



# Columbitization of fluorcalciopyrochlore by hydrothermalism at the Saint-Honoré alkaline complex, Québec (Canada): New insights on halite in carbonatites

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## ABSTRACT

Niobium (Nb) in carbonatite is mainly hosted in fluorcalciopyrochlore and columbite-(Fe). Information related to Nb petrogenesis is useful for understanding the processes related to Nb mineralization and carbonatite evolution. The Saint-Honoré, Quebec, alkaline complex offers a rare opportunity for studying these processes as the complex is not affected by post-emplacement deformation, metamorphism nor weathering. Columbite-(Fe) is shown to be an alteration product of fluorcalciopyrochlore (columbitization). Columbitization is characterized by the leaching of Na and F from the A- and Y-sites of the pyrochlore crystal structure. As alteration increases, Fe and Mn are slowly introduced while Ca is simultaneously leached. Leached Ca and F then crystallize as inclusions of calcite and fluorite within the columbite-(Fe). A-site cations and vacancies in the crystal structure of fresh and altered pyrochlores demonstrate that pyrochlore alteration is hydrothermal in origin. Moreover, halite is a ubiquitous mineral in the Saint-Honoré alkaline complex. Petrographic evidence shows that halite forms in weakly altered pyrochlores, suggesting halite has a secondary origin. As alteration increases, halite is expelled by the hydrothermal fluid and is carried farther into the complex, filling fractures throughout the carbonatite. The hydrothermal hypothesis is strengthened by significant enrichments in Cl and HREEs in columbite-(Fe). Chlorine is most likely introduced by a hydrothermal fluid that increases the solubility of REEs.

## 1. Introduction

Carbonatites are important rocks for understanding the Earth's evolution as they provide a window into the mantle and its dynamics. As such, they have been abundantly studied (Chakhmouradian et al., 2015; Decrée et al., 2015; Le Bas, 1981; Mitchell, 2015; Wyllie, 1966; among many others). However, the crustal evolution of carbonatites, either by fractional crystallization, hydrothermal activity, carbothermalism or weathering can obliterate or modify much of the initial information recorded by these rocks. Thus, an understanding of the evolution of carbonatites is essential to better constrain any interpretation of their formation. Moreover, carbonatites are important economic rocks as they host strategic metals such as rare earth elements (REEs) (Chakhmouradian and Wall, 2012; Giebel et al., 2017), Nb (Mariano, 1989; Wall et al., 1999) and, in some cases, base metals (e.g. Cu; Heinrich, 1970). A rapidly increasing demand for Nb (Roskill, 2017) in emerging countries (Mackay and Simandl, 2014), requires a better understanding of the mineralization processes within carbonatites to develop avenues of possible exploration. The petrography and

chemistry of columbite and pyrochlore are often key for understanding the genesis of Nb mineralization.

Within carbonatites, minerals from the pyrochlore group host Nb mineralization and act as recorders of carbonatite petrogenesis (Atencio et al., 2010; Hogarth et al., 2000; Lumpkin and Ewing, 1995). The pyrochlore mineral group contains more than a dozen species although fluorcalciopyrochlore [(Ca,Na)<sub>2</sub>(Nb,Ti)<sub>2</sub>O<sub>6</sub>(O,OH,F)] (Hogarth, 1977) is the end-member that is usually exploited for Nb. A second economically important mineral in carbonatite is columbite-(Fe) [(Fe,Mn)(Nb,Ti)<sub>2</sub>O<sub>6</sub>]. Columbite is generally found as a primary mineral in granites and pegmatites (e.g. Cerný, 1989; Lumpkin, 1998). It is generally uncommon in carbonatites except for a few occurrences as a secondary mineral (Mackay and Simandl, 2015; and references therein); its presence as a primary mineral is rare (Mariano, 1989). Columbite can be a primary or an alteration product from pyrochlore through an igneous or hydrothermal event (Chakhmouradian et al., 2015; Heinrich, 1966; James and McKie, 1958; Mariano, 1989). James and McKie (1958) were the first to describe the alteration process from pyrochlore to columbite in carbonatite, later named columbitization

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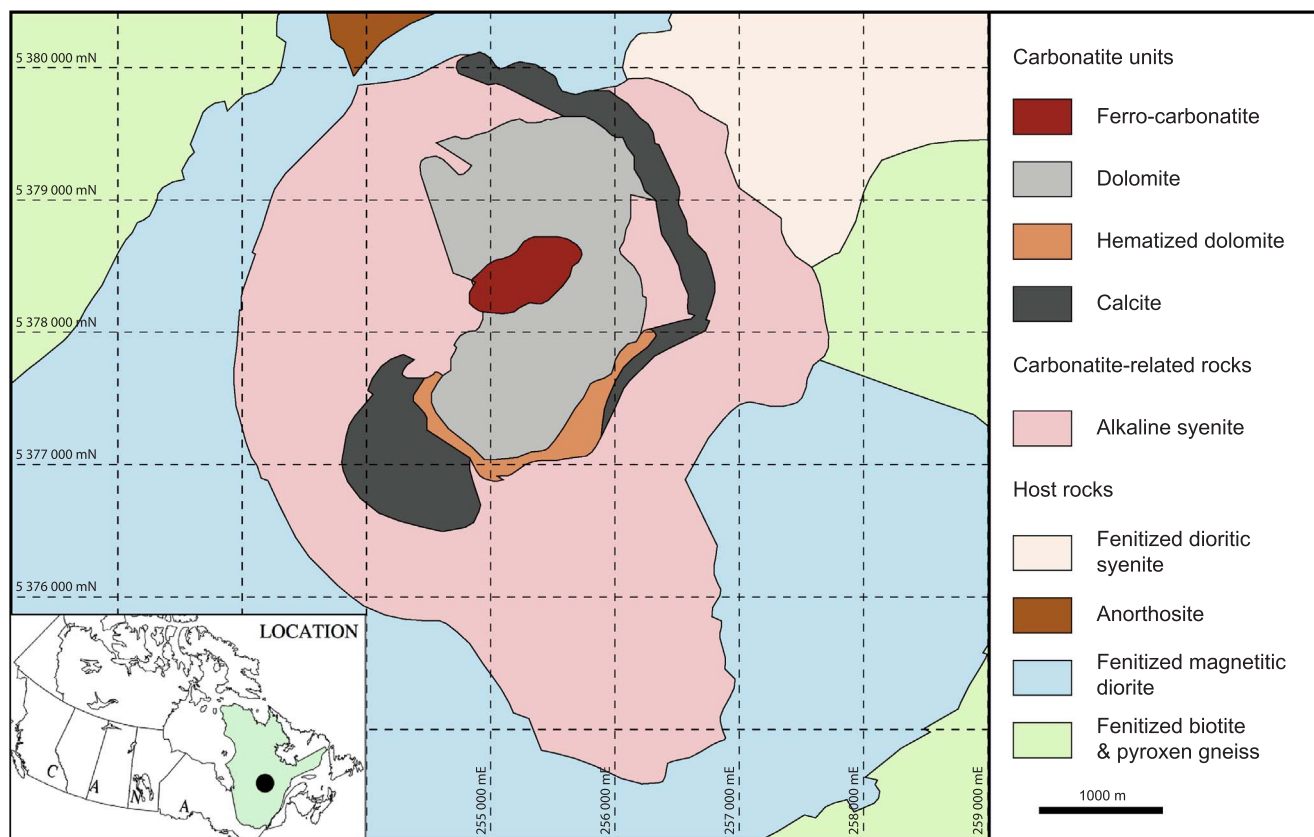


Fig. 1. Simplified geological map of the Saint-Honoré alkaline complex (modified from Vallières et al., 2013). The economically Nb-bearing unit is the dolomitic carbonatite unit.

(Heinrich, 1966). Alteration of pyrochlore has been studied recently (Chakhmouradian et al., 2015; Cordeiro et al., 2011; Lumpkin and Ewing, 1995; Nasraoui and Bilal, 2000; Mitchell, 2015; Wall et al., 1996) in lateritic and relatively fresh carbonatites. These works highlight an origin of columbite from the alteration of pyrochlore, although none of the studies showed the conservation of all major elements between pyrochlore and columbite-(Fe), minus the release of Na.

The economic viability of a Nb exploitation is influenced by the variation in size, shape and chemistry of Nb-bearing minerals as well as by the distribution of different Nb-bearing phases within a deposit. Therefore, a thorough understanding of the mechanisms that control Nb-hosting phases and their alteration are crucial for a comprehension of mineralization associated with carbonatites and potential causes of metallurgy issues, such as the alteration of minerals that can hinder economic exploitation.

The Saint-Honoré alkaline complex is an ideal setting for the study of carbonatites and their related Nb-hosting minerals as it is currently exploited and accessible to a depth of 808 m (2650 feet). Studies regarding mineralization (Fortin-Bélanger, 1977; Thivierge et al., 1983) have been conducted in the weathered upper portion of the carbonatite and more recent studies of the Saint-Honoré carbonatite focused on REE mineralization (Fournier, 1993; Grenier et al., 2013; Néron, 2015; Néron et al., 2013) or the origin of ubiquitous halite (Kamenetsky et al., 2015). The main minerals exploited for Nb are fluorcalciopyrochlore (using the pyrochlore classification of Atencio et al. (2010)) and columbite-(Fe). There are four other pyrochlore species (e.g. Sr, Th or U-rich) present in the Saint-Honoré carbonatite (Belzile, 2009; Clow et al., 2011), but they are of minor importance.

Columbite from the Saint-Honoré carbonatite is part of the iron end-member and hence is classified as columbite-(Fe) (Burke, 2008) (previously named ferrocolumbite). With depth, columbite-(Fe) increases in abundance, becoming a major Nb-bearing mineral. This pattern with depth provides new insights on the genesis of carbonatites, but also

presents extractive metallurgy issues for exploitation. In this study, the petrogenesis of pyrochlore and columbite-(Fe) is investigated. The puzzling presence of halite grains observed in minute cavities of weakly altered pyrochlores provides clues about sodium remobilization in carbonatites and the contribution of halite to fenitization.

### 1.1. Geological setting

The Saint-Honoré alkaline complex is located in the Saguenay region, Quebec (Canada). The regional bedrock is the Canadian Shield and is mainly composed of Mesoproterozoic rocks of the Grenville Province (Dimroth et al., 1981; Higgins and van Breemen, 1996). Dimroth et al. (1981) divided the geological province into three units: 1) a gneiss complex that was deformed and migmatized during the Hudsonian Orogeny (1735 Ma); 2) anorthosite and charnockite-magnetite batholiths dating from pre- to post-Grenville orogeny (935 Ma); and 3) calc-alkaline intrusions that cross-cut the host rocks. This late stage, the Iapetan rift system, is related to alkaline activity (Kumarapeli and Saull, 1966) and includes the intrusion of the Saint-Honoré alkaline complex.

The alkaline complex is composed of a crescent-shaped carbonatite surrounded by alkaline silicate rocks. The host rocks were fenitized by the emplacement of the complex and the fenitization is characterized by sodic-amphiboles, aegirine, sericitized plagioclases as well as green and red carbonate veins (Fortin-Bélanger, 1977). Silicate rocks are represented by three types of syenites: alkaline syenite, nepheline-bearing syenite and syenite foidolites (ijolite-urtite). The presence of xenoliths of altered syenites in the carbonatite suggests silicate rocks are older than the carbonatite. K-Ar dating of the alkaline complex gave an age of 565 Ma (Doig and Barton, 1968). McCausland et al. (2009) report an Ar-Ar age of  $571 \pm 5$  Ma from phlogopite and Kamenetsky et al. (2015) report a two-point Rb-Sr model age of  $564 \pm 8$  Ma. More recently, baddeleyite from lamprophyre dikes associated with the Saint-

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