THE ALTERATION OF RADIOACTIVE MINERALS

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THE soundness of the "lead-ratio" method of determining the age of a radioactive mineral (and hence of the formation where it occurs) is well established, as the lead ratios obtained from different radioactive minerals from the same locality have been practically identical in many cases. Nevertheless it has been observed by different workers that different radioactive minerals from the same locality, and sometimes from the same pegmatite, indicate different ages; also that the uranium minerals generally indicate a higher age than the thorium minerals and these latter have lead ratios varying greatly among themselves. The case of thorium minerals from Norway and Ceylon is prominently illustrative.

From a study of lead accumulated in thorium minerals Boltwood\(^1\) and later Holmes and Lawson\(^2\) came to the conclusion that lead was not the end product of the disintegration of thorium, and even if it was so, a part only of the lead was stable and the remainder further disintegrated into thallium. Soddy\(^3\) and Lawson\(^4\) considered that the thorium minerals were of a secondary nature and necessarily gave lower age indications than the primary minerals with which they were associated. Against this, Joly\(^5\) appeared to have found evidence that uranium might have disintegrated more rapidly in earlier times than to-day, and consequently uranium minerals would give higher age indications than normal, whereas thorium minerals disintegrating at a constant rate would give correct age values. None of these ideas have been found to be satisfactory and they have been dropped in course of time. The formulation of the above ideas shows the complex nature of the puzzle and how the disagreeing lead ratios of radioactive minerals have affected the development of the fundamentals of radioactivity.

In the opinion of the present writer, the essential condition about the unaltered nature of the mineral was lost sight of by many investigators. We shall consider in this paper how the radioactive minerals undergo alteration and how the lead ratio of such minerals are affected in consequence of the process of alteration. We shall also discuss some relevant points in the light of the ideas presented here.
The radioactive minerals have a great tendency to occur in the metamict state. This phenomenon was first observed by Brögger in the case of the rare earth minerals and was thought by him to be due to hydration. Later Mügge considered that the radioactive substances occurring in these minerals by their disintegration were responsible for the metamict state of the minerals. Goldschmidt noted that minerals that usually occur in the metamict state are mostly combinations of weak acids and weak bases and according to him, the percolating waters cause hydrolysis in the minerals, the components of which then exist as a solid solution of the oxides, and consequently the minerals occur in the metamict state. Vegard thought that such minerals were probably formed under very high pressure, and as they are exposed to the ordinary pressure the crystal lattice breaks up and the change to the metamict state is reached gradually. Probably all the above factors are involved in the transformation of a mineral to the metamict state, and such a mineral is very much likely to be altered in the process of this transformation or subsequent to it by external agencies.

I would like to point out that a mineral in the metamict state is not necessarily in the altered condition. The changes to the metamict state and to the altered condition are due to two different sets of causes. The change to the metamict state signifies a change in the physical condition of the mineral and its composition is unaffected, whilst the alteration of a mineral always changes the composition of the mineral, some of its constituents being removed and replaced by other substances. The transformation to the metamict state is brought about purely by physical agencies, such as, by the radiations from the radioactive elements present in the mineral, and is also probably partly due to a release from immense pressure. These two processes may lead to an increase in volume which destroys the arrangement of atoms in the crystal lattice, thus causing the mineral to behave like an amorphous substance, i.e., the mineral is then in the metamict state.

The presence of solutions is essential for altering a mineral. These solutions may contain metallic salts, silica and also some CO dissolved in them. When a radioactive mineral becomes subject to the action of such solutions any of the following changes may occur: (1) The solution may exert a solvent action on some of the constituents of the mineral. (2) Chemical reactions between dissolved substances in the solution and the mineral may occur. (3) Sorption of substances in the solution by the minerals may take place. (4) Ion exchange may take place. We shall discuss these possibilities individually.

(1) There is an old observation of Brande that hydrated uranium trioxide dissolves in an aqueous solution of CO. That this solvent action