PELLETIZING METHOD FOR CONCENTRATED WASTES GENERATED FROM FUEL REPROCESSING PLANTS

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ABSTRACT

A pelletized form of the radioactive wastes is considered to be the most suitable for the interim storage of radioactive wastes in Japan. The simulated concentrated wastes from the reprocessing plant were dried and pelletized under uniaxial pressure using the binder of the inorganic constituents which was prepared for this pelletizing treatment. The binder incorporated the dried powder of the concentrated wastes into a stable pellet block with the condensation polymerization of the colloidal-silica constituent and hydration of the cement constituent. The properties of the pellets were investigated from the viewpoint of the safety during the interim storage. The measurements of their mechanical and thermal properties, and the durability test under the gamma ray irradiation were carried out and the sufficient properties of pellets for the safe interim storage were confirmed.

INTRODUCTION

The low and intermediate level concentrated wastes generated from fuel reprocessing plants contain alpha, beta and gamma nuclides. The alpha nuclides are biologically hazardous in comparison with the other nuclides. So the disposal of these wastes should be strictly regulated.

In Japan, the regulations for disposal of these wastes have not been prepared. These contaminated wastes are, therefore, to be stored at the storage facilities in the reprocessing plants for the interim period until the regulations are prepared and the site of final disposal is established. The most suitable form of these storage wastes is considered to be pellets and the pelletizing system are planned to be developed.

The requirements for the pellets are as follows:

1. The small volume of radioactive waste.

2. No dispersion of radioactivities to the environment during the interim storage.

3. The flexibility of the various packaged forms satisfying the final disposal conditions.

We developed the inorganic binder for pelletizing these wastes and made it possible to produce stable pellets. By use of a binder, pellets are expected to be improved on their properties such as the mechanical strength, the containment of radioactivities and the durability under various conditions. Pellets produced in this process are also expected to be packaged in a drum or a canister with such agents as cement grout for the final disposal.

The properties of the pellets obtained in this experiment are report here.

INORGANIC BINDER

The inorganic binder shown in Table I consists of solution and powder. The solution whose main component is colloidal silica makes the binder adhesive. The powder mainly consisting of cements initiates and promotes the reaction in the binder. The reaction begins by mixing both components of the solution and the powder. The reaction of this binder is schematically presented in Fig. 1. Colloidal silica is condensed to become an inorganic polymer. The particles of powdered wastes adhere with the polymer. The water, which is introduced with the solution of binder and formed in the condensation reaction of colloidal silica, is absorbed into cements in powder with the hydration reaction.

EXPERIMENTAL

The flow diagram of the drying and pelletizing process are shown in Fig. 2. The concentrated liquid wastes were dried into powder with the vertical wiped film dryer. The contents of the simulated concentrated wastes are shown in Table II. The concentrated liquid wastes mainly consist of $NaNO_3$ and Na_2CO_3. $NaNO_3$ comes from HNO_3 neutralizing with NaOH and Na_2CO_3 comes from the solvent recovery process in reprocessing plants. We prepared five different samples of simulated concentrated

TABLE I

Consitituent of Inorganic Binder and Its Reaction

	Form	Main Component	Main Reaction
Adhesive	Solution	Colloidal silica	Condensation
Accelerator	Powder	Cement	Acceleration of Condensation and Water Absorption

wastes because the composition of the concentrated wastes depended on plant operation.

The powdered wastes and inorganic binder were mixed with Henshel's type mixer. The mixture containing 10% binder by weight was pelletized. The pellet was a cylindrical shape, 30 mm in diameter and 30 mm in length.

In order to evaluate the volume and properties of pellets, the bulk density and the compressive strength were measured under non-irradiation and irradiation by ^{60}Co gamma ray and the differential thermal analysis (DTA) was also carried out.

RESULTS AND DISCUSSION

The relationship between the compressive strength of pellets and the curing time is shown in Fig. 3. The pellets consisted of inorganic binder and powdered wastes. The composition of the wastes is shown in Table II as Run No. 1. The compressive strength of pellets increased with curing time until 28 days and was considered to continue the gradual increase after 28 days. The strength of the pellet is considered to come from the cohesion of the powder of itself and the combining strength of the binder due to its adhesiveness. Just after the powder was pressed for pelletizing, the pelletized powder showed the strength due to its

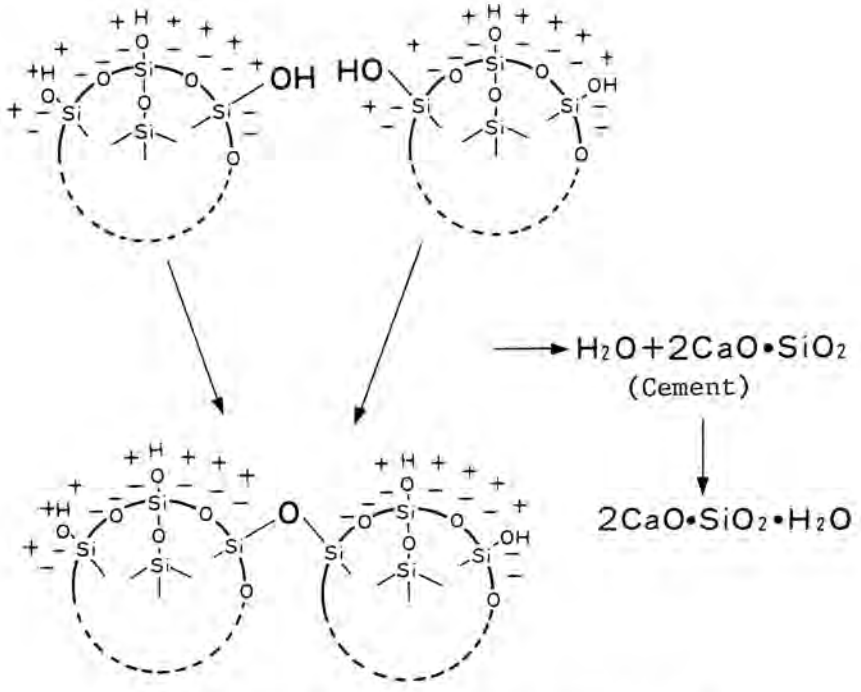

Fig. 1. Condensation Reaction in Inorganic Binder.

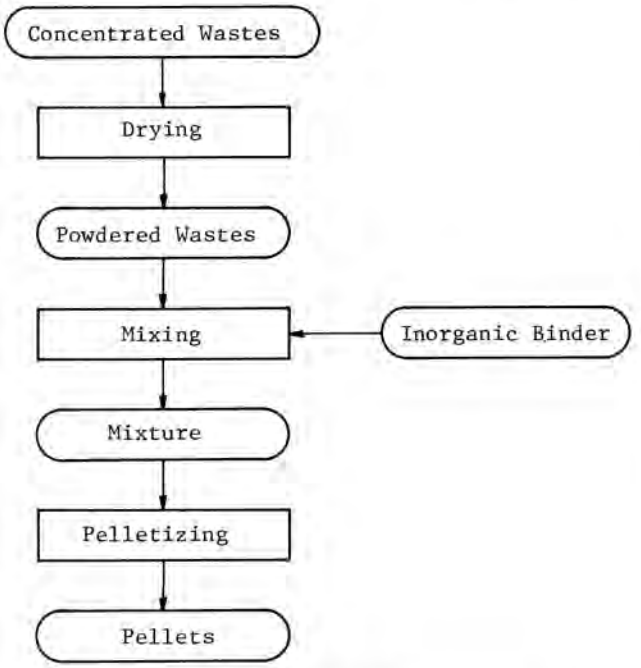

Fig. 2. Flow Diagram of Drying and Pelletizing Process.

cohesion. The binder began to polymerize with condensation and adhere to particles in the powder after pelletizing, but reacted relatively slow and it took some time to complete its reaction. This was the reason for the increase of the compressive strength of pellets after the pelletizing. It was found that the condensation reaction of this binder continued after 28 days and it took a long aging time just as it took a long aging time for cement to complete its aging.

The analysis of the pellet with EPMA (Electron Probe Micro Analysis) containing the same powdered wastes mentioned above (Run No. 1) is shown in Fig. 4. These photos in this figure were observed at the same place in the pellet. The secondary electron image of the pellet is shown in Fig. 4 (a). Characteristic X-ray image of Na-Kα and Si-Kα are shown in Fig. 4 (b) and (c) respectively. The particles shown in Fig. 4 (a) are presumed to be powdered sodium nitrate, because characteristic X-ray of Na-Kα shown in Fig. 4 (b) comes from bulks of these particles. On the other hand, in that of Si-Kα, Si is a main component of binder, and comes from the boundaries of these particles as seen in Fig. 4 (c). From these observations, the binder exists between the particles of powdered wastes and adheres to them.

The bulk densities and the compressive strength of pellets are summarized in Table III. The density of the $NaNO_3$ pellets was larger than those of $NaNO_3$-Na_2CO_3 pellets. And the densities of these pellets depended on their Na_2CO_3 concentration. These pellets were compressed to about 80% of theoretical density (about 2.4 g/cm^3) of the mixture. The compressive strength of pellets obtained in this experiment was larger than 300 kgf/cm^2. The required compressive strength of pellets is calculated to be about 0.5 kgf/cm^2 in case of storage in a 200-liter drum and about 7 kgf/cm^2 in case of storage in the tank whose depth is 10 m

TABLE II

Contents of Simulated Concentrated Wastes* and Properties of Powdered Wastes

Run No.		1	2	3	4	5
Contents of Concentrated Wastes	$NaNO_3$ (wt. %)	30	29	28.5	27	24
	Na_2CO_3 (wt. %)	-	1	1.5	3	6
	Water (wt. %)	70	70	70	70	70
Properties of Powdered Wastes	Apparent Density (g/cm^3)	0.86	0.78	0.79	0.79	0.92
	Average Size of Powder (μm)	35	84	40	41	40
	Angle of Repose (degree)	48	48	46	45	48
	Water Contents (wt. %)	<0.5	<0.5	<0.5	<0.5	<0.5

* density; 1.2g/cm^3

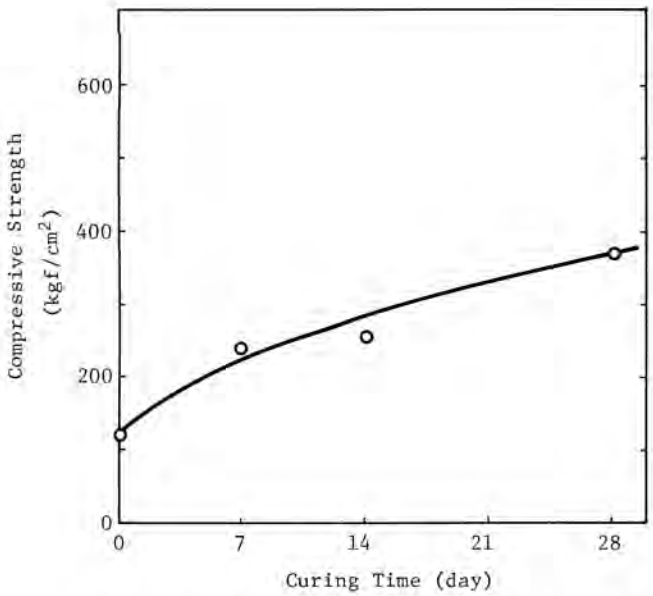

Fig. 3. Relationship Between Compressive Strength of Pellets and Curing Time.

respectively. In consequence, these pellets have sufficient compressive strength during storage.

The effect of radiation exposure on the degradation of compressive strength of pellets is shown in Fig. 5. The pellets from reprocessing plants include radioactivities and are subjected to radiation exposure during the storage period. The cumulative absorbed dose is estimated to be about 10^6 rad. The compressive strength did not change as seen in Fig. 5, even though the absorbed dose was increased up to 10^8 rad. From the above results, we found that the pellets are stable during storage.

The results of DTA are shown in Fig. 6. The curve (A) represents the thermal reaction of pellets including inorganic binder and curve (B) represents that of pure sodium nitrate reagent which is a main component of concentrated wastes. Both curves agree with each other very well. Thus, it was proved that the reaction between inorganic binder and $NaNO_3$ did not occur in pellet. The solution containing colloidal-silica and powder mainly consisting of cements were therefore adopted as an excellent binder since the organic binder might be oxidized by $NaNO_3$ resulting in combustion.

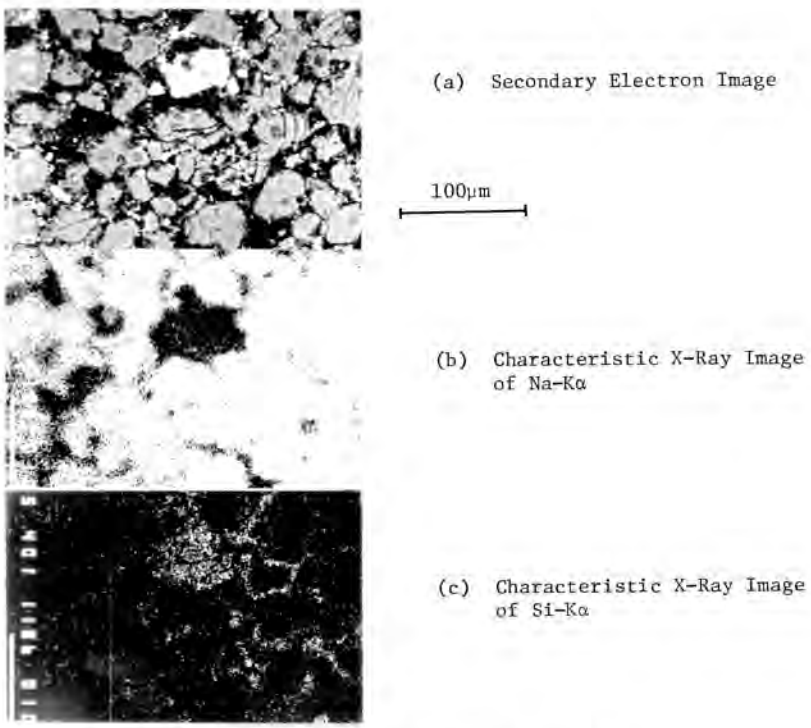

Fig. 4. Analysis of Pellet with EPMA.

TABLE III

Properties of Pellets

Run No.	1	2	3	4	5
Bulk Density (g/cm^3)	1.99	1.91	1.88	1.87	1.86
Compressive Strength* (kgf/cm^2)	370	485	510	520	495

* After 28 days

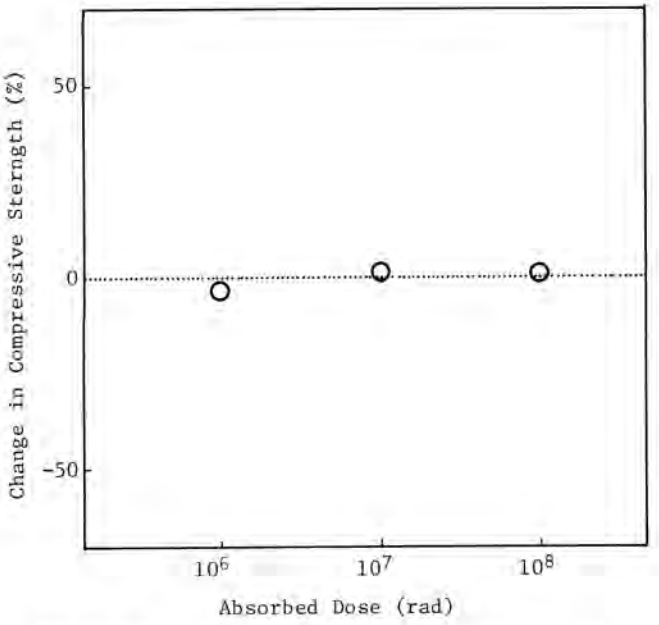

Fig. 5. Effect of Radiation Exposure on Degradation of Eompressive Strength of Pellets (Pellets are irradiated by ^{60}Co-gamma ray).

Calculation from density data in this experiment suggested that the large volume reduction of concentrated wastes was attainable by this pelletizing method and its reduction ratio was about one-third of the original.

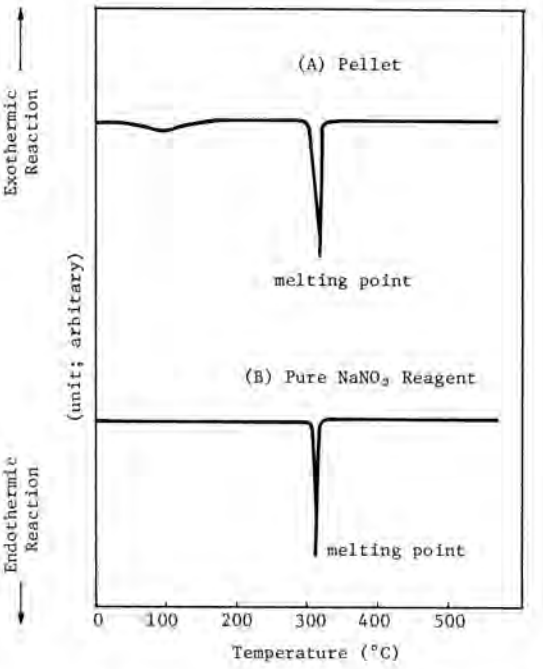

Fig. 6. DTA Curve of Pellet Produced in Run No. 1 and Pure $NaNO_3$

These pellets were encapsulated with cement grout as the disposable homogeneous solid block and its compressive strength was about 80-120 kgf/cm^2.

CONCLUSION

We developed the inorganic binder for pelletizing the concentrated wastes, aiming at the interim storage. This binder was mixed with the powdered wastes and the mixture was pelletized. The properties of these pellets such as the bulk density and the compressive strength under non-irradiation and irradiation by gamma ray were investigated. The differential thermal analysis was also done. From these results, the pellets produced in this experiment have sufficient properties for interim storage. And volume of pellets for storage are reduced about one-third in comparison with the original concentrated wastes. We are planning to develop the pelletizing system for the wastes from fuel reprocessing plants.