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Fenites associated with carbonatite complexes: A review

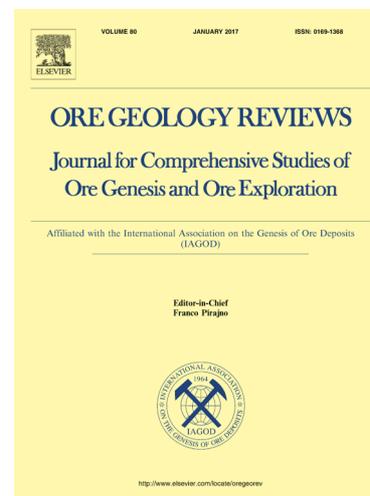
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## Fenites associated with carbonatite complexes: A review

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**Abstract**

Carbonatites and alkaline-silicate rocks are the most important sources of rare earth elements (REE) and niobium (Nb), both of which are metals imperative to technological advancement and associated with high risks of supply interruption. Cooling and crystallizing carbonatitic and alkaline melts expel multiple pulses of alkali-rich aqueous fluids which metasomatize the surrounding country rocks, forming fenites during a process called fenitization. These alkalis and volatiles are original constituents of the magma that are not recorded in the carbonatite rock, and therefore fenites should not be dismissed during the description of a carbonatite system. This paper reviews the existing literature, focusing on 17 worldwide carbonatite complexes whose attributes are used to discuss the main features and processes of fenitization. Although many attempts have been made in the literature to categorize and name

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fenites, it is recommended that the IUGS metamorphic nomenclature be used to describe predominant mineralogy and textures. Complexing anions greatly enhance the solubility of REE and Nb in these fenitizing fluids, mobilizing them into the surrounding country rock, and precipitating REE- and Nb-enriched micro-mineral assemblages. As such, fenites have significant potential to be used as an exploration tool to find mineralized intrusions in a similar way alteration patterns are used in other ore systems, such as porphyry copper deposits. Strong trends have been identified between the presence of more complex veining textures, mineralogy and brecciation in fenites with intermediate stage Nb-enriched and later stage REE-enriched magmas. However, compiling this evidence has also highlighted large gaps in the literature relating to fenitization. These need to be addressed before fenite can be used as a comprehensive and effective exploration tool.

*Keywords:* Fenite, Carbonatite, Metasomatism, REE, Niobium

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## 1. Introduction

Carbonatites and alkaline-silicate (hereafter referred to as alkaline) rocks are the most important sources of rare earth elements (REE) and niobium (Nb) (Wall, 2014; Goodenough et al., 2016), both high profile raw materials critical to technological advancement. Intrusions of both carbonatite and alkaline rocks are typically found in close spatial relationship with aureoles of high temperature metasomatically altered country rock, termed fenites. These alteration aureoles have great potential as exploration indicators. However, the vast majority of prior research has been focused on the intrusions themselves, with little attention paid to the potential for alteration to inform exploration. This contribution seeks to explore that potential by reviewing the current knowledge of fenite types, formation and relationship to economic mineralization.

13 The process of alteration (finitization) is generally viewed to result from mul-  
14 tiple pulses of alkali-rich fluid expelled from cooling and crystallizing carbonatitic  
15 or alkaline melt (Morogan, 1994; Le Bas, 2008). Fenites are typically composed of,  
16 and characterized by, K-feldspar, albite, alkali pyroxenes and/or alkali amphiboles  
17 (Zharikov et al., 2007). However, mineral assemblages are highly variable and de-  
18 pendent on a number of parameters such as protolith mineralogy, permeability and  
19 structure, in addition to fluid composition, temperature and pressure (Dawson, 1964;  
20 Heinrich, 1966; Le Bas, 1987; Platt, 1996). The secondary mineral assemblages asso-  
21 ciated with finitization are often visible in hand sample, whereas other effects such  
22 as the formation of turbidity in feldspars (e.g. Woolley, 1969; Heinrich and Moore,  
23 1969; Vartiainen and Woolley, 1976; Garson et al., 1984; Kresten and Morogan, 1986;  
24 Morogan and Woolley, 1988; Williams-Jones and Palmer, 2002; Le Bas, 2008) or the  
25 modification of primary mafic phases can be more subtle and only visible via op-  
26 tical microscopy. Vein networks often develop early on in the finitization process,  
27 facilitating the passage of fluids expelled from the melt through fractures and joints  
28 in the country rock, to form the outer fenite aureole (Woolley, 1969). The scale of  
29 finitization varies greatly from centimeters to several kilometers from the intrusive  
30 contact. Fenites are often associated with hydraulic fracturing and brecciation, ex-  
31 hibiting both temporal, spatial, mineralogical and chemical variations, resulting in  
32 both horizontal and vertical zoning.

33 Fluids derived from cooling alkaline magmas or carbonatite intrusions transport  
34 high quantities of alkalis and volatiles. These were originally constituents of the  
35 magma that are not retained in the carbonatite rock, therefore descriptions of in-  
36 trusions are incomplete without a description of their associated fenite. Finitizing  
37 fluids emanating from evolved and subsequently enriched magmas, form REE and  
38 Nb micro-mineral assemblages in the country rock (Elliott et al., 2016; Dowman

39 et al., 2017). These can provide information regarding their source magma and any  
40 associated mineral deposits, effectively acting as exploration indicators. Fenites are  
41 vertically and horizontally extensive, forming 3D aureoles around these intrusions  
42 and are therefore likely to be identified at a variety of different erosion levels even  
43 where source intrusions are not necessarily exposed. However, resource exploration  
44 has predominantly focused on intrusive units in alkaline and carbonatite complexes  
45 as these are the primary sources and hosts of REE and Nb deposits (Wall, 2014;  
46 Goodenough et al., 2016). There is a distinct lack of emphasis in the literature re-  
47 garding fenites, with only a few recent publications appreciating the importance of  
48 fenitizing fluids with regard to element mobility within the system (e.g. Arzamas-  
49 tev et al., 2011; Al Ani and Sarapää, 2013; Trofanenko et al., 2014; Dowman et al.,  
50 2017). As such, our comprehension of the relationship between intrusions, fluids  
51 and country rock is relatively restricted. This is in complete contrast to other ore  
52 systems, such as porphyry copper deposits, where alteration patterns and zones are  
53 a standard exploration tool (e.g. Hedenquist et al., 1998; Sillitoe, 2010).

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55 AlkCarb project, funded by the Horizon 2020 Research and Innovation Programme  
56 of the European Union. This project brings together a variety of multi-disciplinary  
57 academic and industrial partners from around the world to develop new geomodels to  
58 explore for 'high technology elements' in alkaline rocks and carbonatites. Although  
59 the locations of many alkaline and carbonatite complexes are well known (e.g. Wool-  
60 ley and Kjarsgaard, 2008b), these exploration models are required in order to better  
61 target unexposed economic deposits. Fenite enrichment is not typically sufficient to  
62 warrant exploitation; however, these rocks do have the potential to be used in the  
63 future as exploration tools to target critical metal resources.

64 *1.1. Rare earth elements and critical metals*

65 New technologies are utilizing an increasingly diverse array of elements in their  
66 manufacture. Amongst these are a sub-group of ‘critical metals’ (European Com-  
67 mission, 2017a,b), so defined as they are economically important metals with a sig-  
68 nificant risk of supply disruption. Carbonatite and alkaline rock-related deposits  
69 dominate the supply of REE and Nb, in addition to being sources of phosphate,  
70 fluorite, copper, titanium, platinum group elements and dimension stone. Alkaline  
71 and ultramafic rocks also provide vermiculite, nepheline and are potential sources of  
72 scandium and tantalum (e.g. Woolley and Kjarsgaard, 2008b; Wall, 2014; Mitchell,  
73 2015; Goodenough et al., 2016).

74 The term rare earth elements (REE) is used in this manuscript to represent the  
75 lanthanide series elements lanthanum (La) to lutetium (Lu), plus scandium (Sc) and  
76 yttrium (Y). Although the division between light and heavy rare earth elements  
77 is variably defined, the European Commission (2014) subdivides these into light  
78 REE (LREE) consisting of La-Sm, and the heavy REE (HREE) consisting of Eu-Lu  
79 including Y. These metals have advantageous paramagnetic, mechanical and spectral  
80 properties (Gupta and Krishnamurthy, 2005; British Geological Survey, 2011; Wall,  
81 2014), making them useful in a wide range of industrial processes.

82 The vast majority of REE production occurs in China (>95 %) (British Geologi-  
83 cal Survey, 2017; European Commission, 2017a), with only three other major mining  
84 operations globally. These consist of a weathered carbonatite at Mt Weld, W. Aus-  
85 tralia (e.g. Hoatson et al., 2011; Pirajno, 2015); a nepheline syenite at Lovozero,  
86 Kola Peninsula, Russia (e.g. Kogarko et al., 2010; Arzamastev et al., 2011; Wall,  
87 2014); and as a by-product of Ti exploitation from mineral sand at Orissa, India  
88 (e.g. Gupta and Krishnamurthy, 2005; British Geological Survey, 2011). Currently  
89 Nb production is dominated by the carbonatite-related laterite deposits at Araxá and

90 Catalão II, Brazil (e.g. Mitchell, 2015); carbonatite at St Honoré, Canada (Mitchell,  
91 2015); and the nepheline syenite at Lovozero, Russia (Linnen et al., 2014). However,  
92 prospective deposits for both REE and Nb exist worldwide (see Fig. 1). Therefore,  
93 to develop a reliable and sustainable supply of these critical metals, complexes and  
94 deposits within politically stable countries must be explored and developed. Fenites  
95 do not contain exploitable quantities of critical metals, but do have great potential  
96 to be used as an exploration tool. Of the commodities listed above, only fluorite,  
97 vermiculite, phosphate and dimension stone have previously been mined from fenite.

### 98 *1.2. Classification*

99 Fenites are described as a metasomatic family by the IUGS (Zharikov et al., 2007),  
100 formed at high temperatures and characterized by a certain suite of minerals, but  
101 no attempt for further subdivision or more detailed nomenclature has been made by  
102 the IUGS. Fenite terms originally used by Brögger (1921) to describe the Fen type  
103 locality, Norway are listed in the IUGS classification of igneous rocks (Le Maitre  
104 et al., 2002). However, names such as tveitåsite and ringite are local, specific to Fen,  
105 and not commonly used elsewhere. Within fenites and their associated intrusive  
106 rocks, other forms of localized metasomatic processes are also observed, for example:  
107 autometasomatism in the peripheral parts of intrusions, contact metasomatism at the  
108 intrusion-country rock interface, and near-vein metasomatism forming symmetrical  
109 zonation on either side of a vein (Zharikov et al., 2007).

110 The classic definition of the term fenitization by Brögger (1921), involves the  
111 removal of silica from the protolith. However, during the metasomatism of undersat-  
112 urated igneous rocks (e.g. Nurlybaev, 1973; Viladkar, 2015) there is no loss of silica,  
113 and often some gain relative to the protolith. As such, Bardina and Popov (1994)  
114 argued that fenitization is marked by an increase in alkali content ( $\text{Na}_2\text{O} + \text{K}_2\text{O}$ ),

115 irrespective of silica content or mobility.

116 Many endeavours have been made in the literature to subdivide fenites further  
117 into categories, dating back to von Eckermann (1948). For example, Kresten (1988)  
118 classified fenites as contact, aureole or veined depending on their spatial relationship  
119 to the source intrusion; Verwoerd (1966) first attempted to use the ratio of Na and  
120 K to classify fenites as sodic, potassic or intermediate, terms still used widely in  
121 the literature; and Morogan (1994) used a continuous scale of fenitization intensity  
122 to classify fenites as low, medium or high-grade. Russian literature names rocks  
123 that retain some of the original texture or protolith mineralogy, using the modifier  
124 “fenitized” added before the protolith name (e.g. Arzamastev et al., 2011; Kozlov  
125 and Arzamastsev, 2015). Additionally, the prefix “apo-” is used to indicate the na-  
126 ture of this protolith following the recommendations of the IUGS for metamorphic  
127 rocks (Fettes and Desmons, 2007) e.g. apogneiss fenite. However, many of these  
128 classification schemes do not provide much information to the reader or have ge-  
129 netic connotations, leading to a history of baffling nomenclature, but reflecting the  
130 complexity of the fenitization process. As such, we recommend that a nomenclature  
131 classification scheme similar to that used by the IUGS for metamorphic rocks (Fettes  
132 and Desmons, 2007) be used to name fenites, by using structural and mineralogi-  
133 cal modifiers e.g. veined aegirine-albite fenite; brecciated orthoclase fenite. Here  
134 it is worth mentioning the collective term ‘alkali metasomatites ’ used in Russian  
135 literature to denote rocks mineralogically similar to fenites, but generated by fluids  
136 of unknown provenance. These rocks are named on the basis of their predominant  
137 mineral, such as albitite or aegirinite (e.g. Es’kova, 1976; Ugryumov and Dvornik,  
138 1984).

139 This manuscript follows the carbonatite nomenclature recommended by the IUGS,  
140 as outlined in Le Maitre et al. (2002), using the prefix of most abundant carbonate

141 mineral present e.g. calcite carbonatite, dolomite carbonatite, ankerite carbonatite.  
142 Names used in the literature have been converted to this scheme; however, where  
143 iron-rich carbonate minerals have not been specifically identified, the term ferroc-  
144 bonatite is used.

### 145 *1.3. Geological setting and models*

146 Carbonatites are an extraordinary group of igneous rocks containing >50 % car-  
147 bonate minerals (Heinrich, 1966; Le Bas, 1981; Woolley and Kempe, 1989) including  
148 calcite, dolomite, ankerite, siderite and magnesite (Woolley and Kempe, 1989). The  
149 term alkaline encompasses a wide variety of rock types and mineralogy (Fitton and  
150 Upton, 1987; Le Bas, 1987) which cannot all be covered within the scope of this  
151 paper. No precise term has been agreed upon to define alkaline rocks, however 'alka-  
152 line' implies a deficiency in silica and aluminium relative to the alkali elements (Na  
153 and K) (Fitton and Upton, 1987; Leelanandam, 1989; Mitchell, 1996). Although it  
154 is recognized that both carbonatites and alkaline-silicate intrusions are capable of  
155 forming fenites, the focus of this paper is carbonatite-related fenitization, and there-  
156 fore alkaline-related fenitization is referred to separately (see Section 5.3).

157  
158 Intracontinental geological settings and rift valleys often host both alkali and car-  
159 bonatitic magmatism, however carbonatites are also found in relation to subduction  
160 processes (Fitton and Upton, 1987). An in depth discussion of carbonatite magma  
161 genesis is beyond the scope of this paper, therefore the reader is recommended the fol-  
162 lowing references for more information. Two opposing theories exist for the origin of  
163 carbonatite magmas. The first suggests that carbonatite magmas are produced from  
164 parent alkaline magmas via a complex combination of liquid immiscibility and/or  
165 crystal fractionation processes (e.g. Le Bas, 1981; Treiman and Essene, 1985; Ander-

166 sen, 1989; Kjarsgaard and Hamilton, 1989; Ray and Shukla, 2004). This is supported  
167 by the fact that 74 % of known carbonatite occurrences are associated with alka-  
168 line rocks (Heinrich, 1966; Woolley, 2003; Le Bas, 2008; Woolley and Kjarsgaard,  
169 2008b), such as nepheline syenites, ijolites and pyroxenites that are considered to  
170 have formed from the immiscible silicate magmas (Le Bas, 1987). The second theory  
171 indicates that some carbonatites are derived directly from the mantle, and therefore  
172 sourced from depths greater than the carbonate solidus at  $\sim 70$  km, below which  
173 dolomite-bearing peridotite can melt to produce carbonatitic magmas (e.g. Bailey,  
174 1989; Harmer and Gittins, 1998; Chakhmouradian et al., 2009).

175 Conceptual models of alkaline and carbonatite magmatic systems have not ad-  
176 vanced dramatically since that proposed by Le Bas (1977) (see Fig. 2). Recent  
177 research driven by resource exploration has progressed our understanding of these  
178 systems and the model is now outdated. The model lacks any emphasis on the forma-  
179 tion and location of potential ore deposits; details of fenite formation and characteris-  
180 tics such as brecciation, veining and zonation; in addition to the role of hydrothermal  
181 fluids in and around the complex. With very few exceptions (e.g. Arzamastsev et al.,  
182 2000; Mikhailova et al., 2016), there is little information on the vertical extent of  
183 individual rock series and their association mineralization (Verplanck et al., 2014).

#### 184 *1.4. Fen, Norway: Fenite type locality*

185 The type locality of fenitization was first described by Brögger (1921) in his  
186 memoir on the Fen carbonatite complex, Norway. Since the initial studies of the  
187 1920s, further investigation of this complex has advanced our understanding of its  
188 petrography (Sæther, 1958; Marien et al., 2017), mineral chemistry (Kresten and Mo-  
189 rogan, 1986; Andersen, 1989), whole-rock geochemistry (Kresten, 1988), and isotopic  
190 signature (Verschure and Maijer, 2005).

191 The Fen Complex (see Fig. 3) consists of a roughly circular 5 km<sup>2</sup> central core  
192 of Neoproterozoic (578 ± 24 Ma) calcite carbonatite and alkaline intrusions of the  
193 melteigite-ijolite-urtite series with subordinate nepheline syenite, that were emplaced  
194 into Mesoproterozoic granitic gneiss country rock (Brögger, 1921; Kresten and Mo-  
195 rogan, 1986; Andersen, 1989; Dahlgren, 1994; Verschure and Maijer, 2005). The  
196 Fen Complex, which has been historically mined for Nb and Fe, lies at the center  
197 of a larger area defined as the Fen Province, which incorporates many small satel-  
198 lite intrusions of dolomite and ferrocarnatite extending over 1500 km<sup>2</sup> (Dahlgren,  
199 1987, 1994). Intrusion of these carbonatite magmas and preceding alkaline rocks are  
200 associated with at least two brecciation events (Kresten and Morogan, 1986) that  
201 facilitated at least two stages of fenitization. The first event created a fenite aure-  
202 ole 700 m wide, and the second >1.5 km wide (Verschure and Maijer, 2005), much  
203 greater than the 200 m previously described by Brögger (1921) which was restricted  
204 to a narrow zone along the intrusion-country rock contacts.

205 Brögger (1921) argued that the fenite at Fen was the product of metasomatism  
206 adjacent to the ijolite-melteigite intrusions during a process he named “Fenitisierung”  
207 or fenitization. However, further study has shown that fenitization is the product  
208 of fluids originating from both the alkaline and carbonatitic intrusions (Kresten and  
209 Morogan, 1986). Based on microscope observations of thin sections, Brögger (1921)  
210 described the progressive metasomatism from the unaltered granite, through to the  
211 ijolite-melteigite contact. This involved the alteration of an original country rock  
212 mineral assemblage of quartz, K-feldspar, oligoclase, biotite and hornblende to an  
213 albite and aegirine-rich fenite near the contact. The stages involved in this progres-  
214 sive metasomatism are shown in Figure 4.

## 215 **2. Fenite Types and Formation**

216 There are many parameters that control the characteristics of a fenite; therefore  
217 it cannot be assumed that a certain protolith will produce a distinct fenite min-  
218 eral assemblage. The complexity of the process is reflected in the diverse range of  
219 nomenclature used in the literature.

220 Protolith characteristics such as mineralogy, texture, structure, porosity and per-  
221 meability have a large bearing on the extent of fenitization. Chemical buffering by  
222 the country rock determines the intensity and extent of fenitization which can vary  
223 greatly from centimeters to several kilometers. Intense fenitization occurs where  
224 there is a large chemical gradient between fenitizing fluids and country rock, such as  
225 granite, granitic gneisses or meta-sediments. In contrast, country rocks dominated by  
226 quartz or carbonate lithologies including sandstone, marbles and limestones, typically  
227 display little evidence of fenitization, reflecting their low degree of reactivity. This  
228 relationship is reflected by the scale of fenitization surrounding carbonatite intruded  
229 into protoliths with varying chemical gradients. For example, fenite in sandstone  
230 surrounding the Amba Dongar carbonatite, India, is <300 m thick (Doroshkevich  
231 et al., 2009), compared to fenites of the Shawa and Dorowa complexes, Zimbabwe,  
232 both of which extend >2 km diameter into the granitic gneiss country rock (Johnson,  
233 1966). Feldspar-rich fenites at Bayan Obo, China only extend 2–4 m into Middle  
234 Proterozoic meta-sandstones and marbles (Le Bas, 2008).

235 The fenite mineral assemblage and chemistry is also dependent on the protolith,  
236 but additionally affected by the composition, temperature and pressure of the feni-  
237 tizing fluid (Dawson, 1964; Heinrich, 1966; Le Bas, 1987; Platt, 1996). Pyroxene and  
238 amphibole chemistry is particularly sensitive to the fenitizing fluid characteristics  
239 (Sutherland, 1969). Mineralogy is also dependent on the intensity of metasomatism;

240 however, this rarely decreases linearly away from the contact. Late-stage intrusions  
241 in the fenite aureole, such as the Jammi dykes in the Sokli carbonatite complex,  
242 Finland (Al Ani and Sarapää, 2009, 2013; Sarapää et al., 2013, 2015), cause high in-  
243 tensity fenitization away from the central intrusion. Additionally, fluids utilize highly  
244 permeable planes of weakness (Ortoleva et al., 1987), causing preferential fenitization  
245 leading to asymmetrical fenite aureoles (e.g. Fen complex, see Fig. 3).

### 246 *2.1. Veined vs Pervasive Fenites*

247 Cross-cutting vein textures and zoned apatite crystals (see Fig. 5A and 5B) ob-  
248 served in fenite aureoles indicate that alkali metasomatism is a multi-phase process  
249 (Le Bas, 2008). The distribution of the metasomatic (or fenite) parageneses can  
250 form two end member macrotextures, either pervasive throughout the country rock,  
251 or localized adjacent to veins that follow pre-existing structural weaknesses, such  
252 as fractures (e.g. Kresten, 1988; Morogan and Woolley, 1988; Sindern and Kramm,  
253 2000; Pirajno et al., 2014). Since fenite mineral assemblages result from the reaction  
254 between fenitizing fluids and the surrounding country rock, these macrotextures are  
255 a response to the distribution of fluids and the dynamics of fluid flow in the host  
256 rock.

257 Fenitizing fluids can flow either along grain boundaries and mineral cleavage  
258 planes (porous flow), and/or within dilated fractures. During porous flow, fluid is  
259 able to interact with a larger surface area of rock than fluid confined to fractures.  
260 This enables pervasive fenitization, causing metasomatic minerals to be distributed  
261 throughout the country-rock (see Fig. 5C and 6A). Pervasive fenites are commonly  
262 observed in the immediate vicinity of carbonatite and alkaline intrusions and extend  
263 radially away from the fenitizing fluid source (Kresten, 1988). Pervasive fenites are  
264 typically divided into zones on the basis of mineralogical and textural changes (e.g.

265 Rubie, 1982; Morogan and Woolley, 1988). The intensity of fenitization typically de-  
266 creases with distance from the source intrusion. This most likely reflects a reduction  
267 in the fluid-rock ratio and changing fluid-rock chemistry gradient with distance. One  
268 example of this process is the Okorusu fenite, Namibia, which displays undulating or  
269 scalloped alteration fronts that formed during the permeation of fenitizing fluids via  
270 porous flow (see Fig. 5E). This fluid forms a reaction front that advances irregularly  
271 through the greywacke country-rock due to differences in porosity and fluid-mineral  
272 reactions across the front (Ortoleva et al., 1987).

273 Fenitizing fluids may exploit pre-existing structural weaknesses in the host rock  
274 or cause the propagation of new fractures by fluid overpressure or volume changes  
275 related to metasomatic mineral reactions (Skelton et al., 2007; Sachau et al., 2015).  
276 During fracture-hosted flow, only a small fraction of the fenitizing fluids can metaso-  
277 matize the country rock adjacent to the host fractures (see Fig. 5D and 6B), forming  
278 veined fenites. The volume of country rock affected by fluid-rock interaction during  
279 fracture-confined fluid flow is limited relative to diffuse porous flow (see Fig. 6).  
280 Fluid hosted in fractures are essentially chemically isolated from the surrounding  
281 country rock and will therefore retain their alkali-rich chemistry over greater dis-  
282 tances. Fractures act as effective conduits, efficiently facilitating transport of fluids  
283 to the outer fenite aureole. The formation of zoning during metasomatic processes  
284 is common, and can result from changes in the chemistry of a single fluid infiltrating  
285 the country rock (e.g. Korzhinskii, 1968; Brady, 1977; Meinert, 1998; Winter, 2001).  
286 However, individual fenite veins can show multiple zones of differing minerals with  
287 clearly defined contacts, mirrored either side of a fracture. These patterns suggest  
288 precipitation from different generations of successive fluids, that repeatedly utilized  
289 the same fracture system during the course of fenitization (see Fig. 5F).

290

291 Fenites of greater complexity display a combination of veined and pervasive fen-  
292 itization, indicating that they have experienced both porous and channelised fluid  
293 flow at various times during evolution of the complex. Shifts in the importance of  
294 flow mechanism most likely reflect adjustments to fenitizing fluid pressure, confining  
295 pressure and porosity/permeability characteristics of the fenite. Subtle changes to  
296 these parameters may result from rock-fluid chemical reactions or mechanical failures  
297 such as fracturing.

298 The dynamics of fluid flow during fenitization are poorly understood. However,  
299 analogies with other fluid-rich geological systems, such as metasomatized porphyry  
300 stockworks (e.g. Ortoleva et al., 1987; Robb, 2005), can be drawn to demonstrate  
301 that the passage and pathways of fluid flow have fundamental consequences for el-  
302 ement mobilization and deposition. One example is the intrusion of the Cornubian  
303 granite batholith, England, which was accompanied by the formation of vein net-  
304 works channelling hot aqueous fluids and vapours expelled from an overpressurized  
305 magma reservoir. The resulting tin-tungsten mineral veins are bound by greisen  
306 wall-rock alteration (e.g. Jackson et al., 1989; Dominy et al., 1995) during a process  
307 broadly similar to metasomatism adjacent to fenite veins. Greisenization is a post-  
308 emplacement metasomatic process leading to remobilization of Si and Al within  
309 the granitoid, which can be preceded or followed by potassic or sodic alteration  
310 (Stemprok, 1987). Alteration in the Cornubian Orefield is categorized into pervasive  
311 and joint-controlled greisens (Stemprok, 1987), forming similar alteration patterns  
312 to those observed in fenites (see Fig. 6). Pervasive greisenization is observed at  
313 Cameron Quarry, St. Agnes, where it caused complete alteration of the granite por-  
314 phyry (Hosking and Camm, 1985). Other sites display limited alteration adjacent to  
315 joint-greisens such as Wheal Prosper, St Austell (Bennett et al., 1981), analogous to  
316 veined fenites.

317 The formation of skarns is another analogy for metasomatism in fenites. Skarns  
318 are metasomatic rocks that typically form at the contact between a silicate intrusion  
319 and a carbonate country rock, forming zones of differing modal mineralogy extending  
320 away from the contact, similar to a fenite. Typical mineralogy consists of Ca-Mg-Fe-  
321 Mn silicates such as forsterite, diopside, phlogopite and garnet (Kwak, 1987; Meinert,  
322 1992; Zharikov et al., 2007). Veins are a common feature of both fenite aureoles and  
323 skarns, resulting from metasomatic fluids confined in bodies or pipes cross-cutting  
324 the intrusion or country rock (Zharikov et al., 2007), the size of which is dependent  
325 on country rock reactivity and fracture permeability (Bussell et al., 1990).

## 326 *2.2. Breccias*

327 Carbonatite emplacement is often accompanied by widespread brecciation and  
328 fracturing of the basement rock (Heinrich, 1966; Tuttle and Gittins, 1966; Le Bas,  
329 1977; Rubie and Gunter, 1983). Swinden and Hall (2012) identified two predominant  
330 types based upon their study of Songwe Hill, Malawi. The first type is a feldspathic  
331 breccia (see Fig. 7A) consisting of alkali feldspar-rich fragments  $\pm$  carbonatite clasts  
332 in a matrix of alkali feldspar, carbonate and oxides with varying proportions of  
333 quartz due to fenite comminution during brecciation. The second is a carbonatite-  
334 rich breccia consisting of carbonatite and subordinate fenite clasts in a carbonate-  
335 rich matrix. The majority of breccias directly related to carbonatite complexes can  
336 be fitted into either of these two categories with varying proportions of clast types  
337 and protoliths. Brecciation is most commonly seen at the carbonatite-country rock  
338 contact, typically consisting of feldspathic breccias intruded by later carbonatitic  
339 dykes or veins (see Fig. 7B). Examples include Sokli (Vartiainen and Woolley, 1976;  
340 Notholt, 1979), Fen (Verschure and Maijer, 2005), Kangankunde and Chilwa Island,  
341 Malawi (Woolley, 1969); Homa Mountain, Kenya (Clarke and Roberts, 1986); Dicker

342 Willem, Namibia (Reid and Cooper, 1992); Toror Hills, Uganda (Sutherland, 1965b)  
343 and Okorusu, Namibia (Bühn et al., 2002). The brecciated transition zone between  
344 fenite and carbonatite at Sokli is highlighted by a reduction in gravity and low seismic  
345 velocities on geophysical surveys (Vartiainen and Paarma, 1979).

346 The presence of breccias at many carbonatite complexes attests to the explosive  
347 release of fluids and volatiles from an evolving magma below (Verplanck et al., 2014;  
348 Croll et al., 2015). At higher fluid pressures, masses of comminuted fenite and/or car-  
349 bonatitic material can explosively breach the surface to form extrusive tuffs (Le Bas,  
350 2008), which was experimentally demonstrated by Lorenz et al. (1991). Explosive  
351 release of these fluids may explain observations of tuffs and lapilli tuffs at Kruid-  
352 fontein, South Africa and Rangwa, Kenya (Le Bas, 2008). Alternatively, Kresten  
353 (1988) interpreted brecciation surrounding carbonatites as the result of volume in-  
354 crease during the fenitization process. The latter mechanism may also be responsible  
355 for the dilation of numerous fractures which host quartz veins.

356 The timing of brecciation around carbonatite complexes is more subjective and  
357 possibly site-specific. Where fully fenitized breccia clasts are entrained in carbon-  
358 atite, it can be hard to determine whether fenitization occurred prior to, or during  
359 their incorporation. Previous sections have presented evidence that carbonatite em-  
360 placement and the release of fluids/volatiles is a multi-stage process. Evidence indi-  
361 cating multiple stages of brecciation and feldspathization between intrusion events  
362 includes, for example, feldspathized breccia blocks incorporated in carbonatite intru-  
363 sions at Chilwa Island (Woolley, 1969) and Sokli (Vartiainen and Woolley, 1976) (see  
364 Fig. 7C). Brecciation often occurs prior to late-stage carbonatite intrusions (see Fig.  
365 7B), as manifested by the *in-situ* injection of carbonatite into previously shattered  
366 country rock at Mountain Pass, USA (Castor, 2008); the intrusion of carbonatite  
367 dykes and veins into intensely brecciated and fractured fenites at Sokli (Vartiainen

368 and Paarma, 1979); and the intrusion of a feldspathic breccia vent by later carbon-  
369 atite sheets at Songwe Hill (Garson, 1965).

370 Brecciation accompanying igneous activity is not exclusive to carbonatites, and  
371 breccias often occur surrounding alkaline complexes. These breccias are often inter-  
372 preted as hydrothermal products resulting from the escape of volatiles from magma  
373 during boiling stages, leading to intense hydraulic fracturing (Burnham, 1985; Pi-  
374 rajno, 2015). Diatremes are often filled with highly permeable and porous vent  
375 breccias and tuffs (Afanasyev et al., 2014; Elliott et al., 2015) that provide efficient  
376 intrusive pathways and hosts for later carbonatite magmas. This process can form  
377 REE-enriched (up to ~3 wt.% TREO) carbonatite stockworks such as the Bull Hill  
378 phonolitic diatreme at Bear Lodge, USA (Moore et al., 2015) and diatreme breccias  
379 at Kaiserstuhl, Germany (Hornig-Kjarsgaard, 1998).

### 380 *2.3. Sodic vs Potassic Fenites*

381 The whole-rock ratio of Na to K is used to classify fenites as sodic, intermediate  
382 or potassic (Verwoerd, 1966). Historically, sodic fenites were attributed to alkali  
383 magmatism such as the formation of ijolites, and carbonatite-related fenitization  
384 was thought to be potassic (Morogan, 1994). However, this is no longer thought  
385 to be the case as many carbonatites are associated with both potassic and sodic  
386 fenites (Woolley, 1982; Le Bas, 2008). The type of fenite formed is not dependent  
387 on carbonatite composition, with sodic and potassic fenites being found adjacent to  
388 intrusions of either calcite or dolomite carbonatite (Le Bas, 2008).

389 The spatial variation between potassic and sodic fenites is distinct at a number of  
390 carbonatite complexes globally. Le Bas (1981) and Woolley (1982) proposed a verti-  
391 cal zonation in fenite chemistry, with more sodic compositions hypothesized to occur  
392 at depth and potassic fenites at shallower crustal levels. This model could explain

393 the presence of sodic fenites around older complexes due to deeper cross sections  
394 being exposed by prolonged erosion (Le Bas, 2008). Very few carbonatite complexes  
395 are sufficiently well-exposed to display this vertical zonation pattern (e.g. Amba  
396 Dongar, India: Doroshkevich et al., 2009), but the majority of carbonatite complexes  
397 show horizontal variations in fenite chemistry. Carbonatites of the Chilwa Province,  
398 Malawi, typically exhibit an outer sodic fenite and inner potassic aureole associated  
399 with intense brecciation (Woolley, 1982), also reported at Wicheeda, Canada (Tro-  
400 fanenko et al., 2014). In addition to spatial controls on fenitization, observations  
401 such as cross-cutting relationships at many complexes such as Bayan Obo (Le Bas,  
402 2008); Chilwa Province (Woolley, 1969, 1982); and Chipman Lake, Canada (Platt  
403 and Woolley, 1990), suggest there is also a temporal control on fenitization with an  
404 early sodic phase typically overprinted by later potassic fenitization.

405  
406 The alkali content in a carbonatite-derived fluid varies dramatically, which is  
407 reflected in the extreme geochemical variations among published fenite compositions  
408 (see Fig. 8). The few available estimates range from up to 3 wt.% dissolved  $\text{Na}_2\text{CO}_3$   
409 in expelled hydrothermal fluids at Oldoinyo Lengai, Tanzania (Carmody, 2012) to up  
410 to 30 wt.%  $\text{Na}_2\text{O} + \text{K}_2\text{O}$  in bulk fluids from inclusions at the Okorusu and Kalkfeld  
411 carbonatites, Namibia (Bühn et al., 2002). This variation in alkali concentration may  
412 result from a difference in the depth of carbonatite magma generation and complexity  
413 of magma evolution. Woolley and Church (2005) suggest that carbonatite magmas  
414 generated by high-level fractionation and immiscibility processes in a peralkaline  
415 crustal magma chamber will have a higher alkali content than those that move rapidly  
416 from a mantle source to the shallow crust.

417 Fenitization is traditionally considered to involve the removal of silica (Brögger,  
418 1921) and the addition of alkalis ( $\text{Na}_2\text{O} + \text{K}_2\text{O}$ ) (Bardina and Popov, 1994), which

419 can clearly be seen illustrated by the general fenitization trends in Figure 8. The  
420 majority of complexes show a substantial increase in alkalis between 0.5–10 wt.% and  
421 a decrease in silica between 0.5–29 wt.%. This trend is most clearly demonstrated  
422 in fenites of the type locality (Fen) (Fig. 8B); Sokli, Finland (Fig. 8C); Silai Patti,  
423 Pakistan (Fig. 5D); and Newania, India (Fig. 8F), all of which replaced protoliths of  
424 granite or geochemically similar metamorphic rocks such as gneiss or schist. In the  
425 majority of fenites, alkalis are enriched in the country rock during fenitization due  
426 to a large compositional gradient between fluids and protolith. Protoliths with a low  
427 initial silica content can experience enrichment in silica during fenitization, such as  
428 the 23 wt.% increase observed in the marble at Lac a la Perdrix, Canada (Fig. 8E).  
429 However, this is not the case where country rock has an initially high alkali content.  
430 For example, aplitic granite, consisting solely of feldspar and quartz (Le Maitre et al.,  
431 2002), loses up to 2.3 wt.% alkalis during fenitization at Meech Lake, Canada (Fig.  
432 8D). Fenitization of nephelinite can cause minor alkali enrichment or depletion (up  
433 to 2.3 wt.%) such as that seen at Amba Dongar (Fig. 8D) and Koga, Pakistan (Fig.  
434 8F).

435 Spatial and temporal fenite variations are thought to be dependent on the tem-  
436 perature, pressure and CO<sub>2</sub> content of the fenitizing fluid (Rubie and Gunter, 1983).  
437 Potassium is considered to be more mobile than Na at lower temperatures and there-  
438 fore transported by fluids to shallower depths, to form shallower potassic fenites at  
439 lower temperatures. Na is preferentially lost from fluids at deeper crustal levels  
440 whilst temperatures are still >600 °C, forming an earlier, deeper sodic fenite (Hein-  
441 rich and Moore, 1969; Le Bas, 1981; Rubie and Gunter, 1983). This is supported by  
442 the common association of brecciation and potassic fenitization, suggesting that the  
443 latter occurred during volatile release and hydraulic fracturing. These processes in-  
444 dicate shallow depth, such as the roof of a magma chamber, and account for vertical

445 chemical zonation of the fenite aureole (Le Bas, 2008). Another model, proposed by  
446 Woolley (1982), involves a high initial Na<sub>2</sub>O:K<sub>2</sub>O ratio in the carbonatite intrusion,  
447 causing initial expulsion of Na to form an early sodic fenite. Continued release of  
448 Na-rich fluids subsequently enriches the melt in K, causing later potassic fenitization  
449 favored at lower temperatures of <450 °C determined by Rubie and Gunter (1983).

450 Processes that cause spatial and temporal segregation of Na and K-rich fluids are  
451 unlikely to conform to the same model at every alkali complex, and many fenites  
452 do not fit the patterns discussed above at all (Heinrich, 1985, – “there are infi-  
453 nite variations on a fenite theme”). Carbonatites surrounded by exclusively sodic  
454 or potassic fenites could be explained by different depths of erosion; for example,  
455 exclusively sodic fenitization is observed at the Newania complex (Viladkar and  
456 Pawaskar, 1989), whereas carbonatites at Rufunsa, Zambia (Woolley, 1969) and  
457 Bukusu, Uganda (Vartiainen and Woolley, 1976) lack sodic fenitization. However,  
458 the presence of K-feldspar crystals rimmed by albite at the Ruri Hills carbonatite,  
459 Kenya, and Koga, as well as K-feldspar rich fenite cut by late albite veins at Bayan  
460 Obo (Le Bas, 2008), suggest that sodic fenitization does not always occur prior to  
461 potassic.

### 462 2.3.1. Sodic fenites

463 One of the most conspicuous features of sodic fenites are the abundant green veins  
464 and veinlets of sodic pyroxenes and amphiboles cross-cutting alkali feldspar (see. Fig.  
465 7B and 7F). Quartz and minor minerals such as calcite, fluorite and apatite are also  
466 observed in small quantities (Le Bas, 2008; Doroshkevich et al., 2009). Pyroxene  
467 and amphibole chemistry is highly dependent on the fenitizing fluid composition,  
468 pressure and temperature (PT), in addition to the composition of their precursor  
469 minerals in the protolith (Sutherland, 1969). Magnesioarfvedsonite coexisting with

470 aegirine is the most common mineral assemblage in sodic fenite veins reported in the  
471 literature (e.g. Sutherland, 1969; Hogarth and Lapointe, 1984; Le Bas, 2008). How-  
472 ever, classification of amphiboles from nine carbonatite-related fenites (Fig. 9) shows  
473 sodic-calcic amphiboles such as richterite to be far more common. In rare cases, the  
474 activity of  $F^-$  in the fenitizing fluid is sufficiently high to facilitate crystallization  
475 of F-dominant amphiboles, such as fluororichterite and fluoromagnesio-arfvedsonite  
476 (Bazhenov et al., 1993, 2000). Nedosekova (2007) noted that the F content of amphi-  
477 boles decreases outwards in fenites of the Il'mensky-Vishnevogorsky complex, Russia.  
478 Hogarth and Lapointe (1984) observed at Cantley, Canada, that during early stages  
479 of fenitization, pyroxene typically replaces quartz, whereas feldspar is replaced by  
480 amphibole.

481 The chemical composition, temperature and pressure of fenitizing fluids changes  
482 over time as the source magma cools and evolves. Therefore, sodic fenites represent  
483 the end product of a series of complicated fluid-mineral reactions resulting from  
484 pulses of fenitizing fluids of differing chemical composition and temperature. During  
485 fenitization, alkalis are typically exchanged for silica in an open system, and mineral  
486 chemistry is continuously adjusted as fluid-rock interactions strive toward equilibrium  
487 (Hogarth and Lapointe, 1984). As a result, early-stage fenite mineral assemblages are  
488 often replaced, partially replaced or rimmed by late stage minerals with a composition  
489 reflecting this fluid evolution. For example, at Cantley, pyroxenes with an augite-rich  
490 core and aegirine-rich rim occur in addition to zoned amphiboles with a Ca-poor rim.  
491 These minerals record the change from an early Ca-rich and alkali-poor fluid to a late  
492 Ca-poor and alkali-rich fluid which may have resulted from decreasing temperature  
493 (Hogarth and Lapointe, 1984). Similarly, aegirine-augite at Fen display erratic core-  
494 rim zoning (Andersen, 1989) and fenite at Silai Patti contains aegirine-augite crystals  
495 rimmed by magnesio-arfvedsonite (Le Bas, 2008). Hogarth and Lapointe (1984)

496 suggested that amphiboles and pyroxenes formed in an open system that allowed  
497 adjustment of mineral composition to reflect fluid chemistry and PT conditions,  
498 would exhibit a curved chemical trend. This evolution of mineral chemistry can be  
499 observed at Fen and Alnö, Sweden (see Fig. 9C and 10). Those formed in a closed  
500 system with no exchange of mass, tend to show little geochemical variation and be  
501 clustered on a graph, similar to those from Cantley (see Fig. 9E and 10).

502 Mineral composition also varies within a fenite aureole depending on proximity  
503 to the source intrusion, and therefore grade of fenitization. In the Fen Complex, am-  
504 phiboles proximal to intrusions have richterite and magnesio-arfvedsonite, alkali-rich  
505 compositions. Distally, the amphibole is dominated by arfvedsonite and riebeckite  
506 compositions with lower alkali content (Andersen, 1989). A similar trend is observed  
507 in pyroxenes at Fen, Alnö and Oldoinyo Lengai. At these complexes, Na-rich ae-  
508 girine is prevalent in high grade fenite proximal to the contact, while distal pyroxene  
509 in lower grade fenite trends toward diopside and hedenbergite (see Fig. 10) (Mo-  
510 rogan and Woolley, 1988; Andersen, 1989; Carmody, 2012). This trend is thought  
511 to be enhanced at Fen, due to compositional difference between multiple successive  
512 carbonatite intrusions expelling fluids with variable chemistry (Andersen, 1989).

513 With relatively few exceptions, a feature common to fenitic pyroxenes and am-  
514 phiboles, including those displayed in Figure 9, is the high and apparently non-  
515 stoichiometric silica content. The majority of amphibole analyses show excess Si  
516 relative to the tetrahedral position, leading to Si/T values  $>8$  (see Fig. 9). This phe-  
517 nomenon has previously been identified by Vartiainen and Woolley (1976); Kresten  
518 and Morogan (1986); Morogan and Woolley (1988) and is typically accompanied by  
519 low Al contents. The Ti content has also been observed to decrease in amphiboles  
520 and pyroxenes with increasing fenite grade (Kresten and Morogan, 1986; Morogan  
521 and Woolley, 1988). One explanation for apparently high Si content in these miner-

als could be the presence of significant light trace element substitutions such as Li,  
hosted in the amphibole C-site (Pfaff et al., 2008). Laser ICP-MS analysis of silicate  
minerals from the Gardar Province, South Greenland, has shown high Li concen-  
trations up to 2895 ppm in amphiboles and 278 ppm in pyroxenes (Marks et al.,  
2004). These minerals are typically analysed by microprobe, which is unable to de-  
tect Li, leading to disproportionately high Si contents during formula calculations  
(Pfaff et al., 2008).

### 2.3.2. Potassic fenites

Potassic fenites can contain up to 90 % K-feldspar in addition to albite, relict  
quartz and a minor mineral assemblage including apatite, pyroxene and rutile (Le Bas,  
2008; Doroshkevich et al., 2009). Alternatively, the addition of Mg to a potassic fen-  
ite can cause the phlogopitization of K-feldspar, forming widespread or localized  
accumulations of phlogopite such as those observed in the Sokli fenite (Vartiainen  
and Woolley, 1976). Intense fenitization can ultimately lead to a rock containing  
high proportions of feldspar, assuming that the original protolith is Si-bearing. This  
process has been called convergent fenitization and is common in potassic fenite,  
but uncommon in sodic fenite (Heinrich, 1966). As discussed in section 2.2, potassic  
fenites are typically brecciated (see Table 1) and intruded by later stage magmatic  
events or sodic fenite veins.

### 2.3.3. Glimmerites

Although not as common as orthoclase-rich fenites, a substantial proportion of  
carbonatites (~ 20 %, see Table 1) are associated with micaceous metasomatic rocks  
(Rosatelli et al., 2003), which have not been well-documented or explained. Rocks  
consisting almost entirely of mica (see Fig. 7D) are conventionally labelled glim-

merite and traditionally refer to ultramafic intrusive units (Khamrabayev et al., 1992; Le Maitre et al., 2002). Therefore many glimmerites associated with carbonatites have also been interpreted as igneous (e.g. Hornig-Kjarsgaard, 1998; O'Brien et al., 2015; Sarapää et al., 2015). However, observations at complexes such as Mt Weld, Australia suggest this viewpoint requires revision. This carbonatite complex is surrounded by a 500 m wide fenite aureole, which gradually transitions from a potassium-rich phlogopite rock into a country rock of mafic volcanics (Hoatson et al., 2011), indicating that the glimmerite is not a primary igneous rock. Carbonatite veins and bodies at Bull Hill in the Bear Lodge Mountains, USA, Aley and Upper Fir, Canada (Chakhmouradian et al., 2015; Mitchell, 2015; Moore et al., 2015) are all surrounded by halos of phlogopite glimmerite at the country rock contact. Phlogopite is also widespread in fenites at Sokli, forming along veins or widely disseminated throughout the fenite and thought to be the result of feldspar replacement due to potassic metasomatism (Vartiainen and Woolley, 1976). Khamrabayev et al. (1992) observe that micas found in association with carbonatites are often devoid of any accessory inclusions such as apatite and zircon that are typical of igneous mica. Textural evidence and alteration patterns therefore suggest that at least some glimmerites associated with carbonatites are the result of metasomatic alteration of Mg-rich country rocks and are not igneous in origin (Khamrabayev et al., 1992; Rosatelli et al., 2003; Jones et al., 2013; Chakhmouradian et al., 2015).

#### 2.4. *Rheomorphic Fenites*

Melting of fenites may occur if volatiles, such as CO<sub>2</sub> and H<sub>2</sub>O, and alkalis are introduced into the country rock by fluids exsolved from carbonatite or alkali-silicate magmas. Continued fluxing of volatiles could sufficiently depress the solidus to cause partial melting and injection of fenites into both the carbonatite and surrounding

572 country rock (King and Sutherland, 1966; Evdokimov, 1982; Haggerty and Mariano,  
573 1983; Cooper and Reid, 2000). Heinrich (1966) suggests that mobilization occurs to  
574 such a degree that the fenite behaves as a magma, however, this does not necessarily  
575 imply that the fenite has melted completely. Partial fenite melting to produce a  
576 crystal mush can occur at temperatures of  $\sim 700$  °C, forming magmatic textures  
577 observed in rheomorphic fenites. The addition of volatiles from CO<sub>2</sub> and H<sub>2</sub>O-rich  
578 fenitizing fluids could therefore depress the solidus of particular country rocks such as  
579 metasediments or ultra-mafics, below the estimated fenitization temperature of 400-  
580 500 °C (von Eckermann, 1966; Currie and Ferguson, 1971; Le Bas, 1981; Haggerty and  
581 Mariano, 1983; Morogan and Martin, 1985; Rosatelli et al., 2003). Rheomorphism  
582 of fenites is very difficult to prove or disprove on the basis of field or petrographic  
583 evidence alone, which may explain the skepticism and controversy that surrounds  
584 the subject and consequent lack of research.

585 Isotopically, fenites display a continuous evolution from initial country rock to the  
586 magmatic source of the fenitizing fluids. Ijolites at Iivaara, Finland are observed to  
587 grade into metasomatized country rock, suggesting that the ijolites were formed, at  
588 least in part, by rheomorphism of the high grade fenite (Kramm, 1994). Differences  
589 in Sr and Nd isotopic signatures and trace element abundances are used to sup-  
590 port a different genetic source between recrystallized and primary magmatic ijolites.  
591 Rheomorphic fenites in the Salitre I complex, Brazil were described by Haggerty and  
592 Mariano (1983) as dykes consisting of aegirine, nepheline and sanidine. They are in-  
593 terpreted to be of non-igneous origin due to their highly oxidized nature, a common  
594 feature of fenite, and the paucity of alkaline pyroxene and magnetite observed in the  
595 local igneous suite. Isotopic methods were used by Kramm and Sintern (1998) to  
596 differentiate between texturally and petrographically similar magmatic syenite and  
597 syenitic fenites at Oldoinyo Lengai, Tanzania. The latter are high grade fenites with

598 a magmatic texture thought to originate by partial melting.

599 The term pseudotrachyte was first introduced by Sutherland (1965a) to describe  
600 trachytes that were not intrusive, but appeared to have formed *in-situ*. The term  
601 was used to describe trachyte-like rocks from Toror, Alnö and Kaiserstuhl. Described  
602 as being derived from brecciated feldspathic fenites that had become locally mobi-  
603 lized (Sutherland, 1965b; Le Bas, 1981), these rocks had all the characteristics of  
604 a primary magmatic trachyte: sheeted, porphyritic, aphanitic groundmass and flow  
605 textures (Sutherland, 1965b). Pseudotrachytes were also described by Cooper and  
606 Reid (2000) at Dicker Willem, alongside Sr, Nd and Pb isotope data that resembled  
607 those of the high grade potassic fenites, not the magmatic suite of rocks. The tran-  
608 sition of a recrystallized feldspathic rock to feldspathic breccia, and then mobilized  
609 pseudotrachyte that intruded the country rock at Tundulu, was traced by Garson  
610 (1962). These rocks consist of small fragments of feldspar, recrystallized to form  
611 clear K-rich feldspar laths in a microcrystalline matrix (Le Bas, 2008).

### 612 **3. Fenites related to economic deposits**

613 Alkaline and carbonatite complexes are often associated with high concentrations  
614 of economically important minerals, enriched in REE, Nb and phosphate (Wall, 2014;  
615 Goodenough et al., 2016). These rocks also host many non-critical resources such  
616 as iron, copper, titanium, fluorite, uranium, thorium and vermiculite, some of which  
617 have been mined since 770 AD (Heinrich, 1966; Mariano, 1989; Pell, 1996).

618 Localities described and discussed in this section have been defined as ‘of economic  
619 importance’, either as a past producer, an active mine or a deposit (i.e. those with  
620 a quantified mineral resource) by Woolley and Kjarsgaard (2008b).

621 *3.1. Fenites at REE-rich deposits*

622 Drawing similarities between fenites surrounding REE-rich carbonatites is prob-  
623 lematic due to the majority of deposits lacking detailed descriptions, including at-  
624 tributes or mineralogy, of any fenite present. Additionally, fenites are zoned, both  
625 horizontally and vertically, and therefore differing levels of erosion can remove im-  
626 portant features or not reveal fenites adjacent to intrusions, causing difficulties when  
627 attempting to model an integrated system.

628

629 Fractional crystallization processes cause magmas to evolve (Thompson et al.,  
630 2002; Ray and Shukla, 2004), typically in a plutonic environment, to form a typical  
631 sequence of multiple carbonatite generations. These generations may not all be  
632 observed at a given site, but typically evolve from a calcite to dolomite and then  
633 a later ferrocarnatite phase (Heinrich, 1966; Le Bas, 1981). Magma evolution  
634 is associated with increasing Fe, Ba, Mn and decreasing Sr (Le Bas, 1981), and  
635 REE tend to be concentrated in the later or last carbonatite differentiates (Heinrich,  
636 1966; Le Bas, 1981; Wall, 2014). Each phase of carbonatitic magmatism is associated  
637 with the release of fenitizing fluids, leading to multiple stages of fenitization which  
638 are represented as different assemblages or vein generations in the country rocks.  
639 As such, the complexity of a fenite could potentially reflect magma evolution, and  
640 therefore the likelihood of REE-enrichment in the source intrusions.

641 The Fen Complex is one such site that displays all three of these carbonatite  
642 species, with the last differentiate (ankerite carbonatite) enriched in Th and REE up  
643 to 1.5 wt.% (Andersen, 1986; Lie and Østergaard, 2011; Marien et al., 2017). This  
644 is reflected in fenite surrounding the complex which increases in total rare earths  
645 (TREE) and HREE with increasing intensity of fenitization (Andersen, 1986; Lie and  
646 Østergaard, 2011). Several stages of carbonatite intrusions can also be observed at

647 Mountain Pass, including calcite, dolomite and a stage with composition intermediate  
648 between these two which contains the most economic REE grades. Fenitized gneisses  
649 can contain up to 9% rare earth oxides (REO) hosted by allanite. These fenites  
650 contain magnesio-riebeckite that replaces earlier amphibole and pyroxene (Castor,  
651 2008).

652 Fenites surrounding REE-enrichmented intrusions typically display both sodic  
653 and potassic metasomatism, suggesting fenitization resulted from multiple fluid pulses.  
654 Amba Dongar is a clear example of this association, displaying vertical zonation of  
655 sodic and potassic fenites (see section 2.1) related to calcite, ankerite and siderite car-  
656 bonatite intrusions. Sodic fenites at this site contain mineral assemblages of aegirine-  
657 augite, alkali feldspar and quartz whereas the potassic fenite consists predominantly  
658 of K-feldspar, quartz and albite with only minor pyroxene. These florencite-bearing  
659 potassic fenites display higher Nd/La ratios than the carbonatite intrusions, indicat-  
660 ing HREE enrichment in the fenites (Doroshkevich et al., 2009). Late-stage carbon-  
661 atite dykes cross-cut both sodic and potassic fenites in the Jammi area of the Sokli  
662 Complex, and host the highest REE content, enriching the surrounding fenites up  
663 to 2 % TREE (Sarapää et al., 2013). Intensely brecciated potassic fenites have been  
664 recorded at the Lofdal carbonatite-nepheline syenite complex, extending up to sev-  
665 eral metres from carbonatite intrusions and consisting predominantly of K-feldspar  
666 (Bodeving et al., 2017). Calcite carbonatite at Lofdal contains on average 1300 ppm  
667 TREE compared to 356 ppm TREE in the potassic fenite. Brecciated and intensely  
668 metasomatized early sodic fenites several tens of metres wide, consist almost entirely  
669 of albite and quartz and host aggregates of later HREE-rich xenotime (Swinden and  
670 Siegfried, 2011; Dodd et al., 2014).

671

672 As discussed in section 2.2, brecciation events often accompany carbonatite em-

673 placement (Heinrich, 1966; Tuttle and Gittins, 1966; Le Bas, 1977; Rubie and Gunter,  
674 1983). Although incidents of brecciation have not been reported at every REE-rich  
675 carbonatite complex, there is a strong correlation between the presence of breccia  
676 and mineralization. Table 2 shows that 70 % of REE-enriched fenites have breccia-  
677 tion reported in their associated literature. Lack of observation does not necessarily  
678 mean lack of brecciation, due to the literature focus, bias of observations and level of  
679 erosion. Widespread brecciation associated with carbonatite emplacement indicates  
680 some pulses of fluids and volatiles sourced from the evolving magma are released ex-  
681 plosively (Verplanck et al., 2014). The presence of these fluids and volatiles attests  
682 to the evolution and crystallization of the carbonatite magma, the same processes  
683 that concentrate incompatible REEs in the residual carbonatite melt (Heinrich, 1966;  
684 Le Bas, 1981; Wall, 2014), therefore it is logical that an association between breccia-  
685 tion and mineralization would exist.

686 The Chilwa Alkaline Province comprises at least 14 carbonatite and 15 alkaline  
687 intrusions (Woolley, 2001). Exposures of breccia can be found at six of these intru-  
688 sive complexes, the majority of which consist of feldspathic breccia (see Fig. 7A)  
689 in close proximity to the intrusion-fenite contact. The Kangunkunde carbonatite  
690 is surrounded by a feldspathic breccia which grades into an intermediate to sodic  
691 fenite with distance from the intrusion. However, the Chilwa Island breccia exhibits  
692 a sharp contact and much higher concentrations of potassium (Woolley, 1969; Dow-  
693 man, 2014). Breccias at Songwe Hill, Malawi can be divided into carbonatite and  
694 fenite breccias, the latter of which can contain metasomatized clasts several metres  
695 in diameter and hosting up to 1.37 Mt of mineralized material at a grade of 1.61  
696 % TREO (Croll et al., 2015). Brecciation is not restricted to complexes in Malawi,  
697 but is observed surrounding carbonatites worldwide. Intensity of brecciation often  
698 decreases with distance from the intrusion-fenite contact such as the multi-stage

699 brecciation at Fen (Verschure and Maijer, 2005; Lie and Østergaard, 2011), Sokli  
700 (Vartiainen and Paarma, 1979) and Lofdal. Quarrying within the Lofdal complex  
701 has exposed breccias containing clasts fenitized to varying degrees, thought to rep-  
702 resent multi-phase gaseous release events preceding the intrusion of a large body of  
703 carbonatitic magma and numerous vein systems (see Fig. 7B).

704

705 The role of ligands in the transportation of REEs into the fenite is discussed in  
706 greater detail in section 4. Fenitizing fluids are often carried away in vein networks  
707 and dissipate, providing little evidence beyond rare fluid inclusions to support their  
708 theoretical composition. However, some complexes contain minerals directly precip-  
709 itated from the fenitizing fluids that provide clues as to their chemical composition.  
710 One example is the presence of minerals such as fluorapatite in the fenites of Lofdal,  
711 Namibia that attests to the presence of  $F^-$  and  $PO_4^{3-}$  (Wall et al., 2008) in the feni-  
712 tizing fluids. Late stage HREE-enriched fluorite at Okorusu, Namibia is thought to  
713 result from the growing dominance of carbonate over fluoride ligand complexing with  
714 time (Bühn et al., 2002), resulting in overprinting of the fenite near the carbonatite-  
715 country rock boundary with 7–10 Mt of 35 wt.%  $CaF_2$  (Deans et al., 1972; Mariano,  
716 1989). Enrichment of  $PO_4^{3-}$ ,  $CO_3^{2-}$ ,  $HCO_3^-$  and  $F^-$  in the fenitizing fluids of the Alnö  
717 Complex, facilitated the mobilization of REE out of the carbonatite intrusion and  
718 into the surrounding fenite aureole, enriching the adjacent potassic fenite by 102-499  
719 ppm TREE (Morogan, 1989). The Amba Dongar carbonatite complex has been ac-  
720 tively mined for fluorite since 1972. The 11.6 Mt deposit at 30 wt%  $CaF_2$  is hosted  
721 in fenite veins and fractures formed by the interaction of carbonatite-derived fluids  
722 and surrounding Cretaceous sandstones (Deans et al., 1972; Mariano, 1989; Palmer  
723 and Williams-Jones, 1996). This florencite-bearing fenite was enriched in REE be-  
724 fore late stage fluorite precipitation (Doroshkevich et al., 2009) resulting from the

725 interaction of F-bearing carbonatite-derived fluids and low temperature Ca-bearing  
726 meteoric fluids (Palmer and Williams-Jones, 1996).

727

728 The country rock into which the carbonatite intrudes and size of the resulting  
729 fenite show no apparent correlation with REE enrichment in the source intrusion  
730 or the fenite. Carbonatites can intrude a wide variety of country rock types which  
731 have no bearing on the degree of REE-enrichment in the intrusion or resulting fenite  
732 (e.g. sandstone, Amba Dongar - Doroshkevich et al. (2009); granite gneisses, Sokli -  
733 Appleyard and Woolley (1979); Vartiainen and Paarma (1979); Sarapää et al. (2013);  
734 and sedimentary sequences, Bayan Obo - Le Bas (2008) and Mianning, China - Xie  
735 et al. (2014)). The degree of REE enrichment in the source intrusion is not propor-  
736 tional to the radius of subsequent fenitization. Carbonatite at Dorowa, Zimbabwe  
737 contain <160 ppm TREE, and are surrounded by >1 km of fenitization (Johnson,  
738 1966; Harmer et al., 1998). In contrast, the ankerite carbonatite at Amba Dongar  
739 locally contain >15 wt.% TREE and are associated with <300 m of fenitization  
740 (Doroshkevich et al., 2009).

### 741 3.2. Fenites at niobium-rich deposits

742 Carbonatites and alkaline rocks are the world's largest source of niobium (Mar-  
743 iano, 1989), hosting all currently and formerly operating niobium mines. Niobium  
744 tends to be found in the intermediate stages of carbonatite differentiation, in contrast  
745 to REE which tend to be found in the later, more evolved magmas (Mariano, 1989).  
746 Primary carbonatite-hosted niobium deposits typically contain between 3000–5000  
747 ppm Nb, however, secondary and supergene processes can further enrich niobium  
748 concentration by more than a factor of 10 (Mitchell, 2015). Intense weathering of  
749 the Araxá Complex, Brazil has produced a lateritic residue containing 462 Mt of ore

750 at 2.5 wt.% Nb<sub>2</sub>O<sub>5</sub> with a thickness of >120 m (Mitchell, 2015).

751 The high charge to ionic radius ratio of niobium makes it insoluble in fluids other  
752 than silicate melts, therefore ligands must be present to enhance its mobility (Lin-  
753 nen et al., 2014). Experimental study of the solubility of Nb in aqueous solutions  
754 indicates an increase in solubility with an increased concentration of HF, indicating  
755 ligand complexing to form NbF<sub>2</sub>(OH)<sub>3</sub> (Timofeev et al., 2015). As such, niobium  
756 should be mobilized by fenitizing fluids into the metasomatized country rock (Le Bas,  
757 2008). Evidence of this process has been reported at Kangunkunde in the form of  
758 fenites enriched in Nb to concentrations higher than those of the source carbonatite,  
759 facilitated by the presence of F<sup>-</sup> in the fenitizing fluids (Dowman, 2014). Amphiboles  
760 and biotites associated with niobium mineralization in the Bayan Obo Complex are  
761 enriched in fluorine relative to the rest of the fenite, indicating high HF activity  
762 in the fenitizing fluids (Smith, 2007). Mitchell (2015) also states that pyrochlore  
763 can be found in significant quantities (50 vol.%) in fenite veins associated with Nb-  
764 rich carbonatite complexes such as Sarfartôq, Greenland and Prairie Lake, Canada.  
765 Therefore, Nb is being mobilized in fenitizing fluids in these complexes. Pyrochlore  
766 group minerals are major constituents in the carbonatites, phoscorites and metaso-  
767 matic fenite aureoles of the Sokli alkaline-carbonatite complex (see Fig. 7F) (Lee  
768 et al., 2006).

#### 769 4. Fenite as an exploration indicator

770 Fenitizing fluids contain ligands such as chloride-, fluoride-, sulfate-, phosphate-  
771 and/or carbonate anions, which can form complexes with REE and Nb, substantially  
772 enhancing their solubility (Andersen, 1986; Haas et al., 1995; Williams-Jones et al.,  
773 2012; Tsay et al., 2014) and allowing transport of these elements into the surrounding  
774 fenite aureoles (see Table 2). Here they precipitate to form mineral assemblages of

775 REE, Nb and fenite minerals (see Fig. 7E) (Hogarth, 2016; Bodeving et al., 2017;  
776 Dowman et al., 2017).

777 The strength of different REE and Nb complexes in aqueous solution can vary,  
778 depending on the ligand. Research into this speciation indicates that Cl<sup>-</sup> ligands in-  
779 crease LREE solubility, whereas F<sup>-</sup> and CO<sub>3</sub><sup>2-</sup> ligands form stronger complexes with  
780 MREE and HREE (Williams-Jones et al., 2012; Tsay et al., 2014). The composition  
781 of the fenitizing fluid can therefore lead to LREE/HREE fractionation. Harlov et al.  
782 (2002) indicates that Cl-rich and F-rich fluids could have responsible for the depletion  
783 of Y, REE, Na and Si from apatites at Kiirunavaara, Sweden, and Broom-Fendley  
784 et al. (2016) observed a similar LREE depletion in apatites from the Kangankunde  
785 carbonatite. Dolomitic melts at the Seligdar complex, Russia, evolved brines con-  
786 taining 38–42 wt.% NaCl-eq which likely removed LREE and Th from apatites, and  
787 redistributed these elements into monazite-(Ce) and xenotime-(Y) (Prokopyev et al.,  
788 2017). Calcite carbonatite ring dykes on Chilwa Island were depleted in HREE rela-  
789 tive to high grade fenites and breccias which have higher HREE:LREE ratios. This  
790 fractionation most likely resulted from F<sup>-</sup> in late-stage fenitizing fluids preferentially  
791 mobilizing HREE, as evidenced by fluorite, quartz and barite veining (Dowman et al.,  
792 2017). Similar trends have been observed at Fen and Alnö, both of which display in-  
793 creasing REE fractionation with decreasing intensity of fenitization (Morogan, 1989;  
794 Verschure and Maijer, 2005). Sodic fenites at Alnö display greater HREE enrich-  
795 ment, with La/Lu ratios increasing from low to high intensity fenitization, proximal  
796 to the carbonatite (Morogan, 1989).

797  
798 Micro-mineral assemblages enriched in REE and Nb have been recorded in fen-  
799 ites globally (see Fig. 7E and 7F, Table 2). Locations of many carbonatite com-  
800 plexes have already been well documented (Woolley and Kjarsgaard, 2008a), however

801 drilling is often the most costly stage of exploration. Thus, the features, mineralogy  
802 and geochemistry of a fenite aureole may act as an exploration indicator to determine  
803 the level of REE or Nb enrichment in the source intrusion. In addition, fenites have a  
804 large footprint, sometimes up to several kilometres diameter and extensive in depth.  
805 The distinctive characteristics of the concentric fenite aureole pattern, easily distin-  
806 guished in the field, could act as a vector toward unexposed carbonatite intrusions  
807 and ore-deposits. Recognition of potassic feldspar-pyrite alteration related to gold  
808 mineralization in alkaline igneous rocks has already been used as a key to successful  
809 exploration at Bear Lodge (Noble et al., 2009). Broad zones of similar, pervasively-  
810 altered country rock accompany vein-hosted gold mineralization at Cripple Creek,  
811 USA, which have also been noted for their REE enrichment and the presence of  
812 bastnäsite and monazite (Noble et al., 2009). Alkali metasomatism associated with  
813 potassic syenites has also been used to focus gold exploration in Russia (Dvornik,  
814 2015).

## 815 **5. Discussion**

### 816 *5.1. Stages of fenitization and their timing*

817 Fenitization is described as a polyphase process, resulting from multiple pulses  
818 of fluid released from a cooling and crystallizing intrusion (Morogan, 1994; Le Bas,  
819 2008). These fluids form inner potassic and outer sodic fenite aureoles, however  
820 there is still much debate about how they form. Many theories exist as to whether  
821 each aureole relates to a discrete fenitization event involving fenitizing fluids of dif-  
822 ferent composition, or whether each fluid pulse evolves with distance from the source  
823 intrusion.

824 The elemental exchange that occurs between the fluid and rock during metasoma-  
825 tism can cause the fluid to evolve spatially and temporally (Harlov and Austrheim,

826 2012; Kusebauch et al., 2013). As such, intense fenitization near the intrusion-  
827 country rock boundary could theoretically change the fluid chemistry, forming a  
828 fenite of differing chemical composition distal to the source intrusion. This process  
829 could potentially explain the pattern of inner potassic and outer sodic fenite aure-  
830 oles observed at many carbonatite complexes (see section 2.3). However, the change  
831 in fluid composition would likely be gradual as fenitization reactions progressed. A  
832 gradational boundary between aureoles would therefore be expected, which is not  
833 recorded in the literature or observed by the authors.

834 In contrast, many carbonatite complexes display cross-cutting relationships be-  
835 tween early sodic fenites cut by later potassic stages (Woolley, 1969, 1982). A number  
836 of explanations have been proposed for the expulsion of fluids with different chem-  
837 istry, including falling temperatures or increasing CO<sub>2</sub> levels (Rubie and Gunter,  
838 1983), or an initial Na-rich intrusion which gradually loses Na to fenitization until K  
839 becomes the dominant alkali (Woolley, 1982). Fluid inclusion studies of the Amba  
840 Dongar fenites indicate that Na-rich fluids were expelled first at a depth of >10  
841 km followed by later expulsion of K-rich fluids upon the magma reaching shallower  
842 crustal levels (Doroshkevich et al., 2009). The presence of multiple cross-cutting vein  
843 generations (Le Bas, 2008) and zoned apatites also provide evidence to support fenite  
844 formation by multiple fluid pulses as opposed to a single evolving fluid.

845  
846 The process of fenitization is often considered to involve the metasomatic addi-  
847 tion of alkalis and removal of silica from the country rock (e.g. Brögger, 1921; von  
848 Eckermann, 1948; McKie, 1966). The metasomatic replacement of K-feldspar with  
849 plagioclase also releases excess silica, often causing intergrown plagioclase-quartz  
850 textures such as myrmekite (Becke, 1908). However, the fate of this silica in the  
851 carbonatite-fenite system has been an ongoing conundrum for geologists.

852 Upon studying the Alnö carbonatite complex, von Eckermann (1948) hypothe-  
853 sized that silica removed during metasomatic processes migrated into the carbonatite  
854 intrusion. Woolley (1969) expanded on this idea and suggested that silica was driven  
855 inward and upward, carried either up a central pipe incorporated within the mag-  
856 matic intrusion, or carried to the surface as a brine. This hypothesis is supported  
857 by Skelton et al. (2007) and Vartiainen and Woolley (1976). The latter suggested  
858 that high level extensively silicified Rufunsa Valley carbonatites, Zambia may have  
859 acted as a sink for fenite-derived silica. The presence of wollastonite in the outer  
860 zone of the carbonatite at Alnö, Sweden is thought to have formed via the metaso-  
861 matic reaction of calcite with quartz (Skelton et al., 2007; Putnis and Austrheim,  
862 2010), also indicating an ingress of silica down gradient into the carbonatite. Sil-  
863 ica in the Melteig fenites of the Fen Complex, Norway appears to have a different  
864 destination however. Observations by Kresten and Morogan (1986) indicate that  
865 low grade fenites in this area contain more quartz than the initial gneissic granite  
866 protolith and quartz veins are common, indicating a degree of silicification. Late  
867 stage silica-rich veins are common in many carbonatite complexes including Sokli,  
868 Finland (Vartiainen and Woolley, 1976) and the Wasaki Peninsula, Kenya (Le Bas,  
869 2008), suggesting that fenitizing fluids which have exchanged alkalis for silica during  
870 metasomatic reactions, may then transfer and precipitate quartz in the outer aureole.  
871 The common factor between these complexes is that silicification appears to be the  
872 last stage of fenitization.

### 873 *5.2. Implications of veining and brecciation*

874 Brecciation has significant implications for fenitization and mineralization in car-  
875 bonatite complexes, creating efficient fluid pathways and facilitating fluid movement  
876 (Verschure and Majjer, 2005; Lie and Østergaard, 2011) toward the outer fenite

877 aureole (see Graphical Abstract). The importance of breccias in relation to fluid  
878 movement is demonstrated by feldspar-rich fenites at the Wasaki Peninsula, Kenya.  
879 These contain fractures filled with a mix of iron oxides, aegirine-augite, feldspars,  
880 carbonate and quartz that grade into a feldspathic breccia proximal to the intrusion  
881 (Le Bas, 2008). Brecciation also often precedes mineralization such as F, Ba and U-  
882 Th mineralization (Le Bas, 1987) and is found to occur prior to REE-fluorcarbonate  
883 precipitation at Tundulu, Malawi (Broom-Fendley et al., 2016).

884 Formation of fine-grained mineral phases during fenitization may effectively act  
885 as a seal, retaining fluid in the immediate vicinity of the intrusion. This process is  
886 analogous to the kimberlite alteration model proposed by Afanasyev et al. (2014),  
887 in which fluids flowing through the pipe are proposed to reduce breccia porosity  
888 and permeability by alteration to fine hydrous minerals. This process suppresses  
889 fluid flow and effectively seals off unaltered sections, very applicable to fenites which  
890 display highly heterogenous intensities of metasomatism. Sealing of magmatic and  
891 hydrothermal fluids in the inner complex, by the formation of early fenites may lead  
892 to pressure build up exceeding lithostatic pressure, resulting in hydraulic fracturing  
893 and the formation of shock breccias. Rapid decompression following the failure of  
894 country rock can lead to boiling and separation of H<sub>2</sub>O and CO<sub>2</sub> phases, causing  
895 destabilization of complexing ligands and subsequent mineral precipitation (Robb,  
896 2005). Multiple pulses of these fluids form veins of varying composition through-  
897 out the lifetime of the complex (see Graphical Abstract). Early stage sodic fenite  
898 veins containing pyroxenes and amphiboles are superseded by intermediate stage  
899 Nb-bearing veins followed by late stage REE-bearing veins. The micro-mineral as-  
900 semblages precipitated from these fenitizing fluids can provide information regarding  
901 element enrichment in the source intrusion.

902

903 The style of fenitization could also be used to determine the approximate extent  
904 of erosion and relative depth in a volcanic system. High pressures prevalent at deeper  
905 crustal levels resist the pervasive migration of fluid through the rock, and therefore  
906 favour flow in structural conduits such as veins (Robb, 2005). Deep crustal vein  
907 systems indicate that fluid is channelized on a coarse scale, although a small degree  
908 of pervasive diffusive scale can occur between veins (Newton, 1989). Intrusion of  
909 magma into very shallow crust, and the subsequent release of fluids, is typically  
910 accompanied by intense fracturing of the host rock (Meinert, 1992). Therefore the  
911 presence of breccia in a fenite aureole is likely to be indicative of very shallow crustal  
912 levels. Knowledge regarding the relative depth of erosion and position of the fenites  
913 in the volcanic system is important to determine where the source intrusion lies  
914 relative to the current surface, and whether there is potential for the presence of an  
915 associated mineral deposit. However, these fluid pathways are not mutually exclusive  
916 and veining can be spatially extensive and form throughout the intrusion's cooling  
917 history (Misra, 2000).

918 The fenite fluid flow pathways and the resulting fluid-rock interaction has im-  
919 portant consequences for the morphology of reaction fronts and the distance over  
920 which elements can be transported, as demonstrated by skarn and porphyry stock-  
921 work systems (Ortoleva et al., 1987; Kelemen et al., 1995; Robb, 2005). Migration  
922 of fluids in fracture or vein networks limits the volume of country rock interacting  
923 with the fluid (Cartwright and Barnicoat, 1999), therefore maintaining chemical gra-  
924 dients over larger distances. Better understanding of the fluid flow mechanism and  
925 dynamics in fenites has important implications for understanding element redistri-  
926 bution from source intrusions into surrounding fenites. The presence of a fenite vein  
927 network could transport fluids further from the source, enriching the outer aureoles  
928 in elements such as REE and Nb. This geochemical signature and resulting indicative

929 mineral assemblages could be used during exploration to find the source intrusion.

### 930 *5.3. Properties and chemistry of fenitizing fluids*

931 There have been a number of studies attempting to provide a chemical and ther-  
932 modynamic framework for fenitization processes. Understanding the chemical and  
933 physical properties of the fenitizing fluids can assist in determining the nature of  
934 the magmatic source and any associated element enrichments, potentially indicating  
935 the presence of an economic mineral deposit. The multiple pulses of fluid associ-  
936 ated with fenitization cause differences in the distribution and abundance of REE in  
937 sodic, intermediate and potassic fenites (Morogan, 1989; Dowman, 2014) relating to  
938 evolution of the source melt.

939  
940 Experimental work undertaken by Zaraiskii (1989) (summarized by Bardina and  
941 Popov (1994); Zharikov and Rusinov (1998)) indicates a lower fenitization tempera-  
942 ture limit of  $500 \pm 30$  °C at 1 kbar, defined by the stability of the aegirine-augite-  
943 microcline-perthite paragenesis. The upper limit of fenitization is not as well con-  
944 strained, but probably approaches 700 °C, based on consensus of measured tempera-  
945 tures between 600–700 °C at Amba Dongar (>700 °C, Williams-Jones and Palmer,  
946 2002); Iivaara, Finland (660 °C, Sindern and Kramm, 2000); and Alnö (640 °C –  
947 Skelton et al., 2007). This temperature is supported by the stability of alkali amphi-  
948 boles such as riebeckite and arfvedsonite, which are constrained between 650–800 °C  
949 (Kovalenko et al., 1977).

950 Fluids are considered to be highly oxidizing, causing significant quantities of  $\text{Fe}^{3+}$   
951 to be incorporated into the fenite mineral assemblage, including feldspar, pyroxene  
952 and hematite crystals (Haggerty and Mariano, 1983). This trait is easily recognized  
953 using cathodoluminescence, due to ferric iron substituting in the tetrahedral sites

954 of feldspar crystals, functioning as an activator for bright red cathodoluminescence  
955 (see Fig. 5C and 5D) (Mariano, 1983; Finch and Klein, 1999; Mariano and Mariano,  
956 2014) - this is termed the “alkali ferric iron effect”(Carmichael and Nicolls, 1967).

957

958 Fenitizing fluids expelled from carbonatites are highly variable. Fluids are pre-  
959 dominantly aqueous with relatively low activities of subordinate CO<sub>2</sub>, but CO<sub>2</sub>/H<sub>2</sub>O  
960 ratios can vary greatly (Drüppel et al., 2005; Le Bas, 2008). Fluid inclusions of  
961 carbonatite-derived fluid at Kalkfeld, Namibia, analysed by Böhn and Rankin (1999),  
962 display a wide range in composition from CO<sub>2</sub>-rich to Cl<sup>-</sup> and HCO<sub>3</sub><sup>-</sup>-bearing aque-  
963 ous fluids. However, they are typically thought to be enriched in alkalis, halogens,  
964 CaO, MgO and FeO, with varying proportions of Sr, Ba, Nb and REE (Morogan,  
965 1989; Sindern and Kramm, 2000; Drüppel et al., 2005; Skelton et al., 2007). These  
966 fluids are also thought to be deficient in SiO<sub>2</sub> and Al<sub>2</sub>O<sub>3</sub>, thereby removing these  
967 elements from the country rock with subsequent redistribution in the fenite aureole  
968 (Morogan, 1989; Sindern and Kramm, 2000; Skelton et al., 2007).

969 Mass balance calculations of element transfer during fenitization have indicated  
970 a substantial volume loss during the process, calculated as a 6 % volume loss in the  
971 highest grade fenite at Alnö (Morogan, 1989) and a 20 % volume loss at Iivaara (Sin-  
972 dern and Kramm, 2000). This loss in volume is problematic when determining the  
973 true elemental gains/losses during fenitization, as observed changes may solely be  
974 due to loss of mass. To remedy this issue, Gresens (1967) developed an equation to  
975 determine the mass transfer during metasomatism, accounting for both volume and  
976 compositional changes. Grant (1986) later adapted this concept into the isochron  
977 diagram which uses elements with no apparent gain/loss during metasomatism as a  
978 reference to determine changes in volume and elemental concentrations.

979

980 Fluids related to the vertically zoned fenites of Amba Dongar have been exten-  
981 sively studied and modelled by Williams-Jones and Palmer (2002) based on fluid  
982 inclusion data. The first fluid to exsolve from the calcite carbonatite is thought to  
983 be at  $>4$  kb and  $>700$  °C, the same temperature as the carbonatite solidus, represent-  
984 ing expulsion at a depth of  $>10$  km to form the lower sodic fenites. Upon reaching  
985 shallower crustal levels of 3–5 km depth, the last fractions of fluid exsolved. These  
986 fluids were solute-rich, displayed an intermediate K/Na ratio and were enriched in  
987 CaO, Al<sub>2</sub>O<sub>3</sub> and SiO<sub>2</sub>, forming the upper potassic fenite. Fenitization described  
988 at Alnö by Skelton et al. (2007) indicates an original mineral assemblage of biotite,  
989 quartz and oligoclase that was metasomatized to a secondary mineral assemblage of  
990 albite, K-feldspar, arfvedsonite and aegirine-augite by the addition of K<sub>2</sub>O, Na<sub>2</sub>O,  
991  $\pm$  CaO, MgO, FeO causing the release of SiO<sub>2</sub> and H<sub>2</sub>O.

992  
993 The genesis of nepheline resulting from metasomatic alteration of a silicate min-  
994 eral assemblage, via nephelinization, has caused controversy since von Eckermann  
995 (1948) published his memoir regarding Alnö. Revisiting this problem, Tilley (1957)  
996 discovered feldspar textures, such as rounded cores surrounded by nepheline rims  
997 and vermicular nepheline growths, that suggested a metasomatic origin. In contrast,  
998 Vartiainen and Woolley (1976) state that no nephelinization has been observed in  
999 fenites related to carbonatite intrusions. The albite-orthoclase join of the quartz-  
1000 albite-orthoclase ternary diagram most likely represents the most extreme extent of  
1001 de-silicification of which carbonatite-derived fenitizing fluids are capable. Although  
1002 the metasomatic origin of the mineral is still debated, observations of nepheline in  
1003 fenite aureoles have been recorded at many carbonatite complexes e.g. fenitization of  
1004 granite at Oldoinyo Lengai (Morogan, 1994); fenitization of gneisses at Oka, Canada  
1005 (Samson et al., 1995); in rheomorphic fenites surrounding the Paraná Basin car-

1006 bonatites, South America (Haggerty and Mariano, 1983); and fenitization of granite  
1007 gneisses at Gifford Creek, Australia (Pirajno, 2015).

#### 1008 *5.4. Timescales of fenitization*

1009 Very little is understood in relation to the timescale over which fenitization occurs.  
1010 The duration of fluid-rock interaction at Alnö has been calculated by Skelton et al.  
1011 (2007) using the disparity between metasomatic reaction and isotope fronts, taking  
1012 into account permeability enhancement by metasomatic reactions. An estimated  
1013 timescale of  $10^2$ – $10^4$  years has been proposed, based on the calculation that fluid  
1014 diffusivity was  $10^{-7}\text{m}^2\text{s}^{-1}$  at 600 °C.

1015 No other studies have been undertaken as a comparison, however analogies can  
1016 be drawn between fenitization and metamorphism in the presence of a fluid phase.  
1017 Thermodynamic modelling undertaken by Wood and Walther (1983) indicates that a  
1018 prograde dehydration metamorphic reaction at 700 °C of 2 mm crystals, would reach  
1019 completion in 70 years, with the H<sub>2</sub>O/CO<sub>2</sub> ratio of the hydrothermal fluid having no  
1020 effect on timescales. This large discrepancy between estimated timescales reflects the  
1021 lack of understanding relating to fenitization processes, and identifies new research  
1022 avenues to further our understanding of fluid-related mineralization.

#### 1023 *5.5. Why do fenites associated with alkaline and carbonatite intrusions differ?*

1024 A detailed discussion of fenitization related to alkaline intrusions goes beyond the  
1025 scope of this paper, however the extensive mineralization associated with large com-  
1026 plexes such as those of the Kola Peninsula, Russia and Gardar Province, Greenland,  
1027 emphasizes their importance in understanding these systems. Little research has  
1028 been dedicated to the comparison of alkaline and carbonatite-related fenitization,  
1029 mostly likely due to the ambiguity of fluid sources created by the close temporal and

1030 spatial association of these intrusions (Woolley, 2003; Le Bas, 2008).

1031

1032 Among the different magmas capable of generating fenitizing fluids, melteigite-  
1033 ijolites and carbonatites appear to be associated with the most voluminous metaso-  
1034 matic overprint. However, peralkaline intrusions produce comparatively small fen-  
1035 ites. For example, fenitization extends 1–2 km from the central Sokli carbonatite  
1036 intrusion (Al Ani and Sarapää, 2009), whereas fenitization surrounding large, highly  
1037 evolved, agpaitic nepheline-syenite intrusions such as Khibiny and Lovozero, Kola  
1038 Peninsula, and Ilímaussaq, Greenland, does not typically exceed 100 m from the  
1039 intrusion (Ferguson, 1964; Gorstka, 1971; Arzamastev et al., 2011).

1040 This striking disparity was interpreted to result from a greater solubility of H<sub>2</sub>O  
1041 in peralkaline melts (Kogarko, 1977; Burnham, 1979; Giehl et al., 2013, 2014), lead-  
1042 ing to the effective retention of volatiles in small batches of evolved residual melts  
1043 enriched in alkalis, ligands, high field strength elements (HFSE) and other incom-  
1044 compatible elements such as REE (Marks et al., 2003; Arzamastev et al., 2011; Giehl  
1045 et al., 2014; Kozlov and Arzamastsev, 2015; Marks and Markl, 2015). These are  
1046 hypothesized to cause limited fenitization, due to fluids exsolving after the bulk of  
1047 the intrusion has already solidified (Arzamastev et al., 2011). Fluids expelled from  
1048 these melts contain variable amounts of CH<sub>4</sub> or CO<sub>2</sub>, but are typically aqueous  
1049 and Na-rich, often with high salinities up to 30 wt.% NaCl equivalent (Konnerup-  
1050 Madsen, 2001; Krumrei et al., 2007; Graser and Markl, 2008). Fig. 11 indicates  
1051 no difference in the ratios of Al–Na + K–Fe + Mg between carbonatite-derived and  
1052 alkaline-derived fenites, and as in carbonatite systems, these fluids have the capacity  
1053 to mobilize HFSE and REE (Migdisov et al., 2009; Timofeev et al., 2015).

1054

1055 Fluids released from the magma migrate through the periphery of crystallized in-

1056 trusions, often causing intense metasomatic reactions and hydrothermal alteration.  
1057 Secondary assemblages of minerals are formed at the expense of primary magmatic  
1058 minerals (see Fig. 12) (Mariano, 1983; Chakhmouradian and Mitchell, 2002; Marks  
1059 et al., 2003; Graser and Markl, 2008; Schilling et al., 2009; Borst et al., 2016), of-  
1060 ten causing mobilization of REE and Nb-rich phases (Chakhmouradian and Mitchell,  
1061 2002). This is the same process termed ‘internal fenitization’ by Mariano (1983). Au-  
1062 tometasomatic reactions and subsequent decrease in volume of fluid released, could  
1063 therefore explain the limited fenitization observed around nepheline syenites (Arza-  
1064 mastev et al., 2011). The Ilímaussaq nepheline syenite complex, Greenland displays  
1065 plenty of evidence for these autometasomatic processes acting in and along the com-  
1066 plex margins, but with limited effect on the surrounding country rock. Within the  
1067 intrusion, autometasomatism can be seen as hundreds of square meters of albitized,  
1068 analcimized and hematitized zones (Ferguson, 1964; Marks and Markl, 2015); min-  
1069 erallogically diverse hydrothermal veins (Engell et al., 1971; Markl, 2001; Markl and  
1070 Baumgartner, 2002); and secondary mineral assemblages of albite, aegirine, fluorite,  
1071 analcime, pectolite, catapleite, gittingsite and other HFSE and REE phases. These  
1072 features result from the release of aqueous fluids variably enriched in Na, Cl<sup>-</sup>, F<sup>-</sup>,  
1073 Ca and Sr during the later stages of crystallization (Markl and Baumgartner, 2002;  
1074 Graser and Markl, 2008; Borst et al., 2016). Fenitization of the granitic country rock  
1075 itself is limited however, extending no further than 120 m from the intrusion-country  
1076 rock contact (Ferguson, 1964).

## 1077 **6. Conclusions**

1078 Recognizing similarities between fenites related to mineralized intrusions is prob-  
1079 lematic due to the lack of detail reported. However, there is evidence of strong  
1080 trends between the complexity of fenite textures, such as multiple stages of vein-

1081 ing and mineral assemblages, and associated mineralization. This is due to Nb and  
1082 REE enrichment typically being discovered in intermediate and late stage carbon-  
1083 atite magma generations, each of that exsolve multiple pulses of fenitizing fluids.  
1084 The presence of brecciation in a carbonatite or alkaline complex also indicates the  
1085 explosive release of fluids and volatiles which exsolve during evolution, cooling and  
1086 crystallization of the magma. Therefore, brecciation indicates the presence of more  
1087 evolved magma generations and therefore an increased likelihood of Nb and REE  
1088 enrichment in the source intrusion.

1089 Although this manuscript compiles and reviews our current knowledge of fen-  
1090 ites and associated metasomatic processes, it also highlights gaps in the literature.  
1091 Many previous attempts have been made to categorize and name fenites, however it is  
1092 recommended that the IUGS metamorphic terminology be used to described predom-  
1093 inant mineralogy and textures. Alteration patterns and zones have been successfully  
1094 used as exploration tools to explore for mineralized systems such as porphyry copper  
1095 deposits in the past. Although fenites have large potential to be used as exploration  
1096 indicators to find new REE and Nb prospects, there are many areas of research that  
1097 still need to be addressed (Table 3) before fenites can be used as an integrated and  
1098 effective exploration tool.

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## 1741 9. Figure and table captions

1742 **Graphical Abstract:** Block diagram summarizing the predominant relation-  
1743 ships and timing between fenitization events and features as discussed in the liter-  
1744 ature reviewed (Heinrich and Moore, 1969; Woolley, 1969; Vartiainen and Woolley,  
1745 1976; Notholt, 1979; Woolley, 1982; Rubie and Gunter, 1983; Platt and Woolley,  
1746 1990; Williams-Jones and Palmer, 2002; Verschure and Maijer, 2005; Le Bas, 2008;  
1747 Doroshkevich et al., 2009; Hoatson et al., 2011; Moore et al., 2015).

1748 **Figure 1:** Classification of global REE deposits based on current exploitation  
1749 status and geological setting. Contains British Geological Survey materials ©NERC  
1750 [2017]. (Colour, 2 columns)

1751 **Figure 2:** Conceptual model of fenitization and brecciation associated with al-  
1752 kaline and carbonatitic magmas from Le Bas (1977). (Greyscale, 1 column)

1753 **Figure 3:** Geological map of the Fen Complex, Norway as a type site for feniti-  
 1754 zation. Map adapted from Verschure and Maijer (2005). (Colour, 1.5 columns)

1755 **Figure 4:** Progressive stages of fenitization and mineral assemblages at Fen,  
 1756 Norway as described by Brögger (1921). Intense fenitization is proximal to intrusion  
 1757 and unfenitized country rock, distal. (Greyscale, 1 column)

1758 **Figure 5: A:** Cathodoluminescence (CL) image showing zoned euhedral apatite  
 1759 (green-purple) with adjacent pyroxene vein (black) and calcite (yellow-orange). **B:**  
 1760 Veined fenite at Nkalonje, Malawi with hammer for scale. **C:** CL image showing  
 1761 growth of new feldspar (pink-red) during fenitization at Chenga, Malawi around  
 1762 a pre-existing feldspar core (grey) **D:** CL image of incipiently fenitized gneiss at  
 1763 Kangankunde, Malawi. Introduction of ferric iron along fractures by fenitizing fluids  
 1764 luminesces red in non-luminescent amphiboles and green unaltered feldspars. Fe-  
 1765 rich carbonate luminesces orange. **E:** Progressive pervasive sodic fenitization front  
 1766 metasomatizing greywackes at Okorusu, Namibia with pen for scale. **F:** XPL image  
 1767 of fracture used by multiple pulse of fenitizing fluid at Sokli, Finland forming a vein  
 1768 with multiple layers of minerals mirrored either side of vein. (Colour, 2 columns)

1769 **Figure 6:** Schematic diagram illustrating the difference between pervasive fen-  
 1770 itization (A) and veined fenitization (B). Fluid interacts with a larger volume of  
 1771 country rock during diffuse porous flow than fluid flowing through fracture. (Colour,  
 1772 1 column)

1773 **Figure 7: A:** Fenite breccia consisting of K-feldspar rich clasts at Chenga, pe-  
 1774 ripheral to Songwe Hill, Malawi. **B:** Carbonatite dyke intruding sodic fenite breccia  
 1775 at Lofdal, Namibia. **C:** K-feldspar rich fenite clasts incorporated within carbonatite  
 1776 intrusion at Songwe Hill, Malawi. **D:** Hand sample of glimmerite fenite adjacent  
 1777 to carbonatite intrusion at Bayan Khushu, Mongolia. **E:** SEM backscatter electron  
 1778 image of fenite sample from Songwe Hill, Malawi showing REE-fluorcarbonates (FC)

1779 and manganese oxides (MnO) in a matrix of Fe-rich carbonates and exsolved iron  
 1780 oxide (intergrown grey and white). **F:** SEM backscatter electron image of fenite sam-  
 1781 ple from Sokli, Finland showing pyrochlore (Py) in an aegirine vein (Ae) intruding  
 1782 albite (Ab). (Colour, 2 columns)

1783 **Figure 8:** Concentration of silica and total alkalis in fenites and their protoliths  
 1784 surrounding carbonatite complexes around the world. Fenites are divided into sodic,  
 1785 intermediate or potassic categories and arrows indicate general fenitization trends.  
 1786 All data available in Supplementary Table 1. **A:** Fenitization of gneiss protolith at  
 1787 Alnö, Sweden (Morogan, 1989). **B:** Fenitization of granitic gneiss at Fen, Norway  
 1788 (Kresten, 1988; Verschure and Majjer, 2005). **C:** Fenitization of gneiss and schist  
 1789 at Sokli, Finland (Vartiainen and Woolley, 1976; Al Ani and Sarapää, 2013). **D:**  
 1790 Fenitization of nephelinite at Amba Dongar, India (Viladkar, 2015); aplitic granite  
 1791 at Meech Lake, Canada (Hogarth, 2016); and granite at Silai Patti, Pakistan (Le Bas,  
 1792 2008). **E:** Fenitization of granite at Dicker Willem, Namibia (Cooper and Reid, 2000);  
 1793 gneiss and marble at Lac a la Perdrix, Canada (Hogarth and van Breemen, 1996); and  
 1794 Swartbooisdrif, Namibia (Drüppel et al., 2005). **F:** Fenitization of granite at Great  
 1795 Glen, Scotland (Garson et al., 1984); nephelinite at Koga, Pakistan (Le Bas, 2008);  
 1796 and granite gneiss at Newania, India (Viladkar and Wimmenauer, 1986). (colour, 2  
 1797 columns)

1798 **Figure 9:** Classification of amphiboles plotted on graphs of Mg:Fe<sup>3+</sup> ratio and  
 1799 Si calculated in tetrahedral position of formula using the scheme outlined in Leake  
 1800 et al. (1997, 2004). **A:** Calcic amphiboles with CaB  $\geq$  1.50, (Na + K)A < 0.50.  
 1801 **B:** Classification of calcic amphiboles with CaB  $\geq$  1.50, (Na + K)A  $\geq$  0.50. **C:**  
 1802 Classification of sodic-calcic amphiboles with (Na + K)A  $\geq$  0.50. **D:** Classification  
 1803 of sodic amphiboles with (Na + K)A < 0.50. **E:** Classification of sodic amphiboles  
 1804 with (Na + K)A  $\geq$  0.50. All data available in Supplementary Table 2. Data sources:

1805 Alnö, Sweden (Morogan and Woolley, 1988); Bayan Obo, China (Le Bas, 2008);  
 1806 Cantley, Canada (Hogarth and Lapointe, 1984); Fen, Norway (Kresten and Morogan,  
 1807 1986; Andersen, 1989); Great Glen, Scotland (Garson et al., 1984); Newania, India  
 1808 (Viladkar and Wimmenauer, 1986); Pollen, Norway (Robins and Tysseland, 1983);  
 1809 Silai Patti, Pakistan (Le Bas, 2008); and Sokli, Finland (Vartiainen and Woolley,  
 1810 1976). (Colour, 2 columns)

1811 **Figure 10:** Ternary diagram showing the proportions of end members analysed  
 1812 in pyroxenes from fenites surround carbonatite complexes around the world. All  
 1813 data available in Supplementary Table 3. Data sources: Alnö, Sweden (Morogan  
 1814 and Woolley, 1988); Bayan Obo, China (Le Bas, 2008); Cantley, Canada (Hogarth  
 1815 and Lapointe, 1984); Fen, Norway (Kresten and Morogan, 1986; Andersen, 1989);  
 1816 Great Glen, Scotland (Garson et al., 1984); Meech Lake, Canada (Hogarth, 2016);  
 1817 Okorusu, Namibia (Shivdasan, 2003); Pollen, Norway (Robins and Tysseland, 1983);  
 1818 Sarambi, Brazil (Haggerty and Mariano, 1983); Silai Patti, Pakistan (Le Bas, 2008);  
 1819 and Sokli, Finland (Vartiainen and Woolley, 1976). (Colour, 1 column)

1820 **Figure 11: A:** Ternary diagram plotting  $\text{Al}_2\text{O}_3\text{-Na}_2\text{O} + \text{K}_2\text{O-FeTot} + \text{MgO}$   
 1821 concentrations of fenites related to carbonatite complexes around the world, cate-  
 1822 gorized based on sodic, intermediate or potassic fenitization. **B:** Ternary diagram  
 1823 plotting  $\text{Al}_2\text{O}_3\text{-Na}_2\text{O} + \text{K}_2\text{O-FeTot} + \text{MgO}$  concentrations of fenites related to alka-  
 1824 line complexes around the world, categorized based on sodic, intermediate or potas-  
 1825 sic fenitization. All data available in Supplementary Tables 1 and 4. Data sources:  
 1826 Alnö, Sweden, (Morogan, 1989); Bayan Obo, China (Le Bas, 2008); Borralan, Scot-  
 1827 land (Woolley et al., 1972; Martin et al., 1978); Chilwa, Malawi (Woolley, 1969);  
 1828 Dicker Willem, Namibia (Cooper and Reid, 2000); Fen, Norway (Kresten, 1988; Ver-  
 1829 schure and Maijer, 2005); Kaiserstuhl, Germany (Wimmenauer et al., 1977); Iivaara,  
 1830 Finland (Kramm et al., 1993; Sindern and Kramm, 2000); Ilmeny-Vishnevye Gory,

1831 Russia (Dvornik, 2015 - unpublished data); Ishimovsky, Russia (Dvornik, 2015 - un-  
1832 published data); Kangunkunde, Malawi (Woolley, 1969); Khibiny, Kola Peninsula  
1833 (Arzamastev et al., 2011); Koga, Pakistan (Le Bas, 2008); Lovozero, Kola Peninsula  
1834 (Arzamastev et al., 2011); Murun, Russia (Dvornik, 2015 - unpublished data); Oz-  
1835 ernaya Varaka, Kola Peninsula (Kozlov and Arzamastsev, 2015); San Vicente, Cape  
1836 Verde Islands (Le Bas, 2008); Silai Patti, Pakistan (Le Bas, 2008); Sokli, Finland  
1837 (Vartiainen and Woolley, 1976; Al Ani and Sarapää, 2013); Ryabinovskiy, Russia  
1838 (Dvornik, 2015 - unpublished data); Turiy Mys, Kola Peninsula (Evdokimov, 1982);  
1839 Wasaki, Kenya (Le Bas, 2008). (Colour, 2 columns)

1840 **Figure 12:** Schematic diagram illustrating the process of autometasomatism.  
1841 Crystallizing melt within the magma chamber or intrusion expels alkali-rich fluid  
1842 that migrates through the periphery of already crystallized alkaline rock, autometa-  
1843 somatizing primary magmatic crystals. As a result, fenitization of country rock is  
1844 limited due to restricted flow out of intrusion. (Greyscale, 1 column)

Complex	Country	Location	Carbonatite Age Types	Country Rock	Fenite Size	Fenite Characteristics	Fenite Mineralogy	Mineral-References
<b>Alnö</b>	Sweden	62.4500, 17.1833	Calcite, dolomite: di-atrems, sheet dykes	Gneisses	500-600 m	Sodic, intermediate, potassic, pseudo-trachytes, 6 % volume loss	Albite, K-feldspar, arfvedsonite, aegirine-augite	Morogan (1989); Skelton et al. (2007)
<b>Amba Dongar</b>	India	21.9875, 74.0617	Calcite, ankerite, siderite: diatrems, ring dykes	Quartzitic sandstone	<300 m	Deeper sodic, shallower potassic	Sodic: aegirine-augite, feldspar; Potassic: K-feldspar, albite.	Doroshkevich et al. (2009)
<b>Araxá</b>	Brazil	-19.6333, -46.9333	Dolomite, calcite: intrusions, veins	Quartzites, schists	2.5 km	Glimmerite from fenitization of pyroxenites		Traversa et al. (2001); Mitchell (2015)
<b>Bayan Obo</b>	China	41.80194, 109.9900	Dolomite: intrusions	Meta-sandstones, dolomite marbles	2-4 m	Early sodic, later potassic	Sodic: amphibole, albite, iron oxides, microcline. Potassic: K-feldspar cut by albite veins	Le Bas (2008); Xu et al. (2008); Wall (2014)
<b>Bear Lodge</b>	USA	44.4833, 104.4500	- Calcite: veins and intrusions in diatremes	45 Granite		Glimmerite, potassic		Moore et al. (2015)
<b>Chipman Lake</b>	Canada	49.9667, 86.2000	- Dolomite, ankerite: intrusions in stockwork	Granites, metavolcanic greenstones	Broad	Early sodic, later potassic	Sodic: alkali feldspar, amphiboles, bonate. Potassic: phlogopite, alkali-amphiboles.	Platt and Woolley (1990)
<b>Dicker Willem</b>	Namibia	-26.4667, 16.0167	Calcite: cone sheets, intrusions, plugs, breccia pipes	49 Granitoids, granite gneisses		Breccia, pseudo-trachytes, potassic	Orthoclase, hematite, calcite	Reid and Cooper (1992); Cooper and Reid (2000)

Complex	Country	Location	Carbonatite Types	Age	Country Rock	Fenite Size	Fenite Characteristics	Fenite Mineralogy	Mineral-References
<b>Fen</b>	Norway	59.2716, 9.3066	Calcite, dolomite, siderite: ring complex, intrusions, veins, intrusions in diatreme	Neoproterozoic 578 Ma	Granitic gneiss	>1.5 km	Breccia, potassic, sodic. At least 2 alkali stages.	Potassic: mica, amphibole, apatite. Sodic: feldspar, aegirine-augite, sodic-amphiboles (Magnesioarfvedsonite, riebeckite, ferrosichterite).	Kresten and Morogan (1986); Andersen (1989); Dahlgren (1994); Ver-schure and Majer (2005)
<b>Kangunkunde</b>	Malawi	-15.1240, 34.9103	Dolomite, ankerite: vent, veins	Cretaceous: 136-123 Ma	Amphibolites, quartz reefs, granitic pegmatites	>1 km	Breccia, potassic, sodic	Orthoclase, gioclase, amphiboles (riebeckite, magnesioarfvedsonite, crossite, tremolite), aegirine, calcite, ilmenite, phlogopite	Woolley (1969)
<b>Lofdal</b>	Namibia	-20.3500, 14.7500	Calcite, dolomite, ankerite: plutonic, dyke swarm, diatremes	Proterozoic: 765 Ma	Gneisses, schists	>3 m	Breccia, potassic, sodic	K-feldspar, albite, calcite	Wall et al. (2008); Bodeving et al. (2017)
<b>Mountain Pass</b>	USA	35.4833, 115.5333	- Calcite, dolomite, ankerite: intrusions	Proterozoic: 1375 Ma	Schists, gneisses	>200 m	Breccia, potassic	K-feldspar, phlogopite, carbonate, magnesio-riebeckite	Castor (2008)
<b>Mt Weld</b>	Australia	-28.8635, 122.5475	Calcite, dolomite: vent, stocks, intrusions	Proterozoic: 2025 Ma	Sedimentary- volcanic sequences	500 m	Breccia, glimmerite	Phlogopite-rich	Hoatson et al. (2011)

Complex	Country	Location	Carbonatite Age Types	Country Rock	Fenite Size	Fenite Characteristics	Feniteogy	Mineral-References
<b>Okorusu</b>	Namibia	-20.0331, 16.7667	Calcite: plu-tonic, intrusions Cretaceous: 137-124 Ma	Greywackes, quartzites, schists, limestones	>1 km	Breccia, potassic, sodic, pyroxenite	Potassic: feldspar, aegirine-augite, calcite, biotite, sodic-amphiboles, albite	K- Böhn et al. (2001, 2002)
<b>Sokli</b>	Finland	67.7973, 29.3221	Calcite, dolomite: pipe, intrusions, veins in stockwork Devonian: 370-360 Ma	Gneisses, amphibolites, schists, ultramafics	1-2 km >30 km <sup>2</sup>	Breccia, potassic, sodic	Amphibole, gopite, carbonate	Appleyard and Woolley (1979); Vartiainen and Paarma (1979); Al Ani and Sarapää (2009); Sarapää et al. (2013)
<b>Songwe Hill</b>	Malawi	-15.6897, 35.8153	Calcite, ankerite: vent, intrusions	Granulites, gneisses	>1 km	Breccia, potassic, sodic (Chenga)	Potassic: feldspar, aegirine-augite, sodic-amphiboles, albite.	K- Croll et al. (2015)
<b>Tundulu</b>	Malawi	-15.5391, 35.8069	Calcite, ankerite, siderite: pluton, intrusions, vents, possible diatreme Cretaceous: 133 Ma	Granulites, gneisses, granites	2-3 km	Breccia, potassic, sodic	Potassic: ortho-clase, microcline, aegirine, sodic-amphiboles, albite, orthoclase, calcite.	Garson (1965); Ngwenya (1994); Woolley (2001); Broom-Fendley et al. (2016)
<b>Wasaki Peninsula</b>	Kenya	-0.4667, 34.3833	Calcite, siderite: trusions, veins, diatreme Miocene: in-16-13 Ma	Granodiorite	200-300 m	Breccia, potassic	Potassic: thoclase, albite, aegirine-augite.	Woolley (2001); Le Bas (2008)

Table 1: Summary of fenite characteristics for key carbonatite complexes used in this manuscript. Fenite size describes distance from carbonatite to outer detected limit of fenite.

Complex Commodity	Carb enrichment	Fen enrichment	Fen characteristics	Fluid chemistry	Notes	References
<b>Alnö</b>	REE <1463 ppm	Potassic: 102-499 ppm TREE	Sodic, intermediate, pseudotachytes, 6 % vol. loss	PO <sub>4</sub> <sup>3-</sup> , CO <sub>3</sub> <sup>2-</sup> , HCO <sub>3</sub> <sup>-</sup> , F <sup>-</sup> rich, 640 °C	Increasing fractionation with decreasing fenite intensity	Morogan (1989); Hornigton et al. (1998); Skelton et al. (2007)
<b>Amba Dongar</b>	REE, F 15 wt.% TREE	HREE, 11.6 Mt at 30 wt.% CaF <sub>2</sub> potassic	Deeper sodic, shallower potassic	F-rich, 600-700 °C	Fenite Nd/La ratio >carbonatite ratio	Deans et al. (1972); Mariani (1989); Palmer and Williams-Jones (1996); Doroshkevich et al. (2009)
<b>Araxá</b>	Nb 1.6 wt.%	Laterite: 462 Mt at 1737-16906 ppm TREE	Glimmerite		Supergene laterite enrichment of Nb	Traversa et al. (2001); Mitchell (2015)
<b>Bayan Obo</b>	REE 750 Mt at 4.1 wt.% REO		Early sodic, later potassic	Fenite amphiboles and biotites enriched in F		Smith (2007); Le Bas (2008); Xu et al. (2008); Wall (2014)
<b>Bear Lodge</b>	REE, Gold 13.3 Mt at 3.2 wt.% TREE (Bull Hill Diatreme)	LREE	Glimmerite, potassic	F <sup>-</sup> , SO <sub>4</sub> <sup>2-</sup> , MREE rich		Andersen et al. (2013); Moore et al. (2015)
<b>Chipman Lake</b>	REE 1-17 % in early bankite		Early sodic, later potassic	Early Na and F-rich fluid		Platt and Woolley (1990)
<b>Dicker Willem</b>		32-352 ppm Nb, 168-515 ppm TREE	Breccia, potassic, pseudotachytes	HCO <sub>3</sub> <sup>-</sup>	Low temperature hydrothermal overprint	Reid and Cooper (1992); Cooper and Reid (2000)
<b>Fen</b>	REE, Nb 1.03-4 wt.% TREE, 0.35-0.45 wt.% Nb <sub>2</sub> O <sub>5</sub>	385-585 ppm TREE	Breccia, potassic, least 2 stages.	CO <sub>3</sub> <sup>2-</sup> , PO <sub>4</sub> <sup>3-</sup> , F <sup>-</sup> rich	Increasing REE fractionation with decreasing fenite intensity	Lie Andersen (1986); Lie and Østergaard (2011); Mitchell (2015)
<b>Kangunkunde</b>	REE 0.37-10 wt.% TREE		Breccia, potassic, sodic	F <sup>-</sup> , PO <sub>4</sub> <sup>3-</sup> , Cl <sup>-</sup> rich		Woolley (1969); Woolley and Kempe (1989); Wall et al. (2008); Downman (2014)

Complex Commodity	Carb enrichment	En-Fen enrichment	Fen enrichment	Enrichment	Characteristics	Fluid chemistry	Notes	References
Lofdal	REE 0.43-0.64 wt.% TREE, HREE enriched	10-25 HREO + Y, 1393 ppm Nb	% Breccia, potassic, sodic		Breccia, potassic, sodic	Early rich contains LREE, later HREE	F- fluid late-stage drothermal activity	Wall et al. (2008); Bodeving et al. (2017)
Mountain Pass	REE 20 Mt at 8.9 wt.% TREE	<9 wt.% TREE			Breccia, potassic			Castor (2008)
Mt Weld	REE, Nb, P 17.5 Mt at 8.1 wt.% TREE; 37.7 Mt at 1.07 wt.% Nb2O5; 77 Mt at 13.5 wt.% P2O3				Breccia, glimmerite		Supergene laterite enrichment	Hoatson et al. (2011)
Okorusu	F, REE, P 0.2 wt.% TREE; 1-3 wt.% P2O5	80-285 ppm REE in fluorite; Mt at 61% CaF2; Mt at 49% CaF2			Breccia, potassic, sodic, pyroxenite	Up to 3 wt.% REE and 3 wt.% F. stage fluids HCO3-		Bühn et al. (2001, 2002)
Sokli	REE, P 0.1-1.83 wt.% TREE	1-2 wt.% TREE			Breccia, potassic, sodic		Late stage dykes have highest REE potential	Al Ani and Sarapää (2013)
Songwe Hill	REE 8.4 Mt at 1.6 wt.% TREE	1.37 Mt at 1.61 wt.% TREE			Breccia, potassic, sodic (Chenga)		Late stage black carbonatite av. 3.7% TREE	Croll et al. (2015)
Tundulu	REE, P 2.3 Mt at 12.5 wt.% P2O5; Mt at 2.4 wt.% TREE (Nathace Hill)				Breccia, potassic, sodic	Acidic, containing F, P, Cl	Late stage HREE enrichment	Ngwenya (1994); Broomfield et al. (2016); Kruger (2016)
Wasaki Peninsula	La, Ce 322 ppm La, 957 ppm Ce				Breccia, potassic			Bowden (1985); Woolley (2001); Le Bas (2008)

Table 2: Summary of critical metal enrichment in carbonatite and fenite at complex examples used in manuscript.

Number	Question
1	Why do carbonatites release Na-rich and K-rich fluids at different times?
2	How long does it take for a carbonatite to cool, and how does this affect fenitization?
3	How does the fenitization style i.e. pervasive / veined, affect the timescale of metasomatism?
4	Can we use diffusion modelling on rimmed crystals e.g. feldspars and aegirine-augites to better define fenitization timescales?
5	How do fluids evolve during progressive fluid-rock interactions, and what implications does this have for fenitization further from the intrusion?
6	What spatial variations can be seen within fenites? E.g. vein composition proximal vs distal, ratio of LREE:HREE with distance?
7	Are carbonate-rich veins in fenites true carbonatite veins or precipitation from hydrothermal fluids? How can the difference be identified and where is the line drawn?
8	Where do we find REE and Nb mineralization within the fenites? Within veins, adjacent to veins or within pervasive fenites?
9	Does fenitization occur at a lava-country rock contact?
10	Can we estimate the amount of REE or Nb present in the carbonatite, based upon their concentrations in the associated fenite?
11	Does K and Na fenite separation occur at each stage of carbonatite or alkaline magmatism?
12	Can a magmatic syenite be distinguished from a syenite formed by fenitization?

Table 3: Summary of some key highlighted gaps in knowledge and literature in regards to fenites and the process of fenitization