HYDROTHERMAL REACTIONS FOR MATERIALS SCIENCE AND ENGINEERING

An Overview of Research in Japan

HYDROTHERMAL REACTIONS FOR MATERIALS SCIENCE AND ENGINEERING

An Overview of Research in Japan

Edited by

Shigeyuki Sōmiya

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*To Professor Emeritus E. F. Osborn,
The Pennsylvania State University,
University Park, Pennsylvania, USA*

*Without the benefit of study at the Department of Geochemistry,
College of Mineral Industries,
The Pennsylvania State University,
under a Fulbright Travel Grant,
I would not have been able to commence my studies on
hydrothermal reactions*

Preface

According to the late Professor Emeritus Seitaro Tsuboi,¹ the word 'hydrothermal' was used as early as 1849 by a British geologist, Sir Roderick Murchison (1792–1871), in relation to the action of heated water in bringing about change in the earth's crust. The term abounds in later geological literature, and is most frequently met in connection with the processes that take place at a stage near the closing in the course of consolidation of magma. When a cooling magma reaches that stage, the residual liquid contains a large proportion of volatile components, chiefly water, and further cooling results in the formation of minerals of special interest or ore-deposits.

A great concern of Tsuboi's as a petrologist was to elucidate the details of the nature of various actions involved in these 'hydrothermal processes', of which little was known. It is remarkable that, in the last few decades, extensive high-temperature and high-pressure experiments, in which water plays an important role, have become practicable in laboratories, owing to the development of new apparatus and new methods. As a result, the knowledge essential to the elucidation of 'hydrothermal processes' has been improved, but is still far from complete.

On the other hand, we are informed that, apart from the geological interest, research workers in the synthesis of engineering materials are greatly concerned with 'hydrothermal reactions', and that their studies along this line are being carried on efficiently. Armed with this information, Tsuboi considered it highly desirable to provide arrangements for a sort of interdisciplinary conference to discuss subjects of common interest, which would be of benefit to both earth scientists and materials engineers. Over the years, the idea of such a joint meeting occurred repeatedly to him.

In the field of materials science and engineering, ordinary use of the term 'hydrothermal synthesis' usually refers to heterogeneous reactions in aqueous media above 100°C and 1 bar.²

'Hydrothermal crystal growth' is

a process in which an aqueous solution is held at high temperature and pressure in order to dissolve a material (the nutrient) in one part of the system, transport it to another part of the system and there deposit it, either on a seed, or on small crystals attached to the containing vessel by epitaxy. This process is usually carried out in sealed vertical autoclaves with a temperature difference (ΔT) established between the top and bottom of the vessel. Usually the nutrient is placed in the lower, hotter part of the autoclave and the seeds are mounted in the cooler upper part. The seeds are single crystal plates properly oriented and mounted on a suitable wire frame.

Transport of the hot solution from the region containing the nutrient to the region containing the seeds is by convection. Once the solution reaches the growth region it becomes supersaturated with respect to the seed material and deposits on the seeds. The cooler depleted solution then returns to the hotter zone by convection and dissolves more nutrient.³

There are many types of hydrothermal reaction:

1. Hydrothermal treatment.
2. Hydrothermal metamorphism (hydrothermal alternation).
3. Hydrothermal growth.
4. Hydrothermal synthesis (hydrothermal reactions).
5. Hydrothermal dehydration.
6. Hydrothermal decomposition.

7. Hydrothermal extraction.
8. Hydrothermal sintering.
9. Hydrothermal reaction sintering.
10. Hydrothermal hot isostatic processing.
11. Hydrothermal crystallization.
12. Hydrothermal deposition for thin films.
13. Hydrothermal corrosion.
14. Hydrothermal etching.
15. Testing under hydrothermal conditions.

After 1950, Professors E. F. Osborn, O. F. Tuttle and Rustum Roy of the Pennsylvania State University used hydrothermal experiments for mineral synthesis and for phase equilibrium studies. Preparation of homogeneous starting materials is very important in phase equilibrium studies, which is one of the reasons why hydrothermal conditions are used for their preparation.

In Japan, hydrothermal work first appeared in 1926, by Dr Tominosuke Katsurai. His report, entitled 'Soda treatment of the aluminum ore', dealt with hydrothermal extraction. On hydrothermal synthesis, Professor Shoichiro Nagai reported on calcium silicate in 1931. The Morey bomb used after the Second World War was reported by Professor Raisaku Kiyoura in 1952 for hydrothermal alternation and hydrothermal synthesis.

After the mid-1970s there were several reports related to hydrothermal reactions, since such reactions can be considered for use in ceramic processing, such as for powder preparation, reaction sintering, etc. Powder prepared by hydrothermal preparation is not agglomerated, is fine-grained, has a narrow grain size distribution and consists of single crystals. Such processing therefore received wide attention. Many papers have been published in Japan and several companies produce powders such as ZrO_2 under hydrothermal conditions. Reports relating to hydrothermal reactions in such fields as powder preparation, sintering, corrosion, crystal growth, etc., also appeared.

Following the First International Symposium on Hydrothermal Reactions in March 1982, it was decided to maintain communications between workers in different fields, and on 25 September 1982 the Hydrothermal Reactions Study Group was established. Since then papers related to hydrothermal reactions have increased, both in Japanese and in English.

This book is a compilation of English language reports by Japanese workers on hydrothermal reactions related to materials science and engineering. The book shows the development and the present status of R & D in Japan. The papers are scattered in many journals, which is one reason for collecting them together. I am convinced that this book will promote the R & D of hydrothermal reactions.

Shigeyuki Sōmiya

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3. R. A. Laudise, *Solid State Phys.*, **12** (1961) 150.

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oxide Ceramics (1978, Japan)
1st International Symposium on Hydrothermal Reactions (1982, Japan)
Powder Metallurgy Conference (1986, Germany)

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