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THE PROBLEM OF METASOMATISM

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ABSTRACT.— Metasomatism consists of volume-by-volume replacement of minerals and rocks, as evidenced by pseudomorphs, preservation of older textures, and bulging contacts with walls which do not «match». Serious petrographers admit that microscopic textures may often be explained in different ways, and refer to geological and geochemical considerations for arbitration, where in doubt. Therefore, geologists should not take the part for the whole, but check hypotheses of metasomatic origin of ore-bodies against field observations, before accepting them.

Metasomatism, according to the definition given by Lindgren in 1900 (AIME Trans. 30, p. 580) is «that variety of metamorphism which involves a chemical change in the composition of the rock, by addition or subtraction of substance».

According to Correns (Einführung in die Mineralogie, 1949, p. 246), the term comprises all changes of rocks involving at least partial preservation of the solid state, which do not take place on the surface of the earth.

Generally, diagenetic processes are not regarded as «metamorphism», but for the purpose of defining metasomatism they may well be included. Oxidation of sulfides on or near the surface of the earth yields striking examples of metasomatism, such as the occurrence of limonite cubes or pentahedra which clearly are pseudomorphs after pyrite. In this case, sulphur has been removed and water and oxygen added to form limonite in the place previously occupied by a pyrite crystal.

According to Lindgren's definition, loss of water contained in a limestone would constitute a metasomatic process, because the chemical composition of the rock is changed «by subtraction of substance». In fact, Correns (*o.c.*, p. 253) restricts the term «metasomatism» to such processes whereby rocks are wholly or partly replaced by reaction products.

The following definition, however, seems to meet the case: «Metasomatism is a process whereby the individual minerals of a rock are replaced by different ones».

The chemical composition of a rock can be changed in three different ways: (1) By impregnation of porous rocks (preferably sandstones or tuffs'); (2) by removal of soluble minerals or their decomposition products (preferably alkalis, carbonates, chlorides and sulphates); (3) by replacement. —Case (1) and (2) as a rule offer no problem. If vugs or pores in an originally porous rock are impregnated by later minerals (such as quartz, calcite, or sulfides, for instance), the original grain of the parent rock will be recognizable. Similarly, cell structures in limonites, dolomites, or vein-quartz often bear witness of the removal of pre-existing mineral fillings. But replacement on a major scale is often taken for grant-

ed where evidence is insufficient, if not entirely lacking. This remark applies particularly to the so-called «pyrometasomatic ore deposits». Lindgren introduced this term to distinguish a certain type of perimagmatic ore deposits from hydrothermal deposits and from contact-metamorphic deposits. Certainly, Lindgren has shown «beyond question that the introduction of material into previously consolidated rocks by metasomatic processes may give very little indication of replacement according to criteria commonly accepted», but he also endorsed Becker's statement (in AIME Trans. 23, 1893, p. 602) that «the theory of the substitution of ore for rock is to be accepted only when there is definite evidence of pseudomorphic molecular replacements». The gist of this statement holds good to the present day, though the idea of molecule-by-molecule replacement has been abandoned, as will be seen below.

Bastin, Graton, Lindgren, Newhouse, Schwartz and Short (Criteria of age relations of minerals with, especial reference to polished sections of ores. *Ec. Geol.* 26, 1931, pp. 561-610) submitted the results of their combined research, and established rules for replacement. Under the heading «Evidences of replacement» (p. 590), they stated: «If one cannot pick out structures or texture in the host, some of which still remain unmodified by the destroying guest, such as crystal form, banding, etc., the fact of replacement cannot be proven except by geological or geochemical reasoning. It must be admitted that certain intergrowths formed by unmixing and others formed by replacement are so similar that no means of distinguishing between them are at present known».— Thus the matter is referred back to geologist, his field observations and his discretion, as well as his prejudices. Unfortunately, it is when field observations do not permit to arrive at a well-founded opinion on the nature of an ore deposit that the geologist will take refuge in petrographic studies and «geochemical reasoning». Geochemical reasoning, however, it must be admitted, is a highly hypothetical procedure, if not based on, or supported by, conclusive field evidence.

Lindgren illustrated his treatise (AIME 30, 1900) with a number of convincing photos, which show beyond doubt metasomatic replacement.

Gilbert (Oxidation and enrichment at Ducktown, Tenn., AIME Trans. 70, 1924, pp. 998-102) reproduces photos (pp. 1007-1012) showing various types of replacement in polished sections.

How chemical comparisons can be used to define changes of substance in rocks is demonstrated by Lindgren in «Contact metamorphism at Bingham, Utah» (*Bull. Geol. Soc. Am.*, 35, 1924, pp. 507-534): «The equation $\text{CaCO}_3 + \text{SiO}_2 = \text{CaSiO}_3 + \text{CO}_2$ would call for a reduction of volume of 31.48 percent, provided that the silica was solid in the limestone. If the silica was introduced from the outside as fluid, there would be an increase of volume of 10.81 percent. I hold, however, that during replacement there has been no change of volumes in general, the volume of replaced material equalling that introduced. As a matter of fact, if there has been no change in volume, the above equation is incorrect, and the reaction involves a loss of CaO as well as of CO₂. This finds expression in the fact that in the altered rocks CaO as well as CO₂ is carried away. But in metamorphic action like that here described the movement of matter has been intense and variable. Two of the six analyses of altered rock show a strong addition of CaO».—Why Lindgren calls the equation incorrect, is not evident. Removal of

CaCO_3 is independent of the reaction of remaining CaCO_3 with silica, and certainly can go hand in hand with it.

In *Journal of Geology* (26, 1918, pp. 542-554), Lindgren made «Volume changes in metamorphism» the subject of a special paper. In this treatise, he adopts (p. 543) a different definition of metasomatism: «Metasomatism is here defined broadly as any change in composition of a mineral when exposed to conditions under which it is unstable. Solutions, gaseous or liquid, effect the change. A more restricted definition is that metasomatism comprises any change in composition of a mineral in a solid rock induced by a change in the physical conditions and resulting in its space being occupied by another mineral stable under the prevailing conditions.—Replacement is used as equivalent to metasomatism».

«Replacement in solid rocks consists in solution of the host mineral, followed immediately by deposition of an equal volume of the guest mineral.... The volume of the replacing mineral equals the volume of the mineral replaced. Deposition follows so closely upon solution that at no time can any open space be discerned under the microscope.... Replacement proceeds particle by particle but not «molecule for molecule».—This paper contains a lot of interesting information concerning metasomatic processes. Among the examples of replacement cited are: Calcite by pyrite, orthoclase by pyrite, chlorite by pyrite, anorthite by calcite. It is shown (p. 547) that conversion of forsterite to iron-free serpentine involves removal of 0.75 gram MgO and 0.28 gram SiO_2 for each cubic centimeter of forsterite changed into serpentine.—Alteration of orthoclase to sericite requires a great addition of alumina, which is supplied by replacement of aluminous silicates by pyrite, calcite, and chlorite—a process which usually accompanies sericitization of feldspars.

Lindgren took up the subject again in his Presidential Address to the Geological Society of America (*Bull. Geol. Soc. Am.* 36, 1925, pp. 247-261). He began with a new definition of metasomatism: «... an essentially simultaneous, molecular process of solution and deposition by which, in the presence of a fluid phase, one mineral is changed to another of differing chemical compositions. Specifically excepted are processes of paramorphism and unmixing processes. Lindgren cites Goldschmidt (*Metasomatic processes in silicate rocks*, *Ec. Geol.* 17, 1922, pp. 105-123): «Metasomatism is a process of alteration which involves enrichment of the rock by new products brought in from the outside. Such enrichment takes place by different chemical reactions between the original minerals and the enriching substances... (water and silica excepted)». Lindgren disagrees (p. 249): «Metasomatism should be a term applied to individuals», not rocks only. Also «definite chemical reactions» are not deemed necessary by Lindgren, to effect metasomatism. (Example: replacement of calcite or limestone by quartz or cryptocrystalline silica.) Lindgren wants to «call attention to the neglected field of colloidal replacement. Substantially, the process is of the same type, whether a chemical equation can be written for it or not, and whether the product is a crystalloid or a gel».

Boydell, in a «Discussion on metasomatism» in *Ec. Geol.* 21, 1926., pp. 1-55, gives a review on older literature and considers the effect of pressure on solubility of mineral substances. Then he discusses at length properties of colloidal solutions and reproduces a dozen photos showing colloidal structures.

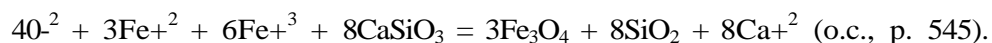
Knox (Criteria of age relations of minerals, *Ec. Geol.* 27, 1932, pp. 95-96) gave a convincing explanation for the fact that pyrite sometimes replaces several different minerals of diverse solubilities simultaneously. In the same year, Butler (*Ec. Geol.* 27, 1932, pp. 1-24) discussed «the influence of the replaced rock on replacement minerals associated with ore deposits».

In his «Mineral Deposits» (1933) Lindgren devotes a whole chapter (pp. 695-745) to Pyrometamorphic Deposits.

In the following year, Schouten (*Ec. Geol.* 29, 1934, pp. 611-658) reported on experiments permitting to study «structures and textures of synthetic replacements in open space», with 38 figures showing textures and structures which are unquestionably due to replacement because they were obtained by experiment under known conditions. But «the solutions used in the experiments were often of such unusual and fantastic composition as to make it necessary to avoid any comparison of these synthetic replacements with natural ones as to mode of origin.» (p. 657). Again (p. 656) The expressions «mineral A projects into» or «cuts» mineral B, are mostly suggestive of original replacement; they do *not* offer conclusive proof concerning the sequence of A and B'. The experiments went to show that structures may develop by the action of solutions on solids which are not typical for metamorphic replacement. In other words, even experiments under known conditions do not yield reliable criteria for the genesis, of replacement ores.

Douglas, Goodman and Milligan (On the nature of replacement. *Ec. Geol.* 41, 1946, pp. 546-553) consider three possible processes : (1) pseudomorphs formed by removal of an already formed mineral, and the filling of the resulting open space by the «replacing» mineral, which exhibits the crystal habit of the original; (2) hydrodynamics: softening and subsequent removal of the host rock and deposition of the «replacing» mineral in the open space so produced; (3) atom shunting: atoms and corresponding «holes» in a crystal lattice move through a crystal. The holes may be filled in by suitable atoms of adequate size and valence. This paper has been discussed and elaborated by «Holser» (Metamorphic processes) in *Ec. Geol.* 42, 1947, pp. 384-395.

In 1949, Ridge (Replacement and the equalling of volume and weight, *Journ. of Geol.* 57, 1949, pp. 522-550) showed that equations can be written for hypogene and supergene replacements, which «are balanced molecularly volumetrically, and electrically». For instance:



It is not maintained that reactions actually take place in this form—«they are, however, possible and in many cases probable explanations of what occurs during replacement reactions». In Ridge's opinion, they demonstrate the truth of Emmons' remark (*Principles of Economic Geology*, 1940, p. 159) that replacement is definitely not a molecule-for-molecule change.

Taupitz (Die verschiedene Deutbarkeit von «metasomatischen» Gefügen auf «telethermalen» Blei-Zink-Erzlagerstätten, *Der Karinthin*, F. 25, 1953, p. 331) showed that ore textures which have previously been considered evidence of the telethermal metamorphic origin of ore deposits in the Northern Alps, Black Forest, and Sauerland, Germany, are chiefly of sedimentary origin (fossilized bacteria, ore psammite, ore breccias).

In 1950, Bastin (Geol. Soc. of America, Memoir 45) presented a comprehensive study on «Interpretation of ore textures». On the subject of metasomatism, he writes (p. 34) : «Usually the processes of solution and deposition go on concurrently through the medium of the same solution and without the development of open spaces—that is, by the process known as replacement or metasomatism..... In other instances, however, minerals are dissolved by one solution, and after an interval, during which open spaces exist, new minerals are deposited in these spaces by solutions of a different composition. This is not replacements Replacement textures are the subject of a special chapter in this book.

SUMMARY AND CONCLUSIONS

The purpose of the preceding review was to trace the subject of metasomatism through geological literature of the past decades in order to elicit points of agreement or dissent between different authors, as well as changing aspects under which the problems involved have been tackled.

Though authors differ in their definition of metasomatism, there is no serious disagreement. Whether supergene processes, that is processes on the surface of the earth (such as limonitization, processes of weathering) should be included under the general term of metasomatism or not, is of no far-reaching consequence. Authors seem to agree that Goldschmidt's definition of metasomatism as a process involving molecule-for-molecule change by definite chemical reactions, is too narrow. Metasomatism differs from metamorphism in so far as a change in the chemical composition of rocks results, whereas metamorphism is a change in the mineral composition of rocks—with or without change in the chemical bulk composition of the rock—preferably under the influence of elevated temperature. If limestone is recrystallized to marble, or marls are converted into hornfels by re-arrangement of the mineral composition, a chemical change is not necessarily involved.

Metasomatism requires addition and subtraction of substance of different chemical composition. Replacement is the process by which metasomatism is effected. Both terms are applicable only if removal of mineral substance is immediately followed by deposition of the replacing substance.

But eminent petrographers admit that replacement cannot in every case be proved by examination of polished sections.

One point which seems to be overlooked, sometimes, is that evidence for replacement on a microscopic scale does not necessarily prove wholesale replacement of large coherent rock masses.

Criteria for replacement which are applicable on a geological scale are (1) pseudomorphs, (2) preservation of pre-existing textures (such as banding, silicified, pyritized or limonitized fossils), (3) irregular contacts with rounded bulges of the replacing minerals into the adjoining host rock (walls that do not «match»).

It has been shown that metasomatism proceeds from tiny cracks, and therefore is a very slow process which cannot be expected to extend very far from the minute cracks which provide channels for the mineralizing solutions.— For instance, where a serpentine is veined by a network of tiny magnesite veinlets

measuring up to a few millimeters, metasomatism may play a significant part. But where magnesite bodies are found with sharp-cut straight walls, forming lenticular coherent veins of several decimeters, if not meters, thickness, filling of open fissures akin to hydrothermal veins seems a more plausible explanation.

Whenever the geologist is tempted to explain the formation of an ore deposit by metasomatism, he should be careful not to overrate the evidence presented by observations in thin slides or polished sections, unless field observations support the assumption that metasomatism occurred on a major scale.

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