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## Crystallographic evidence for simultaneous growth in graphic granite

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### ABSTRACT

Deciphering the graphic texture is important for understanding its origin and the crystallization process of granitic rocks. We present here investigations on petrography, crystallographic relationship, and three-dimensional (3D) relations between quartz and alkali-feldspar in graphic granite from the Fangshan Pluton, north China. Under the petrographic microscope, multiple domains of quartz grains exhibit a nearly simultaneous extinction within a single crystal of feldspar. Electron back scattered diffraction (EBSD) analyses indicate that majority of quartz (Qz) grains are in a topotaxial relationship with the host K-feldspar (Kfs) with  $[11\bar{2}3]_{\text{Qz}}//[001]_{\text{Kfs}}$ ,  $(10\bar{1}0)_{\text{Qz}}//[(110)_{\text{Kfs}}$ , and either  $(10\bar{1}1)_{\text{Qz}}$  or  $(01\bar{1}1)_{\text{Qz}}$  parallel to  $(110)_{\text{Kfs}}$ . Moreover, essentially all quartz grains in the sample are crystallographically related by either the Dauphiné or the Japan twin laws. Synchrotron X-ray computed micro-tomography demonstrated that majority of the quartz grains are in the form of sub-parallel long rods with D-, L-, and U-shaped cross sections. These results provide new quantitative microstructural evidence of simultaneous growth of quartz and feldspar in graphic granite.

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### 1. Introduction

Graphic granite, found predominantly in granitic pegmatite, is a leucocratic granitic rock consisting of an intimate intergrowth of alkali feldspar and quartz with a distinctive texture as ancient cuneiform writing when viewed in certain cross sections. In spite of easy identification and clear illustration in every fundamental geology textbook for such a masterpiece of nature, its origin has been debated in the literature for more than a century (e.g. Wahlstrom, 1939; Simpson, 1962; Černý, 1971; Smith, 1974; Fenn, 1986; Lentz and Fowler, 1992; Stel, 1992; London, 2009; London and Morgan, 2012). Two principal models have been proposed for the formation of the graphic texture, simultaneous growth of quartz and feldspar (e.g. Simpson, 1962; Fenn, 1986; Lentz and Fowler, 1992) and replacement of feldspar by quartz (e.g. Augustithis, 1962; Seclaman and Constantinescu, 1972).

The proposed evidence for the simultaneous growth mechanism includes the following. (1) The relative constancy of graphic quartz and feldspar in a volume ratio of about 1:3 (Simpson, 1962), which has been interpreted as due to rapid crystallization from a melt with a homogeneous composition (Fenn, 1986). (2) Crystallographic control of quartz by the feldspar host. This is indicated by the observation of adjacent quartz grains showing simultaneous extinctions over a broad

area in thin sections (Wahlstrom, 1939). It has been suggested that quartz in graphic intergrowths is oriented with respect to the feldspar host based on the analyses using X-ray diffraction and petrographic microscope (e.g. Fersman, 1928). (3) The reticular connection of graphic quartz observed in thin sections (Seclaman and Constantinescu, 1972). X-ray computerized tomography and imaging (Simpson, 1962; Ikeda et al., 2000) later clearly revealed the skeletal nature of graphic quartz. (4) Remarkably similar graphic textures can be produced by experiments on pegmatite composition (Fenn, 1986) and simulations of crystal growth in hydrous albite-quartz and albite-orthoclase systems (Baker and Freda, 1999).

The replacement origin was argued for the graphic texture in other studies based on different, even contradictory evidence or observations. Schaller (1927) suggested for the first time that quartz rods in graphic granite were formed by replacement of feldspar along weak zones, such as cleavage planes and twinning planes. The supporting evidence for the replacement mechanism includes: (1) Contact relation between quartz and feldspar. It has been shown that quartz tends to preferentially distribute along interfaces between feldspar or other mineral crystals, as well as along cleavages or other crystallographic planes of the feldspar host, and occasionally the same skeletal quartz is included in two or more feldspar hosts (Seclaman and Constantinescu, 1972). (2) In contrast to the observations of fixed quartz/feldspar ratio (Simpson, 1962; Černý, 1971), the observations of large variation of quartz/feldspar ratio in different types of graphic texture are not in accordance with a eutectic crystallization (Augustithis, 1962). (3) Different generations of quartz and feldspar. In some graphic granites, quartz and feldspar

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display different characteristics including the deformation features. It has been suspected that the graphic quartz has a younger age than that of the feldspar (Augustithis, 1962; Seclaman and Constantinescu, 1972). (4) No constant orientation of quartz with respect to feldspar. Although simultaneous extinction is common, in some graphic quartzs no simultaneous extinction has also been observed (Wahlstrom, 1939; Simpson, 1962; Ikeda et al., 2000). Fersman (1928) found a  $42^{\circ}16'$  angle between the *c* axes of quartz and feldspar based on measurements using a U stage, and showed that the prism edge of the feldspar host is parallel to an edge between two adjacent rhombohedral faces of the quartz grains. However, using both X-ray and petrographic microscope techniques, several others (Heritsch, 1953; Heritsch et al., 1962; Choudhury et al., 1965; Bakumenko, 1966) confirmed a number of angular relationships. Moreover, some investigators have concluded that the quartz rods in graphic granite are not oriented according to a fixed crystallographic law nor are their positions rigidly defined by vectorial properties of the host feldspar (Wahlstrom, 1939; Simpson, 1962).

Systematic, quantitative knowledge on the crystallographic relationship between quartz and feldspar in the graphic granite should contribute to solve the above disagreement in the origin of graphic texture. Understanding the graphic texture is important for deciphering the origin and the crystallization process of granite pegmatites in a volatile-rich magma (e.g. Smith, 1974; London and Morgan, 2012). The conventional U-stage petrographic microscopy and X-ray diffraction have proven to be unsatisfactory tools. Contradictory conclusions on the origin of graphic texture were drawn based on incomplete information or limited observations, such as the contact relation between quartz and adjacent quartz or feldspar (e.g. Wahlstrom, 1939; Seclaman and Constantinescu, 1972). New analytical techniques, including the electron backscattered diffraction (EBSD) and the synchrotron-based X-ray tomographic microscopy, have become available for performing such dedicated and complex petrographic analyses. The EBSD technique is capable of determining the complete crystallographic orientation of individual rock-forming minerals with a spatial resolution better than  $1\ \mu\text{m}$  and with an absolute angular resolution better than  $1^{\circ}$  over a relatively large area (regular thin section size) (e.g. Humphreys, 1999; Prior et al., 1999; Humphreys, 2004). It is a powerful crystallographic tool that is applicable to crystalline solids of any crystal symmetry including optically isotropic and opaque minerals. Recent years have seen many applications of this tool for the study of deformation mechanism (e.g. Zhang et al., 2006; Menegon et al., 2011; Ohuchi et al., 2011), the identification of unknown minerals (e.g. Franke et al., 2007; Ma et al., 2012), and the quantification of metamorphic and magmatic microstructures (e.g. Hammer et al., 2010; Obata and Ozawa, 2011) and exsolution microstructure (e.g. Zhang et al., 2011). In addition to the two-dimensional (2D) crystallographic information, the synchrotron-based X-ray tomographic microscopy technique can provide high resolution three-dimensional (3D) tomographic information on contact relations among multiple mineral phases or melts (Rivers et al., 1999; Wang et al., 2005). This study reports 2D crystallographic relationship and 3D contact relations of quartz and feldspar in graphic granite collected from the Fangshan Pluton, China, providing quantitative supporting evidence for the mechanism of simultaneous crystallization.

## 2. Samples and analytical methods

The graphic granite samples were collected from the Fangshan Pluton ( $N39^{\circ}42'26''$ ,  $E115^{\circ}56'38''$ ) in the Western Hills of Beijing, north China. The Fangshan Pluton is a well-known large late Mesozoic (129–134 Ma) intrusion in the North China Craton and is composed of fine-grained quartz diorite, porphyritic granodiorite and mafic dyke (Davis et al., 2001; Cai et al., 2005). More recent studies suggest that this low-Mg adakitic pluton was originated from mixing of mantle-derived magma and partial melts from a molten thickened lower continental crust (Sun et al., 2010; Xu et al., 2012). Granitic pegmatites usually occur as sharply discordant dikes, with widths ranging from

several centimeters to dozens of centimeters, intruding in the fine grained quartz diorite and granodiorite (Fig. 1a). The pegmatite veins are composed of quartz, alkali-feldspar, plagioclase and minor biotite. The crystals of quartz and alkali-feldspar are dominantly subhedral, with grain size varying from medium to coarse grained (i.e.,  $0.5\ \text{cm}$  to  $>10\ \text{cm}$ ). Graphic quartz-feldspar intergrowths are mainly found in coarse grained alkali-feldspar crystals near one side of the veins (Fig. 1b). The studied samples were collected from fresh coarse-grained alkali feldspars from various places in the pegmatitic veins that were recently exposed in a quarry, with minimal later day alteration.

Because the shape of quartz in graphic granite is a function of orientation of the feldspar (e.g. Fersman, 1928; Simpson, 1962; Ikeda et al., 2000), most thin sections were prepared from rock slices with surface orientation subparallel to the (001) or (010) of the matrix feldspar. Doubly polished thin sections were prepared using a series of diamond powers of decreasing grain size from  $9.5\ \mu\text{m}$  to  $3\ \mu\text{m}$  for surface grinding and a  $0.3\ \mu\text{m}$  alpha alumina solution for polishing. Remaining surface damage was then removed by chemical-mechanical polishing using a  $0.05\ \mu\text{m}$  colloidal silica solution for at least 4 h. The major elements of the alkali-feldspar, plagioclase feldspar, quartz and muscovite were analyzed with a JEOL-JXA-8100 electron probe microanalyzer (EPMA) at the State Key Laboratory of Geological Processes and Mineral Resources (GPMR), China University of Geosciences (CUG) at Wuhan. The accelerating voltage, beam current, focused beam size, and counting time were set to 15 kV, 10 nA,  $5\ \mu\text{m}$  and 20 s, respectively.

Crystallographic orientation data were obtained using an Oxford Instruments HKL Nordlys II EBSD detector coupled to a FEI Quanta 450 field emission gun scanning electron microscope (FEG-SEM) at the GPMR of CUG at Wuhan. Thin sections for EBSD analyses were uncoated for better pattern quality in a low-vacuum mode. Working conditions were as the following: 20 kV accelerating voltage, 6 spot size, working distance of about 25 mm,  $70^{\circ}$  sample tilt, and low-vacuum mode (20–30 Pa). Patterns were acquired on rectangular grids by shifting the electron beam with a step size of 40 or  $50\ \mu\text{m}$ . Six to seven Kikuchi bands were used for automatic indexing which compares the obtained intensities of Kikuchi bands with those of computer simulations for trigonal alpha-quartz and monoclinic alkali feldspar sanidine (Keefer and Brown, 1978). To assure data quality, only those measurements with mean angular deviation values below 1.0 (between detected and simulated EBSD patterns) were accepted for analyses. We also confirmed the accuracy of automated EBSD analyses for each sample by comparing results to randomly selected orientation data indexed with the manual interactive mode. High-resolution orientation contrast (OC) images were also taken to reveal sharp changes in crystallographic orientations such as grain boundaries, sub-grain boundaries or structures such as boundaries across mineral phases (Prior et al., 1999). The CHANNEL 5+ software was used for removing erroneous data misoriented from all eight neighboring measurement by more than  $5^{\circ}$  and for replacing non-indexed measuring points by the most common neighboring orientation during an orientation map reconstruction from the raw orientation map to avoid introducing artifacts (Prior et al., 2002).

From hand Sample FS-1, a small cube, approximately  $5.7\ \text{mm}$  (w)  $\times$   $5.8\ \text{mm}$  (w)  $\times$   $5.4\ \text{mm}$  (h), was extracted and examined at ambient conditions by X-ray computed micro-tomography (CMT) on a bending magnet beamline 13-BM-D of GSECARS, at the Advanced Photon Source, Argonne National Laboratory (Rivers et al., 2004). The sample was mounted at the center of the rotation stage of the high pressure X-ray tomographic microscopy (HPXTM) module (Wang et al., 2005) during data collection. A monochromatic X-ray beam (photon energy optimized at 29 kV) was used to collect radiographs of the sample. A cerium doped lutetium aluminate garnet (LuAG:Ce) single crystal scintillator was used to convert X-ray contrast into visible light. The visible images were reflected by an optical mirror, through a  $5\times$  microscopic objective, and recorded by a CoolSNAP HQ2 CCD camera. While rotating the sample along an axis perpendicular to the X-ray beam, 2D radiographic

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