



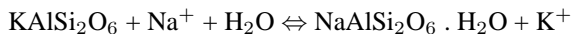
## **<sup>18</sup>O as a Tracer to determine the Mechanism of Replacement of Leucite by Analcime.**

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Leucite,  $\text{KAlSi}_2\text{O}_6$  is a characteristic mineral of K-rich Si poor lavas where it is commonly the dominant phenocryst constituent. It is also an essential component of K-rich ultra-basic rocks. Leucite occurs in rocks of Tertiary or younger age and its absence in older rocks results from its alteration to analcime,  $\text{NaAlSi}_2\text{O}_6 \cdot \text{H}_2\text{O}$ . The reaction:



has been demonstrated experimentally to have a very small activation energy. Experiments using pure crystalline natural leucite in NaCl solutions with sea water concentration at temperatures 150 -200°C show replacement reactions starting almost immediately. SEM observations of textural changes as well as compositional analyses using EDX, XRD, Raman, infrared spectroscopy, and time-of-flight secondary ion mass spectrometry, indicate a replacement mechanism involving recrystallisation.

Both leucite and analcime have an open framework structure. It has previously been suggested that sub-solidus alkali ion exchange accounts for the partial or complete conversion of leucite to analcime, essentially by a simple K-Na substitution. However, previous studies could not explain the large porosity in analcime replacing leucite with no apparent change in external dimensions, i.e., a pseudomorphic replacement, especially in the light of a large solid molar volume increase of about 10% during the replacement process [1]. Recent work on the pseudomorphic replacement of a crystal of KBr by a single crystal of KCl has demonstrated that porosity plays a key role during interface-controlled coupled dissolution-reprecipitation [2,3]. Textural and com-

positional changes in natural systems may be an indication that such a replacement process has taken place. Similar textural characteristics have also been shown in the hydrothermal alteration of Ta-based pyrochlore [4], previously thought to take place by a diffusion-controlled hydration and ion-exchange process.

Using  $^{18}\text{O}$ -enriched  $\text{H}_2\text{O}$ , we show that the replacement of leucite by analcime is not a simple cation substitution but proceeds by a dissolution-reprecipitation process where some structural oxygen atoms of the leucite framework are exchanged and a new analcime structure forms at a moving interface through the leucite parent crystal. The large porosity in the analcime product phase must result from some of the parent phase being lost to the solution to give a volume deficit reaction. If an open framework aluminosilicate structure can be replaced by a new phase by a dissolution-reprecipitation mechanism, there are resulting implications for the understanding of the mechanism of ion exchange processes in general.

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