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Research paper

## Alkali control of high-grade metamorphism and granitization

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## ABSTRACT

We review petrologic observations of reaction textures from high-grade rocks that suggest the passage of fluids with variable alkali activities. Development of these reaction textures is accompanied by regular compositional variations in plagioclase, pyroxenes, biotite, amphibole and garnet. The textures are interpreted in terms of exchange and net-transfer reactions controlled by the K and Na activities in the fluids. On the regional scale, these reactions operate in granitized, charnockitized, syenitized etc. shear zones within high-grade complexes. Thermodynamic calculations in simple chemical systems show that changes in mineral assemblages, including the transition from the hydrous to the anhydrous ones, may occur at constant pressure and temperature due only to variations in the H<sub>2</sub>O and the alkali activities. A simple procedure for estimating the activity of the two major alkali oxides, K<sub>2</sub>O and Na<sub>2</sub>O, is implemented in the TWQ software. Examples of calculations are presented for well-documented dehydration zones from South Africa, southern India, and Sri Lanka. The calculations have revealed two end-member regimes of alkalis during specific metamorphic processes: rock buffered, which is characteristic for the precursor rocks containing two feldspars, and fluid-buffered for the precursor rocks without K-feldspar. The observed reaction textures and the results of thermodynamic modeling are compared with the results of available experimental studies on the interaction of the alkali chloride and carbonate-bearing fluids with metamorphic rocks at mid-crustal conditions. The experiments show the complex effect of alkali activities in the fluid phase on the mineral assemblages. Both thermodynamic calculations and experiments closely reproduce paragenetic relations theoretically predicted by D.S. Korzhinskii in the 1940s.

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## 1. Introduction

Traditionally, pressure (P) and temperature (T), and partial pressure (fugacity) of some volatile components (H<sub>2</sub>O, CO<sub>2</sub>, O<sub>2</sub>) have been considered as the major intensive variables controlling metamorphic mineral assemblages in rocks of fixed bulk

composition (e.g. Spear, 1993). On the basis of paragenetic analysis of rocks from the Precambrian high-grade metamorphic rocks and granitoids of the eastern Siberian region, Korzhinskii (1946, 1962) developed a concept of “mobility of alkalis during metamorphism and granite formation”. According to this concept, chemical potentials of K and Na may be just as important factor and can be considered as intensive variables, as well. Applying this concept Korzhinskii (1946) proposed a number of mineralogical criteria to evaluate the influence of alkali activities in melts and fluids on mineral assemblages from granitic and syenitic rocks. Later, Korzhinskii (1962) concluded that the formation of the orthopyroxene-K-feldspar assemblages after metapelitic gneisses (termed as “charnockitization”) should also be considered as an example of a process controlled by alkali activities in fluids.

Similar to T, P, H<sub>2</sub>O activity, and other conventional intensive variables in metamorphic systems, the variations of alkali activities in fluids should be recorded in the regular changes in mineral

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assemblages, zoning of minerals and reaction textures involving K and/or Na-bearing phases, and, correspondingly, expressed in mineral reactions controlled by those activities. [Perchuk and Gerya \(1993\)](#) applied Korzhinskii's alkali mobility concept to explain textural features and regular compositional trends in coexisting biotite, orthopyroxene and plagioclase related to the development of the K-feldspar micro-veins in charnockitic gneisses and charnockites from central Finland and SW Baikal. Since then, similar textural and compositional relations have been repeatedly reported in granulites from many high-grade terrains ([Hansen et al., 1995](#); [Franz and Harlov, 1998](#); [Harlov et al., 1998](#); [Newton et al., 1998](#); [Harlov and Wirth, 2000](#); [Perchuk et al., 2000](#); [Harlov and Förster, 2002](#); [Montanini and Harlov, 2006](#); [Touret and Huizenga, 2011](#); [Rajesh et al., 2013](#)). These findings have become a basis for a new approach, which invokes complex, poly-ionic aqueous solutions and brines as a factor of high-grade metamorphism ([Newton, 1995](#); [Aranovich and Newton, 1996](#); [Newton et al., 1998](#); [Yardley and Graham, 2002](#); [Newton and Manning, 2010](#); [Touret and Nijland, 2013](#)).

The present paper reviews typical examples of reaction textures, which reflect the passage of fluids with variable alkali activities. It then considers the possible fluid-mineral reactions responsible for the development of these textures and for the accompanying compositional variations in the compositions of the coexisting minerals. These reactions are then thermodynamically modeled in simple chemical systems under variable alkali and H<sub>2</sub>O activities and quantitative estimates of alkali activities presented for a few well documented examples of fluid-rock interaction in intermediate metamorphic rocks and metapelites.

**Mineral and end-member abbreviations:** Ab – albite, Alm – almandine, Amph – amphibole, An – anorthite, Ann – annite, Ap – apatite, Bar – barroisite, Bt – biotite, Cpx – clinopyroxene, Di – diopside, Eas – eastonite, Ed – edenite, En – enstatite, Fs – ferrosillite, Grs – grossular, Grt – garnet, Hbl – hornblende, Hed – hedenbergite, Ilm – ilmenite, Kfs – K-feldspar, Kls – kalsilite, Mt – magnetite, Ol – olivine, Opx – orthopyroxene, Or – orthoclase, Phl – phlogopite, Pl – plagioclase, Prg – pargasite, Prp – pyrope, Qtz – quartz, Sid – siderophillite, Sph – sphene, Tr – tremolite, Ts – tschermackite, Win – winchite

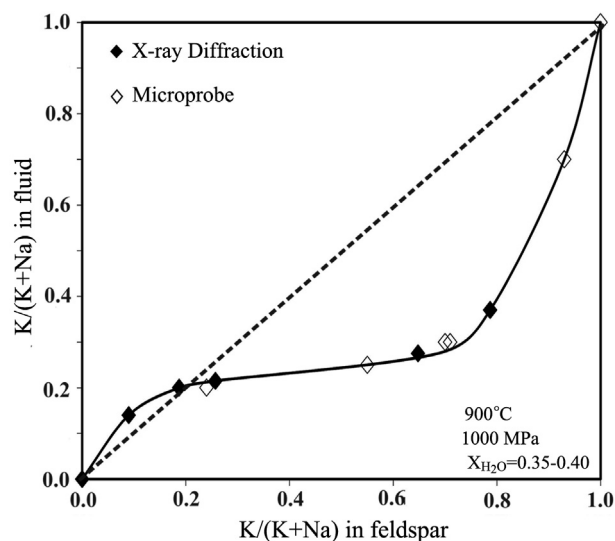
## 2. Mineralogical criteria and reactions indicative of variable alkali activities

### 2.1. Reactions involving feldspars only

[Fig. 1](#) shows the pronounced dependence of the alkali feldspar K content on the K/Na ratio in a coexisting alkali chloride brine at 900 °C and 1000 MPa according to experimental data by [Aranovich et al. \(2013\)](#). In this example, the K/(K + Na) ratio of the coexisting phases is defined by a very well known exchange reaction ([Orville, 1963](#); [Iiyama, 1965](#)):



Even though conditions at 900 °C are considerably super-solvus in the alkali feldspar system ([Yund and Tullis, 1983](#)), the behavior is very similar to the alkali feldspar–brine equilibria at lower temperatures. Thus, very orthoclase-rich alkali feldspars can coexist with a relatively Na-rich aqueous chloride fluid. Slight shifts in the fluid K/(K + Na) ratio (from 0.2 to 0.3) result in large changes in feldspar composition (from Kfs<sub>20</sub> to Kfs<sub>80</sub>), as emphasized by [Orville \(1963\)](#). This mode of K-enrichment in rocks is much more effective than could result from a fluid-undersaturated partial melting of mica- and/or amphibole-bearing rocks ([Aranovich et al., 2013](#)).

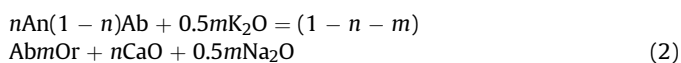


**Figure 1.** K-Na exchange between alkali feldspar and a KCl-NaCl brine according to the experimental data by [Aranovich et al. \(2013\)](#).

Reaction (1) can be re-written without specifying particular alkali species in the fluid as:



where K<sub>2</sub>O and Na<sub>2</sub>O refer to the (virtual) solid oxides in the rock under consideration. This reaction indicates that the K/Na ratio in alkali feldspars may provide an estimate of the  $a_{\text{K}_2\text{O}}/a_{\text{Na}_2\text{O}}$  ratio in the rock and, correspondingly, in the coexisting fluid phase. [Korzhinskii \(1946\)](#) suggested that variations in the K<sub>2</sub>O and Na<sub>2</sub>O chemical potentials (activities) should also affect the composition of plagioclase coexisting with alkali feldspar. He proposed a criterion: *the higher the anorthite content of plagioclase coexisting with K-feldspar, the higher the K activity corresponding to this assemblage due to the reaction*



In two-feldspar assemblages, this reaction simultaneously operates with reaction (1a), indicating that an increase in the K<sub>2</sub>O activity should produce an assemblage of more anorthite-rich plagioclase with orthoclase-rich alkali feldspar. [Fig. 2](#) illustrates these relations in terms of the  $\log(\text{K}^+/\text{H}^+)$  and  $\log(\text{Na}^+/\text{H}^+)$  ratios after [Perchuk et al. \(2000\)](#).

Reaction textures of plagioclase replacement by K-feldspar are common for granitic rocks, and they are usually interpreted as products of sub-solidus interaction of the plagioclase with post-magmatic fluids ([Eskola, 1956](#); [Orville, 1962](#); [Putnis et al., 2007](#)). [Perchuk and co-authors \(Perchuk and Gerya, 1992, 1993; Perchuk et al., 1994\)](#) demonstrated textural evidence of the Korzhinskii's reaction (2) specifically for metamorphic rocks, i.e. the charnockitized gneisses of the Sulkava complex (Central Finland) and the Sharyzhgaysk complex (SW Baikal). Here alkali feldspar rims and micro-veins occur along the plagioclase grain boundaries in contact with both quartz and Fe-Mg minerals. They found that the anorthite content of the plagioclase in contact with the micro-veins increased in accordance with reaction (2), while the orthoclase content of the alkali feldspar increases due to simultaneous reaction (1a). Subsequently, similar textural and compositional relations between plagioclase and K-feldspar have been described in

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