	*-0.80*	**1.00**	*-0.96*
%Liberated	*0.65*	0.05	0.05	-0.56	-0.20	*0.83*	*-0.96*	**1.00**

Table 5. Values for the fitted parameters a, b and c in equation (2)~~(2)~~ of the models shown in Figure 16~~Figure 15~~.

Performance index\Coefficient	a	b	c	R²
Recovery	0.60	-4.43	-11.06	0.94
Grade	1921.17	-2386.91	-1.06	0.99
Enrichment	4.85	-14.51	-6.19	0.96