Engineering Element Evaluation of Dispersible Waste Solidification Using Substitute Waste

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1. Introduction

The encapsulation method using cement as a solidification technique for radioactive waste disposal is the most widely used approach in most advanced nuclear countries due to its advantages, such as low material costs and accumulated technological expertise. However, during the cement solidification process, moisture or hydroxyl groups within the cement can be decomposed by radiation, generating gas. This phenomenon can degrade structural stability and reduce chemical durability, leading to increased leachability. To address these issues, various alternative solidification matrices are being developed [1].

One such alternative currently being researched is a powder metallurgy-based solidification method for final glass composite material (GCM) solidification, targeting dispersed metallic compounds generated in incineration alternative processes. This solidification process includes the homogeneous mixing of target powder materials and additives used in the sintering process, the formation of a green body using a forming mold, and the final heat treatment stage. To overcome mechanical challenges and interference issues between processing elements and the matrix during these processes, the development of powder metallurgy-based engineering technologies is required. Previous studies have investigated the process factors and engineering considerations for solidification devices designed for dispersible radioactive waste [2]. A prototype solidification system for dispersible waste has been developed, and its initial performance was evaluated experimentally [3]. Alongside the development of radioactive waste solidification technologies, a comparative analysis of acceptance criteria for solidified waste at domestic and international disposal facilities has also been conducted [4].

The characteristics of the final solidified radioactive waste material may vary depending on the specific solidification treatment processes employed at each stage. This study describes the types and characteristics of dispersed radioactive waste generated from an integrated dismantling waste treatment process and examines the green bodies and sintered bodies produced using selected glass composite matrix-based substitute waste for the establishment of a dispersed radioactive waste solidification technology.

2. Methods

A substitute waste based on a glass composite material was produced for dispersed waste generated from the integrated dismantling waste treatment process. Using this material, medium-scale green bodies and sintered bodies were fabricated.

2.1. Methods for Producing Substitute Waste

The substitute waste for green body and sintered body fabrication was prepared by mixing Zeolite powder with various additives and distilled water, as shown in Table I.

Table I. Composition ratio of the substitute waste material

Type	Mass[g]	Reference
Zeolite 4A	50.0	500 ml
$K4Fe(CN)_6 \cdot 3H_2O$	38.4	500 ml
$CoCl_2 \cdot 6H_2O$	21.63	500 ml
$CaCl_2 \cdot 2H_2O$	29.28	1 L
$AlCl_3 \cdot 6H_2O$	33.49	1 L
$MgCl_2 \cdot 6H_2O$	17.28	1 L

The substitute waste material in liquid form was subjected to solid-liquid separation using a filter press, followed by drying and particle size classification using a mesh sieve. To meet the disposal site acceptance criteria for compressive strength, 10 wt.% of B_2O_3 was added to the dried material, along with a certain amount of the filtrate (200 mL per 1 L of the raw material).

2.2. Methods for Producing Green Body

The substitute waste material was used to fabricate green bodies using forming molds with diameters of 3 cm (ID30), 5 cm (ID50), and 10 cm (ID100), as shown in Fig. 1. The forming pressure was gradually increased in 10 MPa increments for green body formation, and

the AirBack condition was applied to produce the green bodies. After reaching the desired forming pressure, the pressure was maintained for a specific time (1 minute).

Fig. 1. The green body formation process using forming molds

2.3. Methods for Producing Sintered Body

The sintered body fabrication was performed by increasing the temperature at a constant rate of 8°C per minute up to 650°C, followed by a 2-hour hold at that temperature, and then allowing it to cool naturally, as shown in Fig. 2

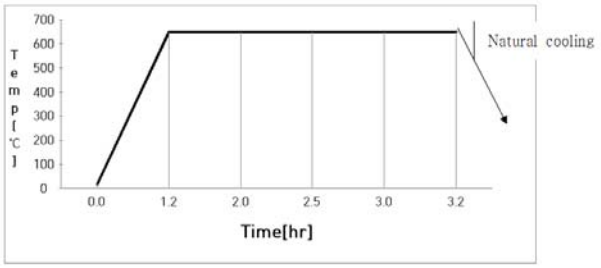


Fig. 2. Methods for increasing sintering temperature

These temperature conditions are identical to the sintering temperature conditions applied to cylindrical green bodies in a lab-scale study using substitute waste materials.

3. Results

3.1. Results of Substitute Waste Production

To ensure the uniformity of the substitute waste material during its preparation, particle size distribution and mixing homogeneity analyses were conducted, along with particle size and tap density measurements. For liquid materials, ICP-MS composition analysis was conducted, while for powder materials, SEM/EDS composition analysis was performed. Table II presents the compositional ratios of the substitute waste materials in cake form for each batch.

Tap density and particle size analysis were also conducted on the classified materials before Green body fabrication. As shown in Fig. 3, the particle size analysis results indicated that all materials had particle sizes within the range of 2–3 μm. Therefore, it was confirmed that the composition and particle size of the substitute waste materials used in the experiment showed no significant differences between materials and remained consistent.

Table II. The composition of the substitute waste material in cake form for each trial.

Element	Trial 1	Trial 2	Trial 3	Average
	wt%	wt%	wt%	wt%
C	15.22	8.80	10.07	11.36
O	6.48	2.64	25.16	11.43
Na	0.23	0.70	2.81	1.25
Mg	0.32	0.23	0.31	0.29
Al	2.08	6.99	12.53	7.20
Si	0.87	7.12	11.68	6.56
Cl	34.70	33.42	12.45	26.86
K	39.57	36.09	17.32	30.99
Ca	NA	2.64	3.62	3.13
Fe	0.53	0.62	1.95	1.03
Co	NA	0.75	2.10	1.43
Totals	100.00	100.00	100.00	

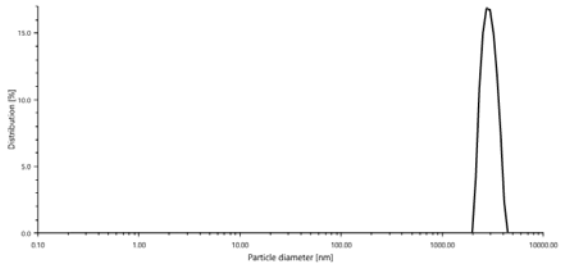


Fig. 3. The particle size analysis results for the substitute waste material

3.2. Results of Green Body

The characterization of the green body using substitute waste materials was conducted based on the size of the forming mold. For a tap density of approximately 1.1 g/cm³, the required material amount was analyzed and fabricated to ensure that the thickness of the green body remained between 0.75 and 0.8 cm. Table III shows the results of green body fabrication based on the material amount and tap density for each forming mold using substitute waste. The volume reduction of the green body using substitute waste materials was confirmed to be approximately 20–24%. The lower volume reduction of the green body fabricated with the ID100 forming mold, compared to the values obtained with the ID30 and ID50 forming molds, is attributed to the slight increase in the tap density of the material.

Table III. Results of green body

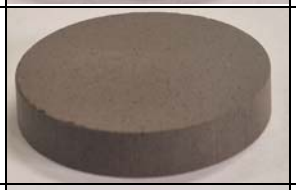
	Forming Mold		
	ID30	ID50	ID100
Material Amount [g]	8	22.24	88
Tap Density[g/cm3]	1.099	1.099	1.150
Thickness[cm]	0.782	0.777	0.769

Volume Reduction[%]	23.46	23.95	20.59

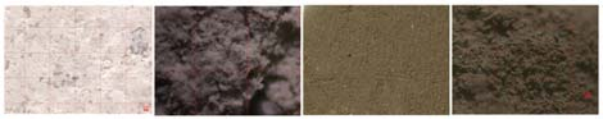
3.3. Results of Sintered Body

The sintering of the green body using substitute waste follows the same sintering temperature conditions as those applied to cylindrical green bodies in a lab-scale study. No cracks were observed in any of the sintered bodies. Table IV presents the results of sintered body fabrication for each forming mold using substitute waste, showing that the volume reduction was approximately 27–33% compared to the raw substitute waste material. For forming mold sizes ID30 and ID50, the mass reduction and volume reduction showed similar results. For the forming mold size ID100, the mass reduction was similar to that of ID30 and ID50; however, a difference was observed in the volume reduction. This was found to be closely related to the tap density of the raw material rather than merely the effect of size.

Table IV. The Results of sintered body

ID	Sintered Body Morphological Characteristics	Mas Reduction [%]	Volume Reduction [%]
ID 30		21.5	31.67
ID 50		21.22	30.64
ID 100		20.85	26.94

The microstructure and morphology of the surface and interior of the green bodies and sintered bodies were captured and analyzed using an optical microscope at 200× magnification, as shown in Fig. 4. In the case of the green body, the particle surfaces were rough and exhibited an irregular structure, with the powder particles appearing to be agglomerated. As a result, the connections between particles were incomplete, suggesting that the strength would be relatively low. In the case of the sintered body, the surface appeared smoother and denser, suggesting that the particles fused together during the sintering process, significantly reducing the porosity.



<Green Body> <Sintered Body>

Fig. 4. Green body and sintered body microstructure analysis results

SEM and EDS analysis of the sintered body fragments indicated that nitrogen (N) volatilized during sintering, while oxygen (O) increased due to oxidation reactions. Lastly, carbon (C) was presumed to have volatilized in the form of CO or CO_2 during the sintering process.

As shown in Fig. 5, the compressive strength of the sintered bodies was measured using a compression strength testing press. It was confirmed that all sintered bodies exhibited a compressive strength of at least 10 MPa.

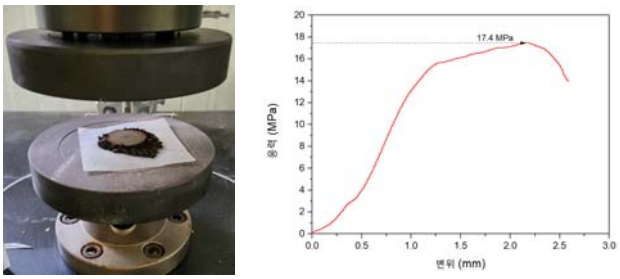


Fig. 5. Compression strength measurement appearance and results

4. Conclusions

In this study, an optimization study on forming and sintering conditions was conducted based on various fundamental experiments and evaluation results. This was done to secure solidification technology by fabricating medium-scale glass composite green bodies and sintered bodies using substitute waste.

First, substitute waste materials were prepared and analyzed for the characterization of green bodies and sintered bodies. The substitute waste materials were manufactured by mixing various additives with an MFC+4A base. The particle distribution and mixing uniformity of the prepared substitute waste materials were analyzed. To ensure that the powder-form substitute waste materials used in the analysis were always utilized under consistent conditions, particle size analysis was conducted. The results confirmed that all materials had similar particle size distributions.

The green bodies were fabricated in three different forming mold sizes (3 cm, 5 cm, and 10 cm). The thickness of all green bodies was controlled to be within 0.75 to 0.8 cm. At this stage, the volume reduction of the green bodies was approximately 20%. Surface imaging using an optical microscope revealed the presence of fine grooves on the surface, which

appeared smooth to the naked eye. The green bodies were sintered by increasing the temperature at a constant rate of 8°C per minute. For sintered bodies of all forming mold sizes, the mass reduction was approximately 20%, while the volume reduction decreased by about 27–31% compared to the raw material. The compressive strength of the sintered bodies was measured, and it was confirmed that all of them exhibited a strength of over 10 MPa. The microstructure of the surface and internal cross-section of the sintered bodies was analyzed using an optical microscope, while SEM and EDS equipment were employed to capture images and confirm the composition of the sintered body fragments. It was confirmed that many of the porosities observed in the green bodies disappeared during the sintering process. Through this study, the optimal composition of the mixture for forming and sintering were determined. The results of this study can serve as important information for the engineering design of unit process technologies such as powder mixing equipment, forming equipment, and sintering equipment at a commercial scale, aimed at producing final glass composite solidified bodies from dispersed waste generated in dismantling and waste treatment processes.

Acknowledgements

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REFERENCES

[1] International Atomic Energy Agency, Strategy and methodology for radioactive waste characterization, IAEA-TECDOC-1537 (2007).

[2] Dong-Sik Jin, Yong-Ho Hong, Hui-Gyeong Kim, Sang-Soo Kwak, Jae-Geun Lee. (2022). Considerations for Designing Solidification Devices of Dispersible Radioactive Waste. 한국방사성폐기물학회 2022 추계학술논문요약집, 261-261.

[3] HyunWook Kang, JaeGeun Lee, YongHo Hong, SnagSu Kwak, DongSik Jin. (2023). The Establishment of Dispersive Radioactive Waste Solidification Devices and Preliminary Performance Evaluation. 한국방사성폐기물학회 2023 춘계학술논문요약집, 270-270.

[4] Mi-Hyun Lee, Young Hwan Hwang, Jung-Kwon Son. (2023). Development of Radioactive Waste Solidification Process Technology and Analysis of Acceptance Criteria for Solids at Domestic and Foreign Disposal Facilities. 한국방사성폐기물학회 2023 춘계학술논문요약집, 267-267.