

METASOMATISM AT A GRANITIC PEGMATITE – DUNITE CONTACT IN GALICIA: THE FRANQUEIRA OCCURRENCE OF CHRYSOBERYL (ALEXANDRITE), EMERALD, AND PHENAKITE: DISCUSSION

GERHARD FRANZ

Technische Universität Berlin, Fachgebiet Petrologie, 10623 Berlin, Germany

H. ALBERT GILG, GÜNTER GRUNDMANN AND GIULIO MORTEANI

Technische Universität München, Lehrstuhl für Angewandte Mineralogie und Geochemie, 85747 Garching, Germany

INTRODUCTION

Martin-Izard *et al.* (1995) described the occurrence of beryl (gem variety emerald), chrysoberyl (gem variety alexandrite) and phenakite at Franqueira, a recently discovered Be-mineral deposit in the Hercynian fold belt of Galicia, in northwestern Spain. They interpreted the formation of this mineral association according to the classical theory of "exometasomatism" (Fersman 1929, Leitmeier 1937, Sinkankas 1989). This exometasomatism is considered to be the result of the intrusion and crystallization of a Be-rich granitic pegmatite-forming magma and its fluid phase into ultramafic rocks. We want to take the description of this new Be-mineral occurrence as an opportunity to draw attention to an alternative genetic model about the formation of Be-minerals, especially emerald, chrysoberyl and phenakite, which was presented in detail by Grundmann & Morteani (1989) and Nwe & Morteani (1993). In this model, a regional multistage tectonic and metamorphic event is essential for the formation of beryl (gem variety emerald) at the contact between Cr-rich ultramafic rocks and Be-bearing K–Na–Al-silicate rocks. We believe that in general, the importance of metamorphic processes in the genesis of mineral deposits has not received

enough attention. Admittedly, we are not familiar with the geology of the Franqueira deposit; nevertheless, the data presented by Martin-Izard *et al.* (1995) are sufficient for such a discussion.

GEOLOGICAL SETTING – PETROGRAPHY – GEOCHEMISTRY

The Be mineralization at Franqueira is located in a several-meter-thick metasomatic zone between pegmatites and dunites, consisting of essentially monomineralic layers of phlogopite, tremolite and orthoamphibole (Martin-Izard *et al.* 1995). The pegmatites form an intrusive dyke system into the schistose country-rocks and ultramafic bodies (ophiolite complexes). The individual dykes of pegmatite are 3 to 40 cm thick and were interpreted by Martin-Izard *et al.* (1995) as formed from a residual melt derived from the granitic pluton underlying the schistose country-rocks. The granites belong to the synkinematic granites of the area, and were affected by the last phase of three tectonic phases (328–339 Ma) in the area. The study area is situated in the Central Iberian Zone of the Hercynian fold belt, which is characterized by thrusting of ophiolite complexes, eclogite-facies and granulite-facies rocks onto a parautochthonous basement (Arenas *et al.* 1986, Ribeiro *et al.* 1990).

We want to emphasize here that all igneous rocks, the peraluminous granites, the pegmatites as well as dunite and gabbro from the ophiolite, have obviously been deformed and metamorphosed, and thus should be called metagranites, metapegmatites, *etc.* However, the grade of this metamorphic overprint cannot be unambiguously deduced from the description given by Martin-Izard *et al.* (1995), as a detailed correlation

E-mail addresses:

fran0938@mailszrz.zrz.tu-berlin.de
agilg@mineral.min.chemie.tu-muenchen.de
grundman@mineral.min.chemie.tu-muenchen.de
and
morteani@mineral.min.chemie.tu-muenchen.de

among the three tectonic phases, metamorphism and magmatism has not been documented. The presence of an anthophyllite rim around tremolite from the metasomatic zone and at the contact of the dunite indicates temperatures of at least 550°C (Spear 1993) after the emplacement of the pegmatites. A regional high-temperature metamorphism in Galicia, characterized by conditions close to anatexis, with the assemblage sillimanite + K-feldspar, was reported by Martin-Izard *et al.* (1995), as well as contact metamorphism to the albite-epidote hornfels facies, with muscovite and andalusite porphyroblasts. The pressure during these metamorphic events is not well constrained.

In our opinion, the relationship between deformed pegmatites, synkinematic granites and migmatites clearly shows the importance of high-grade metamorphism, anatexis and deformation in the area. The pegmatites from Franqueira are characterized by a high $\text{Na}_2\text{O}/\text{K}_2\text{O}$ ratio, and such chemical compositions resemble those of anatectic melts from metabasites (*e.g.*, Rapp *et al.* 1991, Franz & Smelik 1995). Highly differentiated granitic liquids are mostly rich in K (*e.g.*, Lentz & Fowler 1992).

Our main criticism, however, is concerned with the model of Martin-Izard *et al.* (1995) that heat, fluids and some essential elements, such as Be, Ca, and K, involved in a single-stage contact metasomatic event associated with emerald, chrysoberyl and phenakite mineralization, have been supplied by the crystallizing pegmatites. The thickness of individual pegmatite dykes (only 3 to 40 cm) is relatively small compared to that of the metasomatic zones (several meters). The volume relations of metasomatized rock to pegmatite would require a large amount of thermal energy as well as fluid from the small pegmatite veins. Even at the contact of large pegmatite bodies (*e.g.*, the Tanco pegmatite, in Manitoba), the contact-metasomatic reaction zone does not exceed 10 cm in thickness (Gaupp *et al.* 1984). The formation of the broad zone of tremolite (see Fig. 13 in Martin-Izard *et al.* 1995) needs a considerable amount of Ca, which cannot come from the Ca-poor pegmatite (0.33 wt% CaO, their Table 1). In addition, for the formation of phlogopite, large amounts of K and Al are necessary; although Al is mobile under acidic conditions and K-exometasomatism is typical for pegmatite, it is also unlikely that they were derived directly from such small bodies of pegmatite.

Few data are given by Martin-Izard *et al.* (1995) on the behavior of major and trace elements during metasomatism. These data are, however, essential for an interpretation in terms of contact metasomatism, as suggested by Curtis & Brown (1969, 1971).

Furthermore, Be-minerals were found only in the metasomatized rocks and not in the pegmatites, which contains only 18 ppm Be. This would indicate that, if their genesis is explained by exometasomatism, Be was

efficiently transported away from the original source. This is considered unlikely, and contrasts with the common occurrence of significant amounts of beryl within Be-enriched granitic pegmatites (*e.g.*, Černý & Simpson 1977). In greisen zones associated with Be-minerals, beryl, chrysoberyl and phenakite occur, commonly together with euclase, bertrandite and bromellite (BeO), and often with fluorite and topaz (see review by Barton 1986). None of these minerals were mentioned by Martin-Izard *et al.* (1995), and it is therefore unlikely that the Franqueira deposit can be explained in terms of a greisen environment.

FLUID INCLUSIONS

Martin-Izard *et al.* (1995) described three types of fluid inclusions in phenakite and emerald: early, primary and pseudosecondary $\text{H}_2\text{O} - \text{NaCl} - \text{CH}_4$ (plus other volatiles, probably CO_2) (type 1), $\text{H}_2\text{O} - \text{NaCl} - \text{CO}_2$ (plus other volatiles, possibly CH_4 or N_2) (type 2), and late mixed-salt aqueous inclusions ($\text{H}_2\text{O} + \text{NaCl} + \text{other salts}$) (type 3). They consider the two early types of fluid inclusion (types 1 and 2) as being contemporaneously trapped and representing two immiscible fluids in the system $\text{H}_2\text{O} - \text{CO}_2 - \text{CH}_4 - \text{NaCl}$. This interpretation is not considered valid.

Firstly, most inclusions of type 1 homogenize into the aqueous liquid, and only a few homogenize into the carbonic vapor phase; those of type 2 all homogenize into the aqueous liquid phase. However, two types of fluid inclusion representing coexisting immiscible fluids should homogenize in two contrasting ways, one into the aqueous liquid phase, the other into the carbonic vapor phase (*e.g.*, Ramboz *et al.* 1982). Secondly, fluid immiscibility in the $\text{H}_2\text{O} - \text{CO}_2 - \text{CH}_4 - \text{NaCl}$ system should form a gas-poor aqueous fluid and a gas-rich carbonic fluid (Holloway 1984, Diamond 1990, Duan *et al.* 1992). This is not the case in the Franqueira deposit. Thirdly, the complex CH_4 -bearing inclusions (type 1) indicate relatively reducing conditions, whereas the CO_2 -bearing inclusions (type 2) indicate relatively oxidizing conditions. Both fluids cannot coexist (*e.g.*, Holloway 1984, Kreulen 1987, Cesare 1995). Mixing of these two fluids should result in the precipitation of graphite, which is not observed at Franqueira. Thus, the two early fluids can neither represent two immiscible fluids, nor can they be trapped contemporaneously. It seems more probable that one of the two types of fluid inclusion, most probably the much rarer type-2 inclusions, has been subjected to post-entrapment re-equilibration (*e.g.*, H_2 diffusion out of the inclusion). As the early fluid inclusions in emerald and phenakite have not been trapped in a two-fluid-phase field, their temperatures of homogenization represent minimum temperatures of trapping only.

Martin-Izard *et al.* (1995) claimed that the fluid

inclusion assemblage found in the Franqueira deposit "distinguishes these crystals of emerald from the other emerald ores in the world". Such a fluid inclusion assemblage is not unique at all; in fact, very similar assemblages of fluid inclusions, *i.e.*, early $\text{H}_2\text{O} - \text{CH}_4 - \text{CO}_2 - \text{NaCl}$ inclusions and late gas-poor complex brines, also have been found in emerald from the Gravelotte mine, South Africa (Nwe & Morteani 1993) and from the Habachtal area (Nwe & Grundmann 1990).

DISCUSSION: ALTERNATIVE GENETIC MODEL

Detailed petrological, structural, geochemical and microthermometric studies of two classic schist-type emerald deposits, Habachtal (Austria) and Gravelotte (South Africa), were presented by Grundmann (1983), Grundmann & Morteani (1982, 1989), Nwe & Grundmann (1990) and Nwe & Morteani (1993). These studies have shown that the concept of exometamorphism, as suggested by Fersman (1929), Leitmeier (1937) and Sinkankas (1989), is not adequate to explain the formation of these two deposits. On the basis of many similarities between the above-mentioned deposits and the example of Be-mineralization at Franqueira, as described by Martin-Izard *et al.* (1995), and the problems mentioned above, we propose an alternative genetic model for Franqueira.

The first step in the formation of schist-type emerald, alexandrite and phenakite mineralization is the juxtaposition of Cr- and Mg-rich ultramafic rocks with Be- and alkali-rich felsic rocks, such as granitic pegmatite, rhyolite, pelite or their metamorphosed equivalents. The contact between the two geochemically contrasting rock-units can be either intrusive (*e.g.*, Gravelotte, possibly Franqueira) or tectonic, as a mixture of oceanic and continental crustal material (*e.g.*, Habachtal).

The next step involves the development of zones of metasomatic reaction between the two contrasting rock-units, known as "blackwall zones" (Brady 1977). Mass transport along large chemical potential gradients, mainly of Si, Na, K, Ca and Mg, may form thick, essentially monomineralic zones (Phillips & Hess 1936), if sufficient time, heat and fluid are available. The intrusion and solidification of felsic igneous rocks generally do not supply enough energy to form thick contact-metasomatic reaction zones in ultramafic rocks. However, several meter-thick blackwall zones are found in regional metamorphic terranes, where long-lived fluid flow is channeled in shear zones, which developed between the

mechanically contrasting ultramafic and felsic rock-units. The formation of phyllosilicate minerals, such as mica (phlogopite), talc and chlorite, in the contact zone further lowers its shear strength, leading to increased deformation and strong preferred orientation. Note that strong preferred orientation in the metasomatic phlogopite zone at Franqueira (Fig. 3A in Martin-Izard *et al.* 1995) must have been formed during regional metamorphism, possibly after the intrusion of the synkinematic pegmatites. We want to emphasize here that the blackwall formation is commonly a multistage process, which may start from peak metamorphic conditions and which continues during retrograde metamorphism. At Franqueira, long-lived fluid flow may have continued from amphibolite-facies conditions (presence of anthophyllite + olivine), probably down to greenschist-facies conditions (presence of biotite + chlorite).

During such multistage metasomatism, Be-silicate minerals may form. Beryllium may have been present either as pre-existing Be-minerals or as a trace element in common rock-forming minerals (Grundmann & Morteani 1982). The transformation of felsic country-rocks at the contact with ultramafic rocks into blackwall zones can liberate significant amounts of Be, and allows the formation of new Be-bearing species. For example, a mass-balance calculation for Be shows that the observed concentration of emerald in the blackwall zones at Habachtal is consistent with the amount of Be released from the rock-forming minerals during metasomatism (Grundmann & Morteani 1989). In the presence of Cr liberated from the metasomatically altered ultramafic rocks, alexandrite and emerald may form. The early formation of chrysoberyl (alexandrite) or phenakite (or both), followed by late beryl (emerald), is consistent with experimentally determined phase-equilibria (Franz & Morteani 1981, Barton 1986), and with observations in many other instances of regional-metamorphism-related emerald mineralization (Grundmann & Morteani 1982, 1989, 1995, Franz *et al.* 1986, Grundmann 1989).

In summary, we suggest that the occurrence of phenakite, alexandrite and emerald at Franqueira is another example of multistage Be-mineralization related to regional metamorphism.

ACKNOWLEDGEMENTS

Reviews by T. Ottaway, S.D. Dilles and an anonymous reader are gratefully acknowledged.

Received May 7, 1996, revised manuscript accepted October 15, 1996.