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LEUCITE SURVIVAL: THE ALTERATION TO ANALCIME[1]

A. K. GUPTA* AND W. S. FYFE
The University of Western Ontario, London, Canada

ABSTRACT

The rate of conversion of leucite to analcime in salt solutions has been studied between 300 and 150°C. The process is fast and the activation energy is in the order of 8 Kcal. Given sufficient time, leucite will not survive diagenetic processes in most natural waters. Thus leucite-bearing rocks may rapidly be converted to analcime analogues before any major metamorphism occurs.

RÉSUMÉ

Le taux de conversion de la leucite à l'analcime dans des solutions salées a été étudié à des températures entre 300 et 150°C. Le procédé est rapide et l'énergie d'activation est de l'ordre de 8 kcal. En lui attribuant suffisamment de temps, la leucite ne survivra pas aux procédés diagénétiques dans les eaux les plus naturelles. Ainsi les roches contenant de la leucite peuvent se convertirent rapidement en analogues d'analcime avant qu'aucun métamorphisme majeur se produise.

(Traduit par le journal)

INTRODUCTION

Recent petrological research has shown the immense significance of some of the less-voluminous melting products of the earth. Thus rocks such as kimberlites, komatiites and some high-potash mantle-fusion products provide information on the heterogeneous nature of the mantle and on mixing phenomena and their variation with time. One of us (W.S.F.) while working with Dr. T. H. Pearce became intrigued by the analcime-bearing volcanics of the Crowsnest Formation in Alberta (Pearce 1970). While Pearce has argued the case for primary, igneous analcime, we doubt if the possibility of leucite replacement at very low temperatures can be ignored. In the classic areas of Italy, the replacement reaction has been frequently observed. The present study was concerned with a study of the rate of conversion and determination of the temperature coefficient of the reaction rate.

EXPERIMENTAL

A natural leucite from Roccomonfina, Italy was used in this study (SiO_2 54.60; Al_2O_3 22.80; K_2O 18.46; Na_2O 1.18; CaO 0.16; MgO 0.03; Fe_2O_3 2.40). The leucite was ground to 100 mesh and allowed to react with salt solutions. In one set of experiments, 20 mg of leucite plus 20 mg of sodium chloride and 20% water were placed in sealed gold capsules and the reaction studied at 1 kb total pressure. In this case the salt solution will be saturated.

In a second set of experiments, 0.1 gm of leucite was mixed with 2 cc of synthetic sea water (a salt mix of the major species prepared for biological studies). These solutions contain sodium and potassium in normal sea-water concentrations. The results of the experiments are shown in Table 1 and Figure 1. The amount of conversion was estimated from *x*-ray diffractometer traces calibrated with known mixtures of analcime and leucite.

DISCUSSION

From the experimental results it is clear that the reaction is very fast and can be measured easily down to 150°C. As the reaction can be

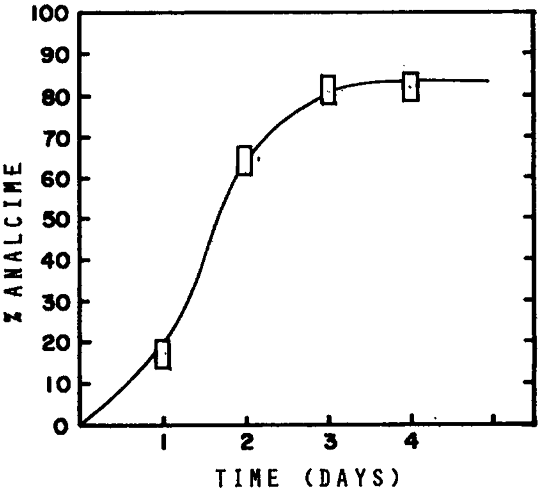

FIG. 1. Rate of the reaction leucite plus sea water to analcime at 150°C.

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*Present address: Institut für Mineralogie, Ruhr-Universität Bochum.

TABLE 1. REACTION RATES

Series A ($Lc_{50}NaCl_{50}$ + 20% H_2O)			Series B (0.1 gm Lc + 2 cm^3 sea water)		
Temp. (°C)	Time (days)	Conversion (%)	Temp. (°C)	Time (days)	Conversion (%)
325	4	100	225	1	81
275	13	100	200	1½	90
225	13	100	175	3	95
225	1	38	150	1	17
200	4	34	150	2	64
150	21(hours)	0	150	3	81
150	13	14	150	4	82

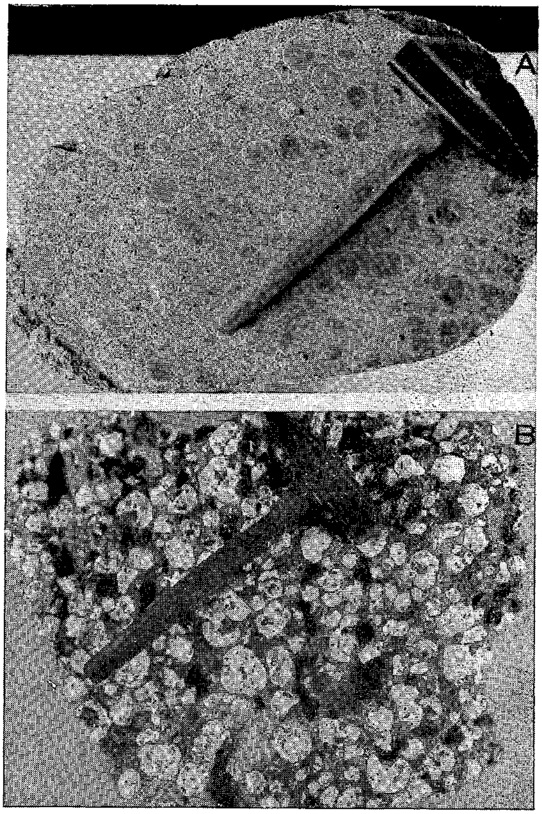

FIG. 2. (A) Analcime from the Crowsnest Formation; (B) Analcime after leucite from Vico volcano, Italy. (The hammer is 8 cm long).

studied over a 100°C range, it follows that the activation energy is small (Fyfe 1973). If we assume that for the early part of the reaction the rate equation is zero order, that is

$$\frac{dc}{dt} \cong K$$

(note the process is almost linear up to 80% conversion), then the activation energy is in the order of 8 kcal mol^{-1}. This is a very low activation energy and the reaction will be rather temperature-insensitive. If we consider that the time constants of diagenetic (burial) processes may be in the order of 10^5 - 10^7 years, then even at 25°C, given appropriate fluids, the reaction would go to completion.

The process being studied is:

$KAlSi_2O_6$ (solid) + Na^+aq + $H_2O \rightarrow NaAlSi_2O_6 \cdot H_2O$(solid) + K^+aq, $\Delta G°_{298} = -1093$ cal mol^{-1} (Robie & Waldbaum 1968). While there are considerable uncertainties in $\Delta G°$, this figure suggests that analcime is stable even when the concentration of potassium exceeds the concentration of sodium ($K^+aq/Na^+aq = 6.4$ at equilibrium). This implies that even normal river water ($K^+/Na^+ = 0.3$) could cause the reaction.

While no nucleation is involved in the reaction (but a 10% volume expansion occurs) a number of steps could be rate-determining. These might include:

(a) diffusion rates of Na^+ or K^+ in the crystal

(b) dehydration rates of ions before entry

(c) diffusion rate of water into the crystal

The overall ΔH (*cf.* Δ*E*) of processes like:

K^+ solid + $Na^+aq \rightleftharpoons Na^+_{solid} + K^+_{aq}$ are small (Latimer 1964). Normal heats of hydration of solids are similar to the activation energy determined in this work. The fact that the more dilute sea water appears to react faster than the saturated salt solutions could indicate that water activity and diffusion are important.

Numerous authors have noted the facile leucite-analcime reaction (see Deer, Howie & Zussman 1963; Bragg *et al.* 1965) (see Fig. 2). The main conclusion of this work is, that if sufficient sodium-bearing water is available, then conversion will occur even at surface temperatures. The chances for leucite survival are small and in fact most reported occurrences of volcanic leucite are in rather recent rocks. It is clear that this process may occur at low temperature when other primary igneous minerals might show little alteration. This possible reaction must be considered before analcime is considered as a primary igneous phase (pace Pearce, 1970).

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