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Unusual evolution of silica-under- and -oversaturated alkaline rocks in the Cenozoic Ambohimirahavavy Complex (Madagascar): Mineralogical and geochemical evidence

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ABSTRACT

The almost unknown Ambohimirahavavy ring complex in the Cenozoic alkaline province of northwestern Madagascar has recently attracted considerable interest because of the discovery of important rare-metal mineralization. The complex consists of arc-shaped bodies made up of silica-under- and -oversaturated syenites and extremely evolved peralkaline granitic dykes, as well as several mafic to felsic volcanic units, including basalt, phonolite and trachyte, all of which have an alkaline affinity. Uranium–lead zircon ages of 24.2 ± 0.6 Ma and 23.5 ± 6.8 Ma have been obtained for nepheline syenites and peralkaline granitic dykes, respectively, which, together with field data and ages of neighboring complexes, support emplacement controlled by regional lithospheric structures, rather than an evolving hot spot.

Whole-rock major and trace-element and Sr–Nd isotopic data for the mafic suite suggest that the parental melt of this complex was generated by low degrees of melting of a metasomatized mantle source with residual amphibole. Fractional crystallization of this alkali basaltic melt likely produced the silica-undersaturated suite. We propose that the silica-oversaturated suite evolved from the undersaturated melt after contamination of the latter by crustal material. Further evolution to peralkaline compositions in both suites is attributed mainly to plagioclase and alkali feldspar segregation.

Nepheline and feldspar compositions, as well as considerations of mineral equilibria among mafic silicates and Fe–Ti oxide minerals indicate crystallization temperatures of 1000 to 700 °C and an oxygen fugacity of 0.4 to 0.8 log units below the fayalite–magnetite–quartz (FMQ) buffer at 1 kbar for the silica-undersaturated melt, and temperatures of 860 to 570 °C and an oxygen fugacity of 1.5 to 3.8 log units below FMQ for the oversaturated syenitic melt. The undersaturated melt evolved towards a more peralkaline composition. Crystallization of arfvedsonite plus aegirine further reduced the melt the evolution of which ended with fluid exsolution. At late stages of crystallization, the oversaturated melt departed from the reducing trend of the undersaturated melt, evolving towards high oxygen fugacity. Very late exsolution of the fluid permitted concentration of the HFSE in the last stages of magmatic evolution with local production of low-temperature pegmatitic phases extremely enriched in these elements.

1. Introduction

In response to an increasing demand by the high-tech industry, exploration for rare metals has received renewed interest in the past few years. Alkaline rocks are known to be major hosts for rare metals, and some of them have been subject to in-depth studies, for example, Ilímaussaq in Greenland (Sørensen, 1992), Strange Lake and Thor Lake in Canada (Salvi and Williams-Jones, 2006; Sheard et al., 2012), the Kola alkaline province (Kogarko et al., 2010), Khaldzan Buregte and Khan Bogd in Mongolia (Kempe et al., 1999; Kynicky et al., 2011). Nevertheless, research aimed to understand the petrological mechanisms that produce such rare-metal-rich magmas is still in its infancy.

Several field and experimental studies have attempted to clarify the origin and nature of the parental magmas of alkaline rocks; invariably, these invoke a mantle source that produced the magmas directly, or

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through evolution by crystal fractionation (Kramm and Kogarko, 1994; Larsen and Sørensen, 1987; Sørensen, 1997). More recently, Pilet et al. (2002) proposed partial melting of a metasomatized mantle source beneath the alkali provinces, and Marks et al. (2011) highlighted the potential role of alkali basaltic and nephelinitic melts in the evolution of peralkaline rocks. However, the processes that control the evolution of alkaline melts during their ascent are not well understood, partly because petrological studies of complexes with a wide spectrum of compositions are rare. Detailed geochemical studies of their phenocrysts, in particular reversely zoned clinopyroxenes, have helped shed some light on these issues, pointing to processes such as crustal assimilation, magma mixing, or several evolutionary stages at different P, T and oxygen fugacity in intermediate magma chambers (Duda and Schmincke, 1985).

A feature that is common to many alkaline igneous complexes worldwide, and is poorly understood, is the close spatial association of silica-undersaturated and -oversaturated rocks. In many cases, field relations indicate a common source from a single parental magma (i.e., a cogenetic origin) (Foland et al., 1993; Riishuus et al., 2008). The explanation that is generally invoked for the formation of silica-oversaturated melts is assimilation of Si-rich crustal material by undersaturated melts together with fractional crystallization (AFC) (Brooks and Gill, 1982; Foland et al., 1993). Understanding the latest phases of crystallization, in particular those leading to the formation of peralkaline rocks, has received particular attention in recent years (Mann et al., 2006; Markl et al., 2010; Marks et al., 2003). Among other things, these authors have recognized the tight link between excess alkali content and oxygen fugacity and their role in controlling the waning stages of melt crystallization to produce such evolved rocks.

The Ambohimirahavavy alkaline complex (AAC) is a very poorly known alkaline ring complex, the largest of several similar complexes in the Ampasindava peninsula of northern Madagascar (Bésairie, 1936; Donnot, 1963; Lacroix, 1915). It is a rare example in which a wide variety of lithologies coexist, including mafic to felsic, silica-undersaturated syenites to -oversaturated and extremely evolved peralkaline pegmatites. In addition, this complex has been recently recognized as potentially valuable for its mineralization in rare elements, in particular, Zr, Nb and the rare-earth elements (REE) (Estrade et al., 2014; Ganzeev and Grechishchev, 2003; Rakotovao et al., 2009), and at present, is at an advanced stage of exploration for the REE by the Tantalus Rare Earths AG company.

In this paper, we investigate the magmatic evolution of this complex. In particular, we: (1) provide precise ages and ascertain the relationship of the complex to regional volcanism and structures; (2) determine the nature of the source and (3) evaluate the role of mixing of differently evolved melts; (4) discuss the parallel formation of silica-undersaturated and -oversaturated melts and propose a model to explain the late peralkaline evolution of the complex; and (5) calculate T–fO_2 paths for the crystallization of the silica-undersaturated and -oversaturated rock suites.

2 . Geological setting and field relations

Most of the island of Madagascar comprises a Precambrian shield basement, which, in the western part, is covered by Mesozoic to Cenozoic sedimentary units deposited during and after separation of Madagascar from the African continent (De Wit, 2003). Two major episodes of volcanic activity have affected the island. Flood basalts of tholeiitic affinity were mostly erupted during the Upper Cretaceous, at the end of the separation of Madagascar from India, and overlie the Precambrian basement along the eastern coast of Madagascar (De Wit, 2003). A second period of volcanic activity occurred during the Cenozoic, when several volcanic complexes of alkaline affinity were emplaced in the north of the island (Melluso et al., 2007a).

The AAC is a Cenozoic annular complex about 18 km in length, located in the Ampasindava peninsula of northwestern Madagascar. It is part of a NW–SE alignment of alkaline complexes intruded in Mesozoic sedimentary units of the Isalo Group (Fig. 1a). The latter is a mixed association of alternating mudstone and marl, locally capped by disrupted limestone blocks, deposited between the lower and middle Jurassic (Papini and Benvenuti, 2008). The AAC was first described by Lacroix (1902), who recognized the Ampasindava region as the most interesting petrographic province known in his time. Detailed petrographic descriptions and numerous chemical analyses are available in the three volumes "Minéralogie de Madagascar" (Lacroix, 1922a,b, 1923). Further work was done by Bésairie (1936) and Donnot (1963). Woolley (2001) provided a brief review of the alkaline rocks occurring in Madagascar.

The AAC consists of a diverse assemblage of intrusive and extrusive rocks. Intrusive units include nepheline syenites, alkali feldspar syenite, quartz alkali feldspar syenite, a biotite granite and a peralkaline granite. Syenitic rocks form two ring-shaped bodies that mark the northwestern and southeastern limits of the complex (Fig. 1b). Our focus is the southeastern part of the complex, partly because it is accessible due to the exploration work of Tantalus Rare Earths AG and partly because this is where the rocks are most varied. The northwestern body consists of several different syenitic centers associated with biotite granite and has the highest relief of the complex. The intrusive body outcropping in the southeast is a cylindrical intrusion with a diameter of ~7 km. Syenites form a high relief ring-shaped exposure (ring-dyke) around a central depression, interpreted as a caldera. Although it is difficult to clearly identify contacts between syenite units, nepheline syenite seems to be confined to the northern innermost part of the ring-dyke. The latter is surrounded by discontinuous peralkaline granitic dykes (PGD) grouped in parallel layers of up to 10 m wide, intruded in mudstone and limestone of the Isalo Group (Fig. 2a). Locally, the PGD cross-cut the external part of the syenitic ring-dyke (Fig. 2b) and are interpreted to be the youngest rocks of the complex (Donnot, 1963; Ganzeev et al., 1989).

Volcanic rocks cover most of the central part of the complex but also occur as dykes intruded in sedimentary units. They consist mainly of heterogeneous volcanic breccias (Fig. 2c) and trachyte plus less abundant peralkaline phonolite. The dykes generally have microgranular textures and are intermediate in grain-size between trachyte and syenite. Two post-caldera domes composed of pitchstone dominate the central part of the caldera (Figs. 1b and 2d). Mafic and intermediate volcanics are less abundant, and occur as dykes injected throughout the complex and in Mesozoic sedimentary units of the Isalo Group (Fig. 2e). They consist mainly of camptonite, alkali basalt, hawaiite and mugearite. It is common to see syeno-trachytic dykes and mafic volcanic dykes closely related in the same dyke system (Fig. 2f).

3 . Sampling and analytical methods

Outcrop data and representative samples were collected during extensive fieldwork carried out over three field seasons by several members of our team. Although a large set of outcrop samples and drill core was collected, only representative specimens are reported for each lithology (see Fig. 1b for location). Rock preparation was carried out in the Laboratoire des Mines in Antananarivo and at the Géosciences Environnement Toulouse (GET) Laboratory, University of Toulouse. Polished sections were studied under transmitted and reflected light as well as by scanning electron microscopy (SEM), in order to determine mineral associations and parageneses. Back-scattered electron (BSE) images were collected with a Jeol JSM 6360LV SEM coupled with silicon

Fig. 1. (a) Geological map of the major alkaline complexes in the Ampasindava area. Modified after carte géologique de Madagascar (2012). BB: Bemarivo block, AB: Anaboriana block. (b) Geology of the Ambohimirahavavy alkaline complex. Sample locations are indicated by white squares.