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Granitic pegmatites - fascinating and puzzling rocks

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Introduction

Of the many different kinds of rocks exposed at the earth's surface, granitic pegmatites are probably the most fascinating rocks owned to very large crystal sizes, attractive textures (e.g. graphic granite) and the common complex and exotic mineralogy including a wide range of valuable gemstones. In addition, pegmatites are economically important sources for a number of rare elements including Li, Cs, Rb, Nb, Ta, Be, REE, and Sn (e.g. Linnen et al. 2012), and the industrial minerals K-feldspar, Nafeldspar, quartz and mica (e.g. Glover et al. 2012). For these reasons it appears surprising that granitic pegmatites are probably the least understood rocks in terms of genesis and, thus, the formation of granitic pegmatites is one of the most controversially discussed topics within the fields of petrology and mineralogy (Simmons & Webber 2008, London & Morgan VI 2012, Thomas & Davidson 2016). The reasons for these controversies are the difficulties in explaining the large and variable crystal size, the mineralogical and chemical zoning of pegmatite bodies, the occurrence of massive, monophase quartz cores in the centre of pegmatites and the extreme enrichments of incompatible elements in some pegmatites. Considering the unsolved genetic issues granitic pegmatites can be considered as "puzzling" rocks. In the following three aspects regarding pegmatite classification and genesis are taken up to illustrate and discuss the challenges which pegmatology - the science of pegmatites - is currently facing.

Challenges of pegmatite classification

The classification of rocks, which is commonly based on the two criteria texture and composition, is a very helpful tool for the identification and better understanding of the origin and formation of rocks. Granitic pegmatites have as their major constituents minerals commonly found in "conventional" igneous rocks, such as granites and rhyolites. Latter have been classified successfully in different ways using the bulk chemistry, modal mineralogical composition, and average grain size. In contrast to these rocks, where crystal sizes are small (commonly <2 cm) and minerals are evenly distributed over large volumes, pegmatites represent the very opposite: crystals are usually very large and heterogeneously distributed within the pegmatite bodies comprising a distinct mineralogical and chemical zoning. In pegmatite bodies the crystal size typically increase from the margins towards the centre. The largest crystals found include for example a 14-m-long spodumene crystal from the Etta mine in the Black Hills, South Dakota and an 18-m-long beryl crystal from a pegmatite at Malakialina, Madagascar (Schaller 1916, Rickwood 1981). Massive quartz cores in pegmatites can reach dimensions of more than $100,000 \text{ m}^3$. In most cases it is impossible to determine the bulk chemistry or modal composition of a pegmatite unless the entire body undergoes an analysis. Thus, classical and currently applied pegmatite classifications use so-called indicator minerals, the emplacement depth, the large-scale structure or the crystallisation temperature of pegmatites as

classification criteria. However, establishinging these criteria are not straightforward and can be affected by subjective opinions.

The first generally recognized classification of granitic pegmatites was established by Fersman (1930), who utilized the estimated temperature of crystallization of pegmatites for classification. Later, the depth-zone classification introduced by Ginsburg & Rodionov (1960) has become widely applied even in the most recent classification by Černý & Ercit (2005). However, this latest classification mixes mineralogical, geochemical, and structural criteria with necessary knowledge of P-T conditions during pegmatite formation and therefore it is difficult to apply. Over the last decade, various studies have shown that some types of pegmatites do not fit into the available schemes of classification. For these reasons, discussions for the establishment of a new classification were initiated recently by an international, informal group consisting of mineralogist and petrologists. So far, the group agrees that the new classification should utilize simple measurable and objective criteria (bulk composition, mineralogy, modal content of type minerals, structure) for the first levels of the classification dendrogram. Genetic considerations, based on measurable attributes, should be secondorder criteria. In addition to characteristic assemblages of minerals, the trace element content of major pegmatite-forming minerals, including feldspar, mica, and quartz, has potential for a chemical classification, even though these minerals commonly show a strong chemical zonation within one pegmatite body.

Emplacement depth and crystallization rate of pegmatite melts

The pressures (emplacement depth) at which pegmatites crystallize are poorly constrained by any chemical or textural features of the pegmatites themselves. Most pegmatites are intrusive bodies, and hence postdate their immediately adjacent host rocks. The pressure and temperature at which pegmatites crystallize may have little or no direct relationship to the conditions of formation and the mineral assemblages of their hosts, except intra-plutonic pegmatites, which are commonly the immediate chemical differentiates of their granitic host. Therefore, the depth-zone classifications of pegmatites (Ginsburg & Rodionov 1960, Černý & Ercit 2005) are difficult to apply because the emplacement depth of pegmatites is in most cases uncertain. A rough estimation of the emplacement depth can be made only if the emplacement age of the pegmatite and the time-resolved P-T path of the host rock are known.

In theory the time of crystallisation of pegmatites is a function of the emplacement depth and body size: the deeper the pegmatite melt emplaces the hotter is the host rock and so slower the pegmatite body cools down and the larger the pegmatite body the more time is needed to cool it down. Until recently, pegmatites were widely believed to be the products of extremely slow cooling because of the paradigm that large crystal size requires long cooling times. However, shallow (<1.5 kbar) emplaced pegmatite melts cooled down evidently fast due to huge temperature difference between melt and host rock (temperature quenching). Besides the temperature quenching (the temperature difference between the host rock and the emplacing pegmatite melt) chemical quenching can have a strong effect on the crystallisation time (Simmons 2014). Chemical quenching may occur when fluxing components are removed from the melt by a crystallizing phase which initiates rapid crystallization, since the melt is no longer fluxed by that component. For example, crystallization of tourmaline- or F-bearing phases can result in melt that is suddenly undercooled even though the surrounding rocks and the melt itself remain at about the same temperature. Concluding, several recent studies (e.g. Černý 2005, Webber *et al.* 2005, Simmons & Webber 2008) proposed that cooling rates of pegmatites are

radically more rapid than previously believed and represent an extraordinary contradiction of the paradigm that large crystal size requires long cooling times.

Pegmatites as the major source of gemstones

Pegmatites are the major source of fine and valuable colored gemstones, including varieties of beryl, tourmaline, topaz, spodumene, and garnet. More than 70 less familiar gemstones also occur in pegmatites (Simmons 2014). Looking at the major and minor element chemistry of pegmatite-hosted gemstones, they contain beside the main constituents Si, Al, Ca, Na, and K commonly Be, B, P, Li, F, and Cs. Latter are incompatible elements with large ion radius and low ionic charge (Cs, F, Li; large ion lithophile elements: LILE) or with small ion radius and high ionic charge (Be, B, P).

The first prerequisite for the formation of gemstones is the availability of these incompatible and rare elements in extremely high concentrations in silica melts or liquids from which the gemstones crystallize. These elements can have enrichment factors greater than 100,000 in some pegmatites compared to the average composition of the continental crust (e.g. Linnen *et al.* 2012). Most scientists agree that these extreme enrichments of incompatible elements cannot be achieved via simply progressive fractionation of a large-volume granitic melt (e.g.Simmons & Webber 2008). Thus, two alternative models have been suggested recently: (1) pegmatite melts are initially supercritical fluids that involve into co-existing silicate-rich and volatile-rich melts and aqueous fluids (Thomas & Davidson 2016) and (2) the constitutional zone refining as driving process of pegmatite crystallisation (London 2009). However, both models have their pros and cons and the processes leading to these extreme element enrichments remain controversially (see also Müller & Thomas 2016 in this volume).

The second prerequisite for the formation of gemstones is the development of reasonable large crystal sizes. Pegmatites are renowned for their spectacular development of giant crystals. However, processes initiating and supporting the growth of extremely large crystals in silica liquids are currently being debated (see previous chapter).

The third prerequisite is a stress-free, relative stable crystallization environment in order to grow clear, fracture- and inclusion-free crystals, the major criteria of gemstone quality. This environment is given in open cavities also called miarolitic cavities or pockets which develop as a result of oversaturation of the melt in volatiles (predominantly H_2O) during the final stage of pegmatite crystallization. The oversaturation leads to the separation of volatiles from the melt which accumulate into bubbles or blobs causing finally the formation of volatile-filled cavities. The cavities provide the space in which terminated crystals can grow stress-free from the cavity walls inward. Two factors are critical in the process of pocket formation (Simmons *et al.* 2012). First, the primary melt must have initially contained sufficient volatiles so that late-stage exsolution occur. Second, the pressure regime must be not too high (<3 kbar), as volatile exsolution is inversely proportional to pressure. Consequently, pockets are most abundant in shallow-level pegmatites and virtually absent in pegmatites formed under high-pressure conditions (>3 kbar). Gem-bearing pockets represent the ultimate concentrations of exotic elements, volatiles, and other fluxes present in pegmatites. However, there are a number of unsolved questions related to the different processes involved in pocket formation.

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