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Hydrothermal Synthesis of Calcite

by

N. Yu. Ikornikova and V. P. Butuzov

Doklady Akad. Nauk S. S. S. R. 111, No. 1, 105 (1956)

Translated by

Paul M. Gruzensky

January, 1962

From literature data it is known that small crystals of $CaCO_3$ (on the order of 0.1 - 0.2 mm) have been obtained: (1) from solutions of chloride salts; (2) from alkaline solutions and chloride salt solutions; (3) by reactions of solutions containing calcium ions and carbonic acid. Large crystals of Iceland spar have not been obtained to date. All deposits of large crystals of calcite have a hydrothermal transformation and are related to the so-called contact-metasomatic type. Crystals generally form here as a result of dissolution and reprecipitation of sedimentary limestones or veiny carbonate material as well as hydrothermal interaction with basic extrusive rocks. The hydrothermal origin of large natural calcite crystals indicates the expediency of experimental hydrothermal synthesis of Iceland spar.

For carrying out a hydrothermal synthesis of crystals of any material it is necessary, first of all, to find its solvents and to determine its temperature dependence of solubility. It is known that the solubility of calcite in various aqueous solutions under normal pressures and low temperatures is extremely small.

N. E. Khitarov established (1) that solutions of sodium chloride or calcium chloride, which are neutral at ordinary temperatures, become acidic at high temperatures (in the interval of 300 - 400°) and intensely dissolve many silicate species. It follows from Khitarov's work that there is a direct relationship between temperature, acidity of the chloride salt solution, and solubility of the substance which is influenced by the chloride salt solution. It can be assumed that hydrolysis of aqueous chloride salt solutions takes place at elevated temperatures, leading to the formation of free HCl. These data indicate that: (1) solutions of chloride salts at elevated temperatures should dissolve calcite to a great extent; (2) in view of the fact that the activity of chloride salt solutions is directly connected with temperature, they are favorable media for hydrothermal synthesis of crystals, especially in the case where hydrothermal synthesis is accomplished by a temperature gradient method. For this reason, aqueous

solutions of various chloride salts were selected for evaluation as solvents for calcite.

We carried out a large series of experiments on the solubility and growth of calcite crystals in various chloride salt solutions and in chloride salt solutions with CO_2 at temperatures from 300 to 550° and pressures up to 1300 atmospheres. It was established that calcite crystal growth takes place most readily in solutions of NaCl and $CaCl_2$.

Results of the work on hydrothermal synthesis of calcite lead to the following conclusions:

1. In aqueous solutions of certain chloride salts under definite pressure and temperature values calcite, in principle, dissolves congruently.*
2. The weaker the basic component of the salt, the more readily the aqueous chloride salt solution hydrolyzes and dissolves calcite.
3. Growth of calcite crystals under temperature gradient conditions is more difficult, the greater the extent of hydrolysis of a given chloride salt.
4. Isometric crystals of calcite 5-6 mm in diameter, as well as needle-shaped crystals up to 1 cm in length, can be obtained from solutions of certain chloride salts by lowering the temperature in an autoclave.

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LITERATURE CITED

1. N. E. Khitarov, Doklady Akad. Nauk 94, No. 3, 519 (1954)

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* In the case of congruent solubility of a substance A, no new solid stable phases are formed in the solution which differ in composition from A. In the case of incongruent solubility of a substance A, a stable solid phase B, differing in composition from A, is formed in the solution.