STABILIZING SOLIDIFICATION II: FUNDAMENTAL TEST OF THE WASTE SOLIDIFICATION PROCESS UTILIZING HYDROTHERMAL REACTION

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ABSTRACT

For the purpose of developing the solidification system able to transform the powder wastes generated from the calciner (or dryer) and incinerator in the nuclear plant to the long-term stable and rigid form, the fundamental test of the hydrothermal solidification process was performed. The results are presented in this paper. From these results we obtained fair prospect of the practical use of this process.

INTRODUCTION

Now, in Japan calcination (or drying) system of liquid wastes is planned in the future plants owing to its high volume reducibility. Products from this new system are in powdery form, therefore they must be solidified to ensure the safe storage or disposal.

The hot-press solidification process utilizing alkaline hydrothermal reaction is one of the hopeful methods to transform the aforementioned powder wastes and incinerator ashes to the long-term stable and rigid form. Hydrothermal reaction is characterized by the extremely active water at relatively higher pressure and temperature than normal condition. By utilizing this reaction, radioactive wastes are solidified with natural solidification material to synthesized rock.

In this research the solidification tests of various wastes were performed and the effects of operational conditions of pressure, temperature etc. on the properties of the solidified form were obtained. These results are presented in this paper. Besides the study on the practical use of this process are also introduced in this paper.

SOLIDIFICATION PROCESS - HYDROTHERMAL REACTION

Solidification process utilizing hydrothermal reaction is shown in Fig. 1. After mixed with natural solidification material (e.g., silica stone etc.), alkali and water, radioactive wastes composed of calciner product, incinerator ashes etc., are solidified by being kept in the hydrothermal condition (high temperature and pressure condition of water). The principle of this solidification is inferred as the following. By this process the radioactive wastes are solidified to the stable and rigid form, where the radionuclides are encapsulated with long-term reliability.

- Ionizing reaction and hydrolysis of water is extremely active on the condition of high pressure and temperature (e.g. ∿ 500 kg/cm^2, ∿300°C). By the medium of this water dissolution and eduction (recrystalization of matrix and waste are highly accelerated. As a result of this reaction, agglomeration of each particle is transformed into crystalline structure solid. (Refer to Fig. 2.)

TEST EQUIPMENT AND CONDITION

The hot-press cell for the hydrothermal reaction is shown in Fig. 3. The cell is a cylindrical type, 20 mm inner diameter and 120 mm height heated from the outside by the electric heater. Material to be solidified is put into the cylinder, pressed by the upper and lower pistons to prescribed pressure, and then heated by the heater to the prescribed temperature. After kept in this condition for prescribed hour, the cylinder is forced-cooled and the solidified product is removed out of the cylinder by push from one direction.

As matrix we used mainly mixture of silicon dioxide (SiO_2) and aluminum hydroxide ($A\ell(OH_3)$) because natural rocks are composed of the above material for the most part and we can make sure reproducibility of test data by using these reagents. In addition to this silica stone-terra alba, coal ashes, glass, glassy volcanic ashes, etc., were tested partially. Sodium hydroxide, calcium hydroxide, barium hydroxide were used as the alkali. Calciner products of boric acid (mainly tested), sodium sulfate and incinerator ashes were used as the simulated waste. The process conditions are shown as follows:

Pressure : 100∿ 500 kg/cm^2

Temperature : 150∿ 300°C

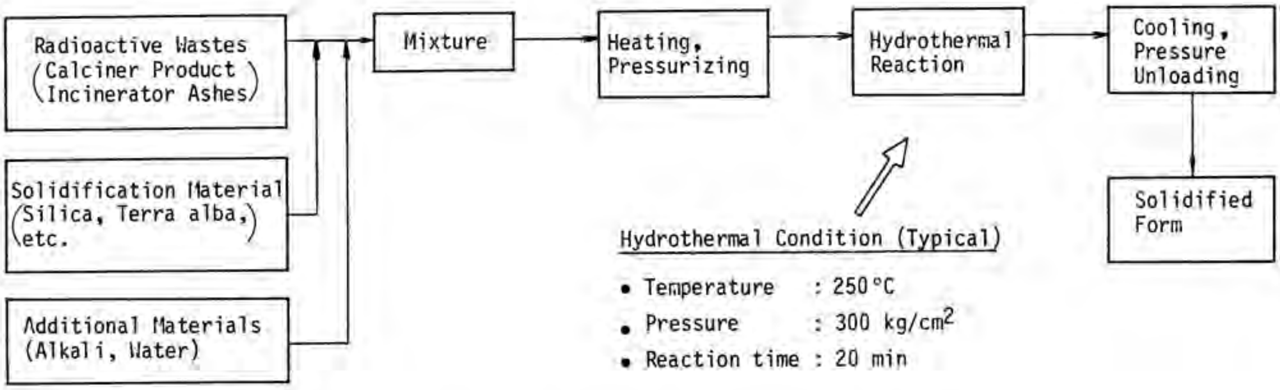

Fig. 1. Hydrothermal solidification process.

Reaction time	: ∿ 30 min
Waste content	: ∿ 50 wt%
Alkali content	: ∿ 20 m mol
Water	: ∿ 10 wt%

TEST RESULTS

Parametric survey was made on the above conditions. From these results, effect of each condition on the properties of solidified form was found. The following are the results of the solidification tests using silicon dioxide-aluminum hydroxide as the matrix and calciner product of boric acid as the simulated waste.

Effect of reaction pressure on the properties of solidified form is shown in Fig. 4. Strength and density of solidified form increase in proportion to reaction pressure.

Effect of reaction temperature on the properties of solidified form is shown in Fig. 5. From this result, the preferable reaction temperature for the stable solidification is found to be above 250°C.

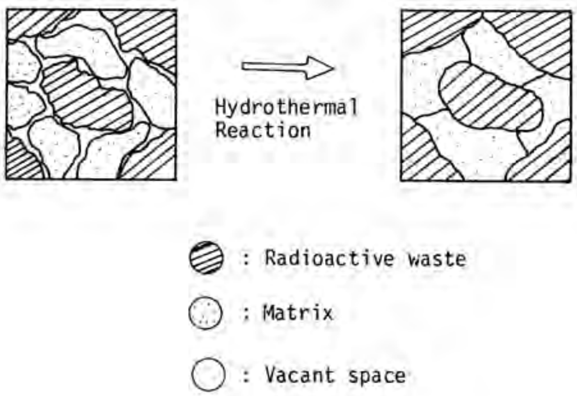

Fig. 2. Conceptual figure of hydrothermal solidification.

Volume change during reaction is shown in Fig. 6. During five minutes from the start of reaction rapid contraction occurrs. After that, gradual expansion occurrs conversely, finally resulting in certain volume in about 15 minutes.

Effect of waste content on the properties of solidified form is shown in Fig. 7. Although compressive strength of solidified form decreases gradually in proportion to waste content, even at 50 wt% content of waste high compressive strength is obtained, which is about 700 kg/cm^2. From this result, it is found that solidification of quite large content of waste is possible. Therefore volume reducibility of this process is found to be high.

Effect of alkali content on the properties of solidified form is shown in Fig. 8. From this result, preferable content of sodium hydroxide (NaOH) is found to be above 5 m mol. In case of calcium hydroxide ($Ca(OH)_2$) and barium hydroxide ($Ba(OH)_2$), compressive strength is slightly lower than sodium hydroxide ($Na(OH)$).

Concerning matrix, it was found that satisfactory solidification by use of the aforementioned natural matrices was possible in spite of some difference in the properties between each matrix.

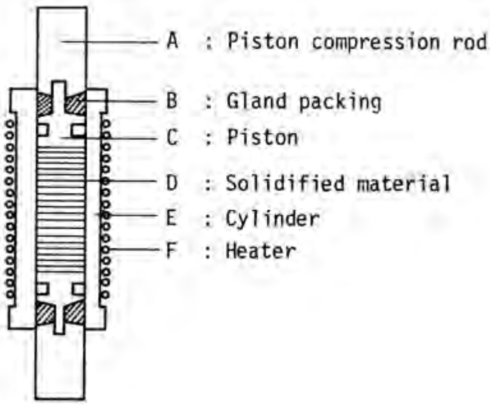

Fig. 3. Hot-press cell.

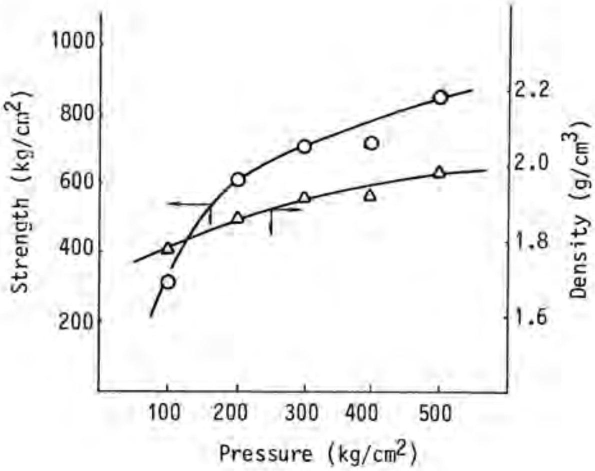

Fig. 4. Effect of pressure.

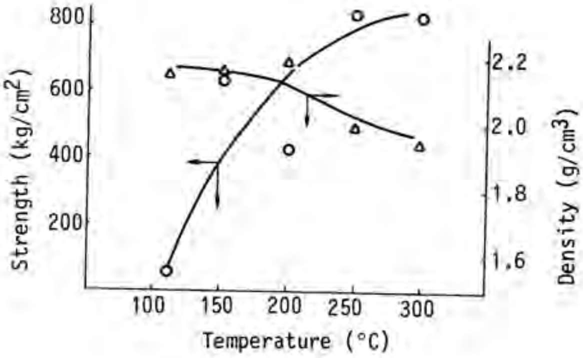

Fig. 5. Effect of temperature.

Besides it was ascertained that satisfactory solidification of not only calciner products of boric acid but also calciner products of sodium sulfate and incinerator ashes were possible.

Concerning leaching rate, precise study and test are now in progress. From the test result until now leaching rate appears to be about $10^{-4} \sim 10^{-5}$g/cm^2 for Cs, <10^{-6} g/cm^2 day for Co on the standard solidification condition; 300 kg/cm^2, 250°C, 20 min, 30 wt% of waste. Besides it is found that leaching rate can be lowered further by use of special additives.

Concerning sacle-up of solidified form, it was ascertained by test that it is possible to produce solidified form up to at least 100 mm outer diameter. Henceforth test concerning larger scale solidified form will be performed.

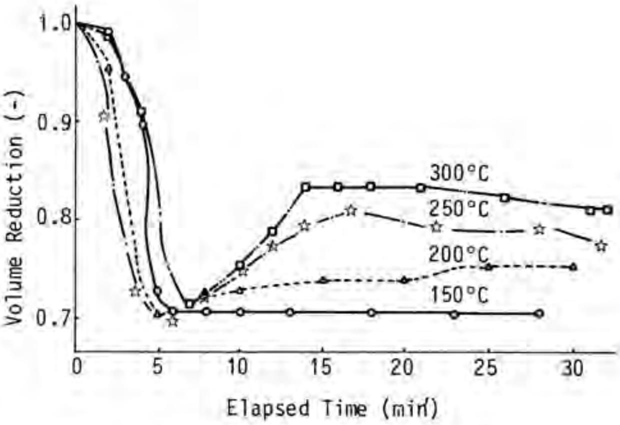

Fig. 6. Volume change during reaction.

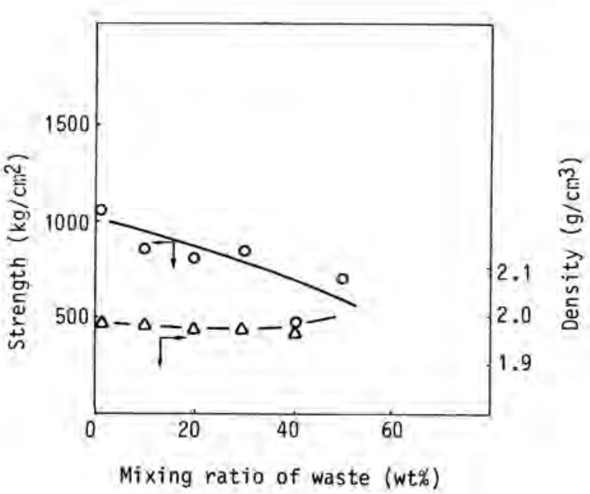

Fig. 7. Effect of waste content.

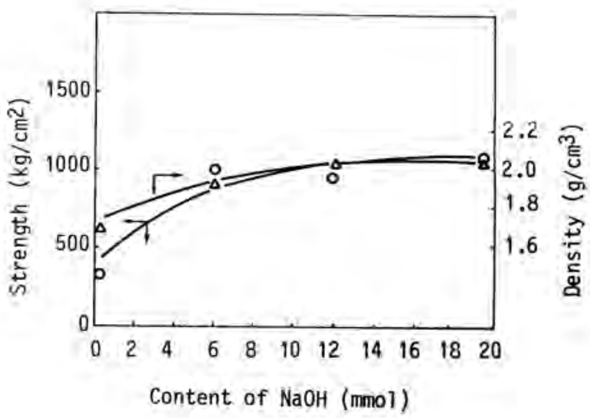

Fig. 8. Effect of alkali content.

STUDY ON PRACTICAL USE

Conceptual study on the practical use of hydrothermal solidification is introduced here. Twin 1200 MWe Class PWR Plants with calciner and incinerator facility is selected as the applied plant. The solidified waste volume produced from the above plants is approximately 20,000 kg/year of the calciner product and 2,500 kg/year of the incinerator ashes. From the above, the soludification capacity per day is estimated roughly to 0.5 ∿ 1 drum (DOT 55 gallon)/day on the standard condition (30 wt% of waste content). Conceptual figure of this solidification process planned on the above estimation is shown in Fig. 9. The solidified form, whose optimization depends on the ongoing scale-up study, is scaled up to DOT 55 gallon drum size. The solidification capacity of this planned equipment is approximately 2 ∿ 3 drums/day.

CONCLUSIONS

It was ascertained that stable and rigid solidified form with high volume reducibility could be produced by the hydrothermal solidification process under the relatively mild conditions, e.g. 300 kg/cm^2 compression pressure, 250°C reaction temperature, 20 minutes reaction time. Besides this solidified form is expected to possess superiority in leachability. From these studies we obtained fair prospect of the practical use of the hydrothermal solidification process. Henceforth larger scale test will be performed to obtain the engineering data for the design of the practical facility, while the evaluation on the physical properties of the solidified product for the final disposal will be also performed.

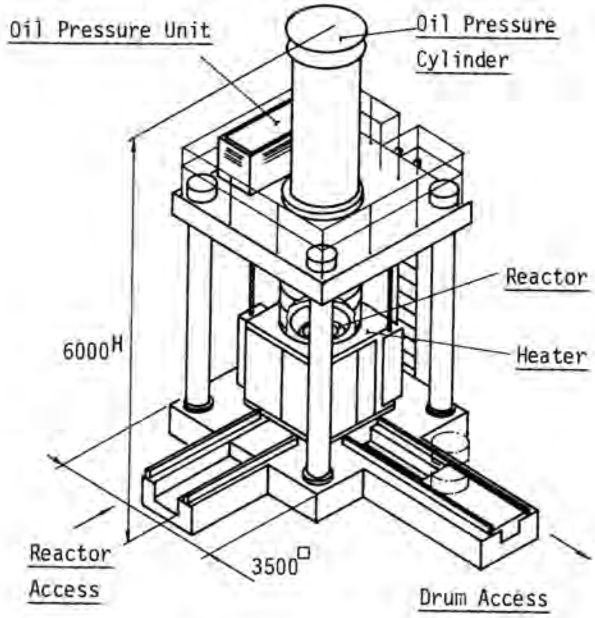

Fig. 9. Concept of full-scale solidification equipment.