PHILOSOPHICAL TRANSACTIONS OF THE ROYAL SOCIETY A
MATHEMATICAL, PHYSICAL & ENGINEERING SCIENCES

The Movement of Uranium during Higher Grade Metamorphic Processes

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The movement of uranium during higher grade metamorphic processes

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The movement of uranium in rocks is largely governed by movement of water and by partial melting.

The movement of uranium at the surface and during diagenetic processes is not considered here. The physical conditions under which higher grade metamorphic processes in the sense of this paper take place are such that rock strength does not contribute significantly to the maintenance of an open pore system. A separate fluid phase can then only exist if its pressure is at least equal to the load pressure. Such conditions exist at a minimum depth corresponding to rock pressures of about 2 kbar (200 MPa) (*ca.* 6 km) and temperatures above 200 °C. These are the conditions of rocks crystallized under greenschist, amphibolite and granulite facies conditions of metamorphism, and it is the behaviour of uranium under such conditions which is discussed here.

Gamma ray spectrometry analyses during the last 15 years have contributed significantly to the quantity of data available on uranium concentration in metamorphic rocks.

Nuclear emulsion and fission track studies combined with leaching experiments have given significant information on how uranium is distributed in rocks and in which it occurs.

Isotope studies of the U–Pb systems and also studies of homogenization of Sr isotopes in metamorphic processes can give valuable information about the movement of uranium under these conditions.

INTRODUCTION

It is necessary first to comment on the physical conditions defining the crustal level(s) in which higher grade metamorphic processes take place.

The movement of uranium in rocks is largely governed by two parameters: (1) movement of water, and (2) partial melting. The movements of uranium characteristic of surficial and diagenetic processes are not considered here. These are the processes during which most of the redistribution of uranium in the crust, and secondary concentration of uranium into ore deposits, take place.

Water is present in all rocks, and movement of water is characteristic of all progressive regional metamorphic processes (dehydration reactions). However, although water may be present as a separate phase in many metamorphic rocks at depths where metamorphic processes take place, the physical state of the system under such conditions suggests that the water phase may be nothing more than a surface film on mineral grains, (adsorbed film), or a filling of microscopic pores. It is important to emphasize that the thermodynamic state of water in such systems may be different from that of water in bulk.

A clear distinction must be drawn between environments where rocks have sufficient strength, or a sufficiently high yield point, to maintain open pores or open fractures, and the conditions under which a separate water phase is only present as an adsorbed film (in many cases apparently just as a monolayer). In the first case the fluid filling the pores and fractures may be at a pressure P_f less than P_l. In a water-filled vertical fracture, or in a continuous open pore system extending down from the surface, the fluid pressure (P_f) would be roughly $\frac{1}{3}P_l$. In discontinuous pore systems, that is porous rocks of zero permeability, the pores may be held open by the fluid pressure, or by the strength of the rocks. The fluid pressure in this case could range from a possible zero (empty or partly filled pores) to a value equal to, or greater than, the load pressure, depending on rock strength. Normally, however, the strength will be low at great depths and will not contribute significantly to the maintenance of an open pore system. Under such conditions a separate fluid phase can exist only if its pressure is at least equal to the load pressure (Fyfe *et al.* 1958). Such conditions exist where rocks crystallize under greenschist, amphibolite and granulite facies conditions of metamorphism (figure 1; Turner 1968). This implies a minimum pressure of approximately 2 kbar† (*ca.* 6 km), and temperatures above 200 °C. It is the behaviour of uranium under these conditions which is covered in this paper.

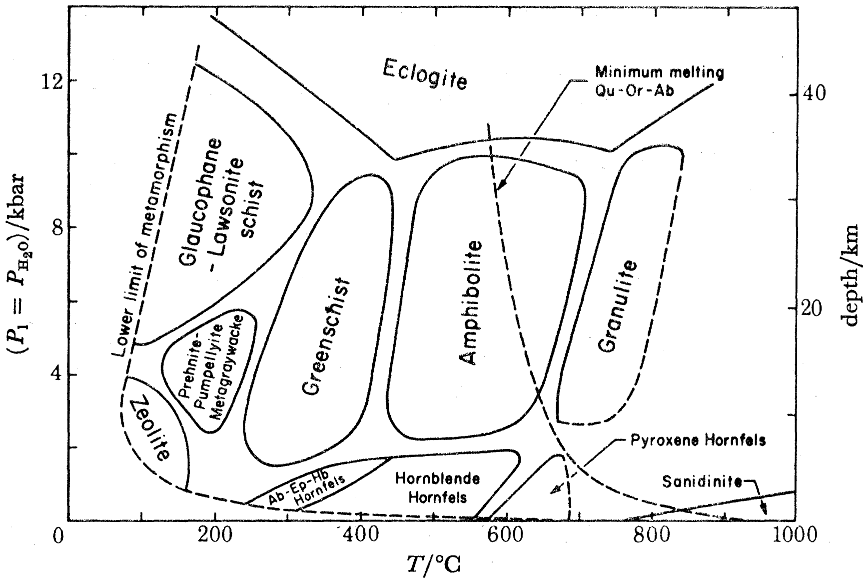

FIGURE 1. Tentative scheme of metamorphic facies in relation to P_l ($= P_{H_2O}$) and T. All boundaries are gradational (from Turner 1968).

2. BEHAVIOUR OF URANIUM IN METAMORPHIC REACTIONS

In spite of all the research which has been carried out concerning the geochemistry of uranium, our knowledge concerning the central question, its behaviour in metamorphic reactions, is remarkably restricted (or non-existent). In the most up-to-date review on the geochemistry of uranium Rogers & Adams (1969) state, 'The mobility of the uranyl ion in surface waters plus the tendency for uranium to occur together with potassium lead to the presumption that uranium is highly involved in metasomatic activity. Unfortunately, data for metamorphic rocks are not yet adequate to test the hypothesis. Heier & Adams (1965) have proposed that high-rank rocks in one area in Norway are impoverished in uranium in comparison with lower-rank rocks owing to upward movement of uranium. This evidence of mobility, however, has not been confirmed in other areas owing to lack of adequate samples.'

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† 1 kbar = 10^8 Pa.

Work in the intervening years has confirmed that medium to high pressure granulite facies rocks are depleted in uranium. This will be discussed later. Otherwise we are as far from understanding its behaviour in metamorphic reactions today as we were then.

3. The occurrence of uranium in metamorphic rocks

Rich *et al.* (1977) in their book on hydrothermal uranium deposits briefly mention the occurrence of uranium in metamorphic rocks with this statement: 'The uranium content of metamorphic rocks is quite variable, and tends to reflect the uranium concentration of their protoliths. However, some very high grade metamorphic rocks are apparently depleted in uranium relative to their lower grade and unmetamorphosed equivalents, and it is possible that uranium moves upward in the crust during granulite grade metamorphism (e.g. see Heier & Adams 1965).'

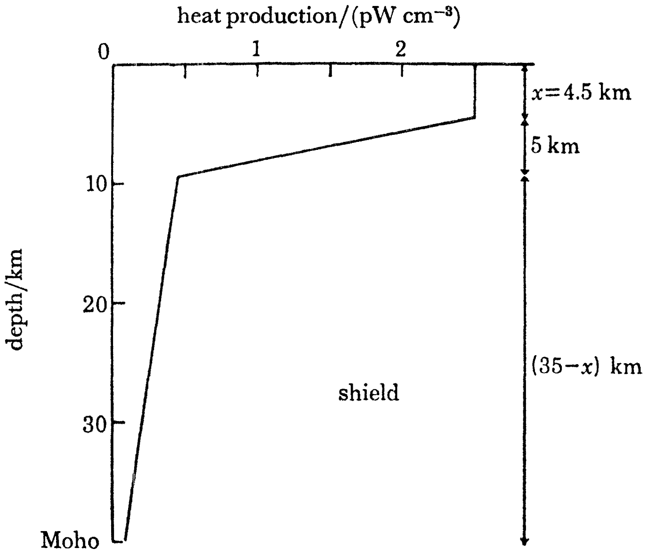

Figure 2. Heat production at various depths within the stable southwest Australian Shield (adapted from Lambert & Heier 1968).

These authors also state, 'average sedimentary rock has about the same uranium content as average igneous rock'. Bearing in mind Barth's statement, 'the diversity of igneous rocks is caused by sedimentary processes', one might simply deduce that metamorphic processes do not significantly affect the uranium concentration of rocks. This is, however, contrary to the overwhelming amount of data collected during the last decade on the distribution of heat producing elements in the crust, and on the occurrence of uranium deposits.

Combined studies of heat flow and surface heat production carried out in the last decade have clearly demonstrated that the uranium of the Earth is strongly concentrated in the upper 10 km or so of the continental crust. References to this are numerous but it will suffice here to refer to the paper by Heier (1974). Based on geological and geochemical data alone an impression is obtained as illustrated in figure 2. The figure clearly demonstrates how heat production in a stable part of the Earth is derived from the upper few kilometres. When this empirical model which assumes a deep continental crust of granulite facies mineralogy and chemistry is compared with the empirical relation combining heat flow and surface heat production,

$$Q = a + bA,$$

an excellent agreement is obtained (figure 3). Here Q is surface heat flow; A is heat production; a is heat flow from the mantle and lower crust (Q_0); and b (the slope of the line) is the distance from surface to the depth for which the radioactivity measured at the surface (A) is constant. The depth of the upper crust is here estimated to be about 7.2 km. On both models it is clearly demonstrated that the heat producing elements, including uranium, are very strongly concentrated in the upper part of the continental crust.

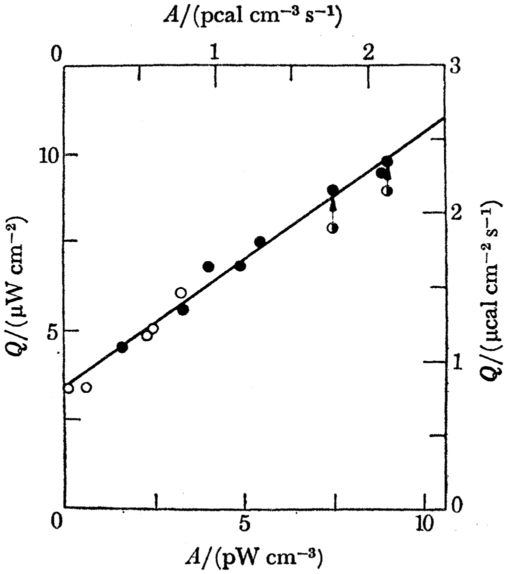

FIGURE 3. Heat flow and heat productivity data for plutons in the New England area (solid circles) and the Central Stable Region (open circles). The line is fitted to both sets of data (adapted from Roy *et al.* 1968).

The uranium depletion in medium to high pressure granulite facies rocks, which may serve as a chemical model for the deep continental crust, has been clearly demonstrated from a number of different areas (figure 4). Economic uranium deposits are found in very low grade metamorphosed sediments as well as in hydrothermal deposits. This again is evidence of the tendency for uranium to concentrate upwards in the crust.

Therefore, even though we do not understand the mechanism(s), the observed facts that uranium is depleted in high grade metamorphic rocks, and that economic uranium deposits occur in the upper parts of the continents, imply that uranium is mobile during metamorphic processes.

Moorbath *et al.* (1969) report lead isotope ratios, together with uranium and lead analyses for thirty-seven whole rock samples from the Precambrian Lewisian basement complex of Northwest Scotland. A fairly representative range of metamorphic and polymetamorphic gneisses, mainly in the amphibolite and pyroxene–granulite facies, has been analysed in this reconnaissance survey. The analysed rocks have a wide range of conventional radiometric dates in the range 2600–1600 Ma, depending on whether they are from the Scourian, Inverian or Laxfordian sectors of the Lewisian complex.

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The leads from these rocks cannot be interpreted by a single stage evolutionary model. The leads fall below the primary growth curve and scatter rather closely about a straight line on a plot of $^{206}Pb/^{204}Pb$ versus $^{207}Pb/^{204}Pb$, which cuts the primary growth curve (with $^{238}U/^{204}Pb = u = 8.68$) at $t_1 + 2900 \pm 100$ Ma and $t_2 = 0$ Ma. This shows that all the analysed rocks underwent varying degrees of uranium depletion at 2900 ± 100 Ma ago, presumably during the early pyroxene–granulite metamorphism. It also suggests that much of the Lewisian basement complex was already in existence about 2900 Ma ago. Later metamorphisms of somewhat lower grade, such as the so-called Inverian and Laxfordian, have probably caused minor redistribution of uranium and/or lead leading to some scatter about the $^{206}Pb/^{204}Pb$ versus $^{207}Pb/^{204}Pb$ lead line, but, in general the uranium/lead ratios of these rocks have always been much lower than that indicated by the primary growth curve.

It is suggested that lead isotope studies on metamorphic and polymetamorphic basement rocks can help to decide whether a given crustal segment represents essentially juvenile addition of material from the upper mantle, or reworked older crust.

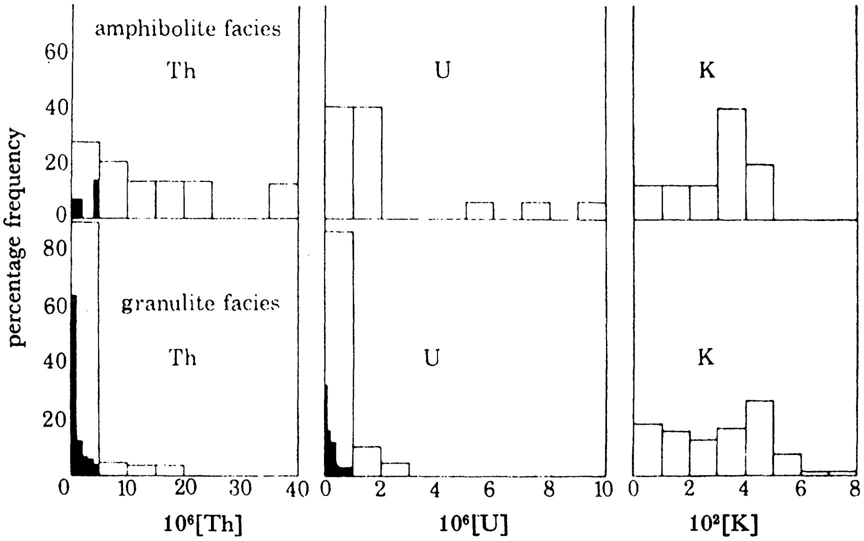

FIGURE 4. Comparison of Th, U and K distributions in neighbouring amphibolite facies and granulite facies terrains, Lofoten–Vesterålen area, North Norway (from Heier 1973).

TABLE 1. URANIUM CONTENT (IN PARTS/10^6) OF IGNEOUS ROCK-FORMING MINERALS

(Data from compilations by Clark *et al.* (1966); Rogers & Adams (1969).)

major minerals	range	accessory minerals	range
quartz	0.1–10	allanite	30–1000
feldspar	0.1–10	apatite	5–150
muscovite	2–8	epidote	20–200
biotite	1–60	garnet	6–30
hornblende	0.2–60	ilmenite	1–50
pyroxene	0.01–50	magnetite	1–30
olivine	–0.05	monazite	500–3000
		sphene	10–700
		xenotine	300–35000
		zircon	100–6000

4. URANIUM IN COMMON ROCK-FORMING MINERALS

The uranium content (in parts/10^6) of igneous rock-forming minerals (data from compilations by Clark *et al.* (1966) and Rogers and Adams (1969) is given in table 1.

The general statement may be made that felsic minerals in igneous rocks normally contain much less uranium than biotite, hornblende and pyroxene. The highest uranium concentrations are found in accessory minerals such as xenotime, zircon, monazite, sphene, allanite, epidote and apatite. Many of the rather high values in common rock-forming minerals are probably the result of solid inclusions of uranium-rich minerals (e.g. zircons in biotite). It is obvious that the tenor of uranium in a rock can be greatly affected by the presence of only small amounts of accessory minerals.

Of the accessory minerals which contain most of the uranium in igneous rocks, we do know that some of them are not stable in high grade metamorphic rocks (granulite facies). These are for instance allanite, epidote and sphene. It is also the author's impression that common accessory minerals like apatite and zircon are not common in high pressure granulite facies rocks. This is just a qualitative observation and it is not known if it has general validity. The possible metamorphic reactions which could cause the breakdown of these two minerals are not understood. Data on zirconium in crystalline rocks indicate that its absolute concentration in granulite facies rocks (lacking zircon) is not less than in igneous and low grade metamorphosed rocks of similar bulk composition containing zircons. Though the data are lacking, a logical assumption might be that zirconium is incorporated in the clinopyroxene structure, and that one may have a reaction involving the breakdown of zircon and the formation of clinopyroxene under granulite facies metamorphic conditions.

As much of the uranium in igneous and sedimentary rocks is present as uraninite, one can easily understand how metamorphic reactions involving dehydration will lead to a loss of much of the uranium originally present and will concentrate it in the upper part of the crust. An explanation for the depletion of uranium in granulite facies rocks should also take into consideration the beginning of incipient melting and the partitioning of uranium and its minerals between the residual rock and newly formed silicate melt. No quantitative data on this are available.

5. HYDROTHERMAL URANIUM DEPOSITS

Perhaps the best impression of the movement of uranium during higher grade metamorphic processes is obtained by a study of hydrothermal uranium deposits. Some points are immediately striking (Rich *et al.* 1977):

(1) Hydrothermal uranium deposits are epithermal or mesothermal in character; the introduced minerals fill open spaces created by structural deformation.

(2) Even though hydrothermal uranium deposits occur in diverse geological environments and in a wide variety of rock types and ages, host rocks in most hydrothermal uranium deposits are competent felsic igneous and metamorphic rocks.

(3) High uranium concentrations usually correlate well with high concentrations of potassium and silica.

(4) The ages of the known hydrothermal uranium deposits range from middle Precambrian to late Tertiary.

All of these observations are consistent with the chemical and geophysical evidence which is for a strong upward increase in uranium concentration in the Earth. Is it possible that the absence of hydrothermal uranium deposits in the Archaean is secondary caused by the processes commented on by Jahn & Nyquist (1976) that 'most of the old crustal segments on the Earth have an imprint of a world-wide thermal event about 2.7×10^9 years ago'? This should be compared with the statement by Moorbath *et al.* (1969) quoted above.

Thus all the examples we know of indicate a strong upward concentration of uranium in the crust. Uranium deposits in high grade metamorphic rocks are unknown. Even though the mechanisms are still not understood it must be inferred that uranium is depleted from these rocks either by leaching (in hydrous solutions as a result of dehydration after progressive regional metamorphism) or as the result of strong partitioning into very early formed anatectic melts.

The first is evidenced by the fact that the richest deposits of uranium ores are found in very low grade metamorphosed sediments (some where oxidizing conditions have prevailed, i.e. sandstones, and others where the concentration of uranium has been caused by reducing activity, (i.e. black shales).

Studies of hydrothermal uranium deposits, so well described by Rich *et al.* (1977) tell the same story. These uranium deposits have formed at temperatures less than 350 °C.

6. Provinces with high Th and U contents

The problem of high-value thorium and uranium provinces has been discussed by Rogers & Adams (1969). There is some evidence for the view that high thorium and uranium concentrations within certain areas are characteristic of igneous rocks formed over considerable periods of time. This, as well as a possible relation between age and Th : U ratios, is a problem that requires further investigation. However, Dahlkamp (1977) demonstrated how statistical studies reveal that certain types of uranium deposits and uranium source rocks prevail in certain geological periods, especially in the Upper Archaean – Lower Proterozoic and Hercynian, and that numerous uranium deposits in sediments and metasediments show a distinct affinity to the uranium rich source rocks, irrespective of age.

In Norway the Precambrian Flå, Iddefjord, and Bohus granites lie along a line striking roughly northwest which crosses the Permian Oslo Province to the southwest of Oslo (figure 5). Radioelement investigations in the three bodies show that they all contain abnormally high thorium and uranium concentrations relative to values in published literature on average radioelement contents of granitic rocks. Trend surface analysis of the radioelement distribution in the Iddefjord granite suggests that there was relative movement of uranium to the east with respect to thorium, possibly as the result of Permian activity in the adjacent rocks. Geological considerations, radiometric evidence and published gravimetric data suggest that the three granites represent a continuous belt enriched in thorium and uranium during the Sveconorwegian orogeny. A portion of the belt was later involved in the Permian igneous activity which produced the igneous Oslo Province. There is some evidence that the Permian Drammen and Finnemarka granites represent a part of the belt which was modified in Permian time.

The radioelement concentrations of the Permian rocks of the Oslo province were studied in detail by Raade (1973). The mean values for 109 samples of the Drammen granite, and 22 samples of the Finnemarka granite are shown in table 2, together with the means for the entire igneous Oslo Province. Raade was able to subdivide the plutonic rocks of the Oslo province into two main groups on the basis of their Th/U ratios. His group 1 has relatively constant Th/U ratios with low mean values (Th/U = 3.5–4.5) while group 2 has high mean values (Th/U = 4.5–6.5) which are very variable (i.e. have high standard deviations). He suggests that 'this clear division of the plutonic rocks undoubtedly reflects a profound difference in the origin and differentiation history of the two main groups.' Group 2 comprises the Nordmarkite, Ekerite and Biotite granites (i.e. the Drammen and Finnemarka granites).

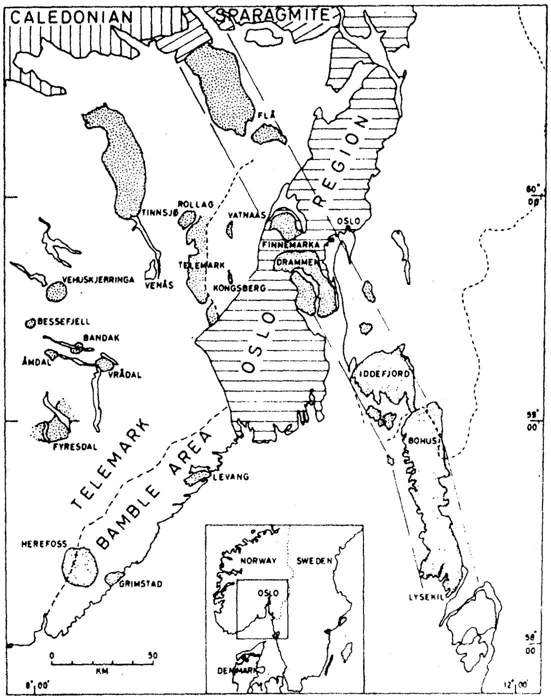

FIGURE 5. Locations of some granitic bodies in southeastern Norway showing the Flå-Iddefjord-Bohus granitic belt cutting across the plutonic rocks of the Oslo region (from Killeen & Heier 1975).

TABLE 2. ARITHMETIC MEANS OF RADIOMETRIC RESULTS FOR TWO PERMIAN GRANITES AND MEANS FOR THE ENTIRE IGNEOUS OSLO PROVINCE (TAKEN FROM RAADE 1973).

							heat production	
granite	10^6[Th]	10^6[U]	K (%)	Th/U	10^4Th/K	10^4U/K	pcal cm^{-3} s^{-1}	pW cm^{-3}
Drammen	27.1	4.7	4.26	6.19	6.43	1.10	0.848	3.54
Finnemarka	21.1	4.6	3.86	5.13	5.48	1.20	0.736	3.08
Oslo Province	19.4	4.3	4.0	—	—	—	0.688	2.88

Raade (1973) explained this group with variable ratios as the result of a process of contamination from overlying Precambrian rocks. He also suggested that selective loss of U by oxidation due to higher oxygen fugacity in the upper crust could explain the highly variable Th/U ratios. The Precambrian rocks which were part of the postulated contamination could very probably include that portion of the Flå–Iddefjord belt which is missing now where it is cut by the igneous rocks of the Permian Oslo region.

7. Conclusions

All of the evidence suggests a very strong upward concentration of uranium in the crust, and that economic uranium deposits are sedimentary or related to relatively low temperature hydrothermal processes. The pronounced depletion of uranium observed in medium to high pressure granulite facies rocks is most probably related to partial melting and a distribution coefficient for uranium strongly in favour of the newly formed melt. However, the most significant factors governing the uranium distribution in the crust are probably low temperature and hydrous conditions. In their book *Hydrothermal uranium deposits*, Rich *et al.* (1977) make these observations:

(*a*) With increasing temperature the solubility of UO_2 in equilibrium with UO_3 probably passes through a maximum near 260 °C. There is a rapid decrease in the solubility of UO_2 below 200 °C and probably also above 300 °C.

(*b*) The pressure of $NaHCO_3$ increases the concentration of uranium in solutions equilibrated with UO_2 and UO_3 throughout the hydrothermal range, although the uranium concentration seems to decrease rapidly with increasing temperatures above 25 °C.

References (Heier)

Clark S. P. Jr, Peterman, Z. E. & Heier, K. S. 1966 In *Handbook of Physical Constants* (ed. S. P. Clark), *Mem. geol. Soc. Am.* **97**, pp. 521–541.

Dahlkamp, F. J. 1977 I.A.E.A. meeting Vienna, 17–21 Nov. 1975 *Publ. IAEA-TC-25/8*, pp. 131–148.

Fyfe, W. S., Turner, F. J. & Verhoogen, J. 1958 *Mem. geol. Soc. Am.* **73**, 259 pages.

Heier, K. S. 1973 *Fortschr. Miner.* **50**, 174–187.

Heier, K. S. 1974 *Centenaire de la Société Geologique de Belgique. Geologie de domaines cristallins, Liege, 1974*, pp. 23–44.

Heier, K. S. & Adams, J. A. S. 1965 *Geo him cosmochim. Acta* **29**, 53–61.

Jahn, B. & Nyquist, L. E. 1976 In *The early history of the Earth* (ed. B. F. Windley), pp. 55–76. London: Wiley.

Killeen, P. G. & Heier, K. S. 1975 *Geochim. cosmochim. Acta* **39**, 1515–1524.

Lambert, I. B. & Heier, K. S. 1968 *Chem. Geol.* **3**, 233–238.

Moorbath, S., Welke, H. & Gale, N. H. 1969 *Earth planet. Sci. Lett.* **5**, 1–12.

Rich, A. A., Holland, H. D. & Peterson, U. 1977 Hydrothermal uranium deposits. *Developments in economic geology*, vol. **6**, 264 pages. Amsterdam: Elsevier.

Rogers, J. J. W. & Adams, J. A. S. 1969 *Uranium, handbook of geochemistry* (ed. K. H. Wedepohl), vol. II–I. Berlin, Heidelberg and New York: Springer-Verlag.

Roy, R. F., Blackwell, D. D. & Birch, F. 1968 *Earth planet. Sci. Lett.* **5**, 1–12.

Raade, G. 1973 Unpubl. cand. real. thesis. (162 pages) University of Olso.

Turner, F. J. 1968 *Metamorphic petrology.* (403 pages.) New York: McGraw-Hill.