

Physical and adsorption characteristics of geopolymers prepared using 1–5 M NaOH solution for immobilization of radioactive wastes

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The temporal variations of compressive strength and flowability (slump-flow), and the adsorption behaviors of Co, Sr, and Cs for fly ash-based geopolymers, were investigated for immobilization of radioactive wastes. The geopolymer samples were prepared by using 1–5 M NaOH solution and cured at ambient temperature. The strength and flow were related to the amount and concentration of NaOH solution. The trends were discussed with the amount of heat generated at an early stage of curation. In addition, the geopolymers adsorbed Co equivalent to cements, Sr and Cs more than cements. The trends of absorption were related to the pH of leachates and the magnitude of x-ray diffraction (XRD) peaks of materials which possess larger distribution coefficients. We considered the possibilities to improve performance and to resolve problems in workability for actual use for immobilization of radioactive wastes according to the results of the present work.

1. Introduction

Low-level radioactive wastes such as filters, metals, spent resin, sludge, etc., are continuously discharged from nuclear power plants. The filters and metals contain radioactive isotopes of ^{14}C , ^{60}Co , ^{63}Ni , ^{129}I , etc. The resin and sludge contain ^{90}Sr , ^{137}Cs , etc.¹ In Japan, these wastes are immobilized with cement in metal drums or containers in order to prevent elution of the radioisotopes if rainwater and groundwater reach the waste. The immobilized wastes are eventually buried for disposal. In the immobilization, the void space in the container is filled with cement for large-volume wastes such as filters, metals, etc. Several drums with a volume of 250 L each in which the wastes are immobilized are accumulated for storing. The immobilized wastes should have a compressive strength enough to stand for the load, although the standard for the strength has not been instituted. Wastes in the form of sludge, spent resin, etc., are uniformly immobilized with cement. A compressive strength higher than 1.47 MPa is required for the immobilized sludge and resin waste. Low elution rates for the radionuclides are essential for the immobilization of these radioactive wastes. Flowability of the filler is an important parameter for filling void space in the container and considering detailed working process. The immobilized wastes which possess lower elution rates for the relevant radioisotopes, i.e. higher distribution coefficients, K_d , are preferred to minimize pollution when water reaches the wastes during long term storage. The standards for the flowability and K_d have not been instituted.

Apart from immobilization using cement, attention has recently focused on geopolymers for the immobilization material of the radioactive wastes instead of cement because of its lower leaching rates for Cs and Sr than cement². Geopolymers are inorganic polymers formed by polymerization of aluminosilicates by activation using an alkaline solution such as

NaOH^{3–5}. The polymerization occurs by a chemical reaction between Si-O-Al-O polymer chains and Al-Si oxides. Main structure has been reported by Davidovits⁶, etc. The structure and detailed chemical process depend on a Si/Al ratio^{7–10}. Numerous studies on geopolymers have been conducted worldwide for the past 100 years to understand their characteristics and utilize them for building materials.

Wang et al.¹¹ and Somna et al.¹² investigated the compressive strength of metakaolin-based and fly ash-based geopolymer cured at ambient temperature. They reported compressive strengths for geopolymers prepared using NaOH solutions with concentrations higher than 4 M. Strength was greater as the concentration increased and showed a maximum at 10–14 M. The strengths were related to curing temperature and Si/Al ratio. The strength increased with curing temperature and reached a maximum at 60–80 °C. The strength then decreased as the temperature rose^{13–15}. Duxson et al.¹⁶ and Lee et al.¹⁷ reported that geopolymer strength varied by Si/Al ratio. The strength increased with increasing Si/Al ratio, showed a maximum at Si/Al = 2.0–3.5, then quickly decreased above 4.0. Generally, higher curing temperature and alkaline solutions with higher concentrations are required to improve the compressive strength of the geopolymer.

An NaOH solution with a concentration higher than 1.25 M is classified as a deleterious substance under Japan's Poisonous and Deleterious Substances Control Law. Special heaters are required for preparation of general geopolymers to cure at high temperatures. Therefore, the workability at sites is reduced by the use of alkaline solutions with higher concentrations and curing at higher temperatures. There are few reports on the physical characteristics of geopolymers prepared using alkaline solutions with low concentration and cured at ambient temperature. The K_d values for the relevant nuclides on geopolymers have been poorly understood in terms of properties of the ingredients. We focused on a fly ash-based geopolymer which comprises industrial wastes excepting the alkaline solu-

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tion. Utilization of lower concentration of NaOH solution provides lower cost for preparation of geopolymer. Blast-furnace slag was also used with the fly ash to ensure a certain strength¹⁸.

The compressive strength, flowability, and adsorption performance related with the ingredient are important for the immobilization of radioactive wastes in terms of long-time storage. In the present work, we showed the compressive strength and slump-flow, which is one of the indicators for flowability, as well as the K_d values of Co, Sr, and Cs, which are representative isotopes in the wastes, on the geopolymers prepared using 1–5 M NaOH solution which have not been reported. These trends were related with the amount of heat generation during curation, pH of leachates, and products observed by an x-ray diffraction (XRD), to understand the phenomena more deeply. Furthermore, we considered the possibilities to improve performance and resolve problems for actual use of these geopolymers according to the results in the present work.

2. Experimental

2.1. Materials

The coal fly ash which fulfills a standard of JIS A 6201 was used for the fly ash (hereafter FA). CERAMENT A, the granulated blast-furnace slag which fulfills a standard of JIS A 6206, purchased from DC Co., Ltd. was used for the blast-furnace slag (hereafter BFS). The slag contained gypsum. The commercial NaOH solutions for the volumetric analysis were used for 1–5 M NaOH solution.

2.2. Physical characteristics

2.2.1. Geopolymer preparation.

Known amounts of FA, BFS, silicate sand, and 1–5 M NaOH solution were mixed for 2 minutes. The composition of each sample is shown in Table 1. The chemical composition of major elements, Co, Sr, and Cs in each ingredient are shown in Table 2 (see section 2.3.4 for analysis method). The slump-flow was measured at a certain time after mixing according to the procedure described in section 2.2.2. Eventually, the samples were transferred into plastic molds of 50 mm in diameter and 100 mm in length. The samples were cured at 20 °C and their compressive strength was measured as described in section 2.2.3.

2.2.2. Slump-flow test. A slump-flow test was carried out at 0 h, 1 h, and 2 h after mixing based on ISO 9597:2008¹⁹ and ISO 679:2009²⁰. Briefly, samples transferred on a stage using a flow cone (upper diameter 70 mm, lower diameter 100 mm, height 60 mm) slumped 15 times from a height of 10 mm using a flow testing machine (MARUTO). Shorter and longer diameters of the samples spread on the stage were measured.

2.2.3. Compressive strength measurement. Compressive strengths were measured using a compression testing machine (UH-500 kN XR, SHIMADZU) at 7 d, 28 d, 91 d, and 1 year after curing, based on ISO 679:2009²⁰.

2.3. Chemical characteristics

2.3.1. Geopolymer and cement paste preparation.

Silicate sand was not mixed into the geopolymers for measurements of chemical characteristics to eliminate its effect. Ingredients of FA, BFS, and NaOH solution with compositions shown in Table 1 were mixed for 2 minutes. Then, the samples were transferred into a plastic container followed by curing at 20 °C. Cement pastes of ordinary Portland cement (OPC) and blast-furnace slag cement type B (BB) were prepared with the compositions shown in Table 1 using the same method used with the geopolymers. Analyses of K_d , pH, and XRD described

TABLE 1: Weight Ratios Used for Preparation of the Geopolymer and Cement Samples. The Compositions for Measurements of the Physical Characteristics are Outside Parentheses, and Those of the Chemical Characteristics are in Parentheses. The Sample Names Indicate NaOH Concentration or Species of Cement, and Weight Percentage of the NaOH Solution or Water to Sum of FA and BFS.

Sample name	FA	BFS	NaOH	Silicate sand
1M-30%	0.7 (0.7)	0.3 (0.3)	0.3 (0.3)	3.0 (0)
1M-35%	0.7 (0.7)	0.3 (0.3)	0.35 (0.35)	3.0 (0)
1M-40%	0.7 (0.7)	0.3 (0.3)	0.4 (0.4)	3.0 (0)
1M-50%	0.7 (0.7)	0.3 (0.3)	0.5 (0.5)	3.0 (0)
2M-50%	0.7 (0.7)	0.3 (0.3)	0.5 (0.5)	3.0 (0)
3M-50%	0.7 (0.7)	0.3 (0.3)	0.5 (0.5)	3.0 (0)
4M-50%	0.7 (0.7)	0.3 (0.3)	0.5 (0.5)	3.0 (0)
5M-50%	0.7 (0.7)	0.3 (0.3)	0.5 (0.5)	3.0 (0)
Sample Name	Cement	Water	Silicate sand	
OPC-50%	(1.0)	(0.5)	(0)	
BB-50%	(1.0)	(0.5)	(0)	

TABLE 2: Elemental Compositions of the Major Elements, Co, Sr, and Cs in the Ingredients

Ingredients	Ca / %	Si / %	Al / %	Fe / %	S / %	Mg / %	K / %	Ti / %	Co / ppm	Sr / ppm	Cs / ppm
FA	6.0	26.0	10.5	5.8	0.3	0.5	1.7	1.1	54.5	589	0.969
BFS	39.9	11.6	5.1	0.4	1.9	1.6	0.3	0.4	0.164	311	0.176
OPC	52.5	6.7	1.9	2.8	1.2	0.3	0.3	0.2	15.8	313	0.649
BB	46.9	9.2	3.0	1.7	1.4	0.9	0.3	0.4	9.94	344	0.317

below were carried out 3 months after the curing started.

2.3.2. K_d measurement. The K_d values were analyzed based on the manner described by Toyoshima et al.²¹. Two grams of the samples pulverized to less than 1 mm were mixed with 20 mL of water including approximately 1 kBq each of ⁶⁰Co, ⁸⁵Sr and ¹³⁷Cs activities. The samples were shaken for 6 h on the basis of JTL-13 for the leaching test in Japan²². The experiment was carried out at ambient temperature of approximately 20 °C. Known amounts of the filtrates, obtained by centrifugation and filtration using a membrane filter with 0.22 μm pore size, were analyzed using a high purity germanium detector (GEM, ORTEC). The amount of each radionuclide was determined by subtracting the amount of the radionuclide in the solution after the treatment from the amount before the treatment. The K_d values were calculated using Eq. (1).

$$K_d = \frac{\frac{C_{\text{before}}}{M_{\text{before}}M_{\text{solid}}} - \frac{C_{\text{after}}}{M_{\text{after}}M_{\text{solid}}}}{\frac{C_{\text{after}}}{M_{\text{after}}V_{\text{liquid}}}} \quad (1)$$

where C_{before} and C_{after} are count rates of gamma ray peaks for each radioisotope in the solution before and after the treatment. M_{before} and M_{after} are weights of the measured solutions before and after treatment, respectively. V_{liquid} is the volume of the aqueous phase (20 mL in the present work), and M_{solid} is the weight of the solid phase (2 g) used in the experiment.

2.3.3. pH measurement. Four grams of each pulverized sample was stirred with 40 mL of deionized water for 6 h. The pH values of the leachates were analyzed using a pH meter (D-72, HORIBA).

2.3.4. Chemical composition and XRD measurements. Approximately 0.2 g of each ingredient was digested with $\text{HNO}_3 + \text{HF} + \text{HClO}_4$ using a microwave oven. Contents of Co, Sr and Cs in these solutions were analyzed using an inductively coupled plasma mass spectrometry (ICP-MS) (iCAP Q, Thermo SCIENTIFIC). In addition, major elements that exceed approximately 0.1% were analyzed using an X-ray fluorescence (XRF) spectrometry (JSX-1000S, JEOL). The XRD spectra (SmartLab, RIGAKU) of the pulverized geopolymer and cement samples were analyzed as well.

2.3.5. Heat generation measurement. The amounts of heat generation during curation at 20 °C from 20 g of 1M-50% through 5M-50% in Table 1 were recorded every 30 minutes for 1 month using a 6-point type conduction calorimeter (TOKYO RIKO).

3. Results and discussion

3.1. Physical characteristics affected by NaOH concentration and composition. Fig. 1 shows the temporal variations of the slump-flows for the samples prepared using different amounts and different concentrations of NaOH solution. The flows of 1M-30% and 5M-50% at 2 h could not be measured because of the rapid solidification. The flow at each time increased with increase of the amount of NaOH solution, among the geopolymers prepared using 1 M NaOH solution (Fig. 1(a)). The flow at each time decreased with increase of the NaOH concentration comparing among the geopolymers which 50% weight of the NaOH solution to FA and BFS was added (Fig. 1(b)). The temporal variations of the compressive strengths are shown in Fig. 2. The strength decreases with increase of the amount of NaOH solution (Fig 2(a)). The strengths of geopolymers prepared using 2–5 M NaOH solution were higher than those using 1 M NaOH solution, but comparable to each other at each curing time (Fig. 2(b)).

Fig. 3 shows the temporal variations of heat generated during curing in the geopolymer samples prepared using 1–5 M NaOH solutions. The cumulative heat showed steep increases at an early stage, and the slopes became gradually gentler with time. The heat flows, which the cumulative heat is differentiated with time, formed peaks earlier in the samples prepared using higher-concentration NaOH solutions than in those with lower concentration. After peaking, the heat flows decreased with time. Generally, the cumulative heat and heat flow are related to the amount of reaction product and its reaction rate, respectively. Comparing the flow of 1M-50% with that of 2M-50%, approximately 5% difference was observed at 0 h and 1 h, but the difference increased to 20% at 2 h (Fig. 1(b)). Ten percent difference was observed in the heat flows of the two geopolymers until 1.5 h, but the difference gradually became larger after 1.5 h. These heat flows indicated that the chemical reaction between the ingredients in 2M-50% was more active than that in 1M-50% after 1.5 h. The flows of the samples prepared using 3–5 M NaOH solution decreased more steeply than those using the 1–2 M solutions because of more active chemical reaction, as seen in the heat flows.

The cumulative heat was comparable between 20 h and 200 h among the samples prepared using 2–5 M NaOH solutions as well as the strengths as shown in Fig. 2(b) and Fig. 3(a). The sample prepared using 1 M NaOH solution showed the lowest cumulative heat at the time, as well as the lowest strength. The cumulative heat indicates qualitatively that the chemical reac-

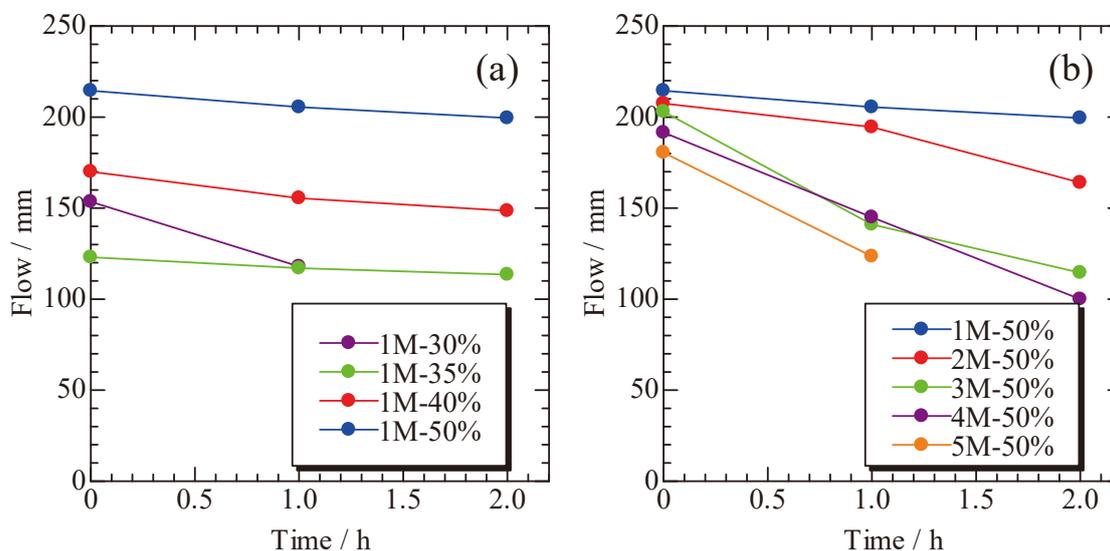


Figure 1. Temporal changes of the slump-flows for samples prepared using various amount of 1 M NaOH solution (a) and using constant amount of 1–5 M NaOH solution (b).

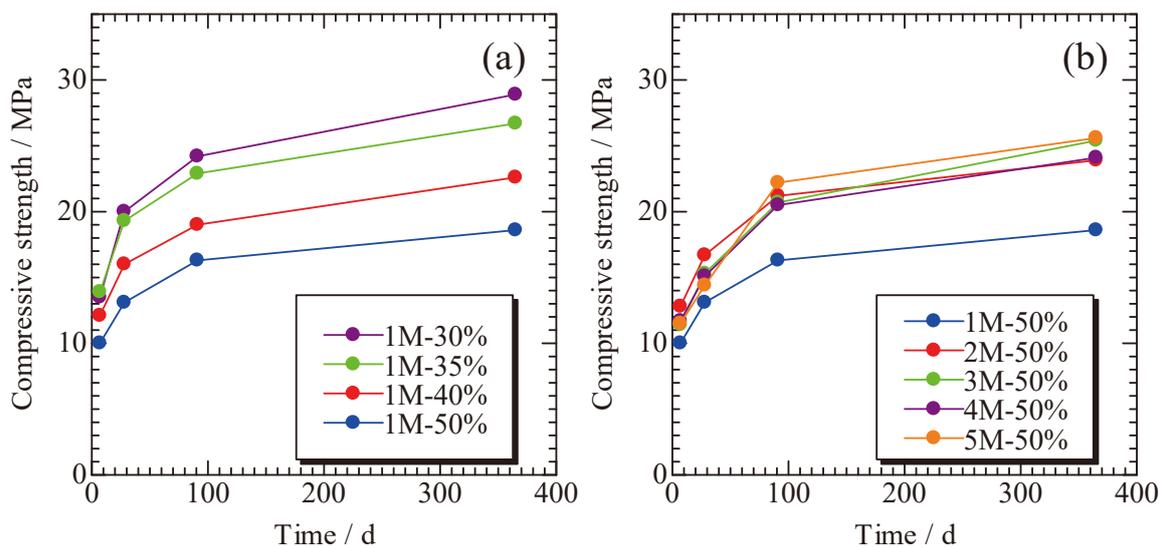


Figure 2. Temporal changes of the compressive strengths for samples prepared using various amount of 1 M NaOH solution (a) and using constant amount of 1–5 M NaOH solution (b).

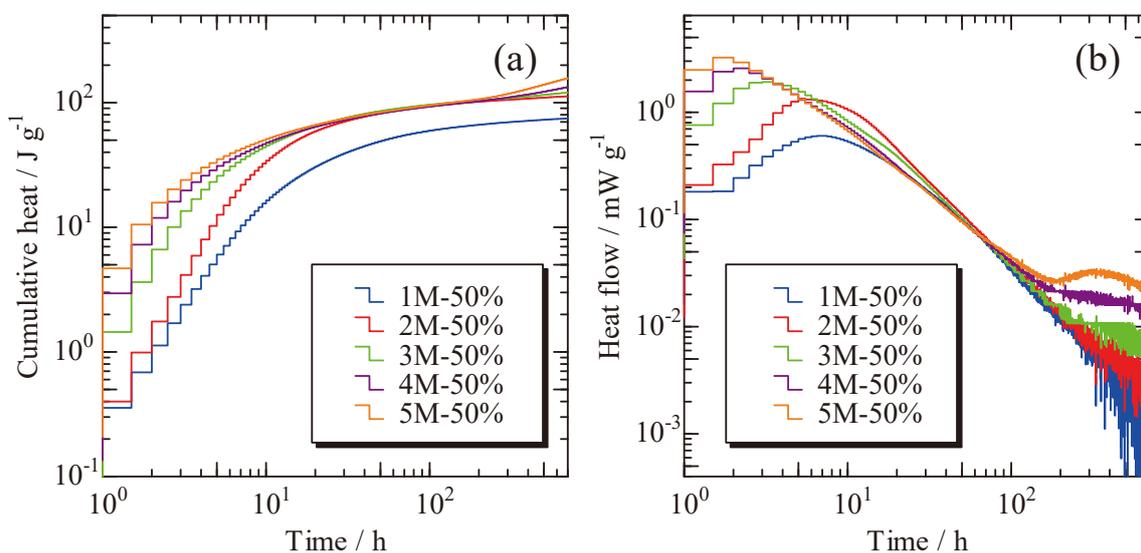


Figure 3. Temporal variations of cumulative heats (a) and heat flows (b).

tion that occurs at 20–200 h dominates the strength. We should note that the strength is dominated by not only the chemical reaction observed as the cumulative heat, but also by the porosity, etc. in the sample.

The cumulative heat increased slightly after around 200 h in the samples prepared using higher-concentration NaOH solutions. The heat flow showed a more prominent peak in the sample with higher alkaline concentrations after 200 h, and then the heat flows decreased with time. Therefore, the cumulative heat reached almost saturation after 1 month (= 700 h). The heat generated at 1 month is not related to the strength because a significant difference was not observed in the trend of their strengths at 7–30 d.

The Si/Al ratio of the geopolymers prepared in this work is estimated to be 2.4 on the basis of the elemental compositions of FA and BFS (Table 2). The Si/Al ratios in general FA and BFS vary between approximately 2 and 3, including the materials used in the present work^{23–27}. The ratio varies depending on the region and factory, etc. where the materials are manufactured. The strength should vary by the ratio^{17,18}. However, the present work shows standard results because the ingredients possess standard chemical composition.

3.2. Adsorption performance characterized by NaOH

concentration and composition. The K_d of Co, Sr, and Cs for each sample are shown in Fig. 4 together with pH of leachate. The K_d values of Co were on the order of 10^3 in all samples including cements. Generally, Co forms precipitate of hydroxide in alkaline solution, including pH range in the present work, because of its low solubility²⁸. Larger K_d results from adsorption of the hydroxide of Co on bodies of geopolymer and cement rather than from a structural mechanism.

The K_d values of Sr and Cs were comparable to each other among the samples prepared using different amounts of 1 M NaOH solution. The amount of the NaOH solution was not related to the K_d . Both cement samples of OPC-50% and BB-50% showed 1 order lower K_d than the geopolymers using 1 M NaOH solution. Comparing samples of 1M-50% through 5M-50%, the K_d for both elements increased with the concentration of NaOH solution. The K_d of the geopolymers increased as NaOH concentration increased. The solubility of CsOH and the solubility product $\text{Sr}(\text{OH})_2$ in water are known to be 395 g/100 mL at 15 °C and $3.2 \times 10^{-4} \text{ mol}^3/\text{L}^3$ at 25 °C, respectively^{29,30}. Approximately 2.2 g of Sr^{2+} can be dissolved in 1 liter at pH = 13.05, equal to OPC-50%. More $\text{Sr}(\text{OH})_2$ can be dissolved in a solution with lower pH than that. The samples of 1M-30% contained 390 ppm of Sr and 0.56 ppm of Cs, based

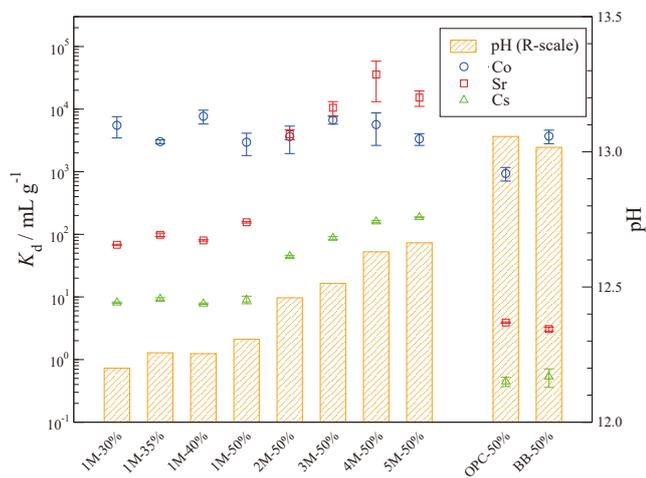


Figure 4. The pH of leachates (right (R) scale) and K_d values (left scale) of Co, Sr, and Cs for the geopolymer and cement samples.

on the chemical composition of the ingredients shown in Table 2. The concentrations in the cements and the other geopolymers were lower than 1M-30%. Even if all of the Sr and Cs in the samples is dissolved during the K_d measurement, the Sr and Cs concentrations are lower than the solubility. The pH does not contribute to the K_d variations for Sr and Cs. Trends of the K_d for Sr and Cs are discussed below with products observed by the XRD.

Fig. 5 shows the XRD spectra of the geopolymer and cement samples. Most of the peaks observed in OPC and BB were assigned to be ettringite and portlandite, which are typical products of cement. The spectra show that less reactive quartz and mullite presented in the FA remained in all the geopolymer samples. Ettringite was detected in the samples prepared using 1 M NaOH solution. The peaks decrease with increase of the amount of the NaOH solution. On the other hand, peaks of chabazite and p-type zeolite were greater with increase of the concentration. The chabazite has been observed in geopolymers investigated by other several groups as well^{31–34}. Naghizadeh et al.³³ investigated the products in the geopolymers prepared using 1–3 M NaOH solution and cured at 80 °C. The chabazite did not form in the samples using the 1 M solution but did form using the 3 M solution. Suksiripattanapong et al.³⁴ reported the temporal variation of XRD spectra in the geopolymers cured at different temperatures. Comparing the geopolymers cured at 65–85 °C, a larger peak of chabazite was observed in the sample cured at higher temperature at an early stage of curation. Takeda et al.³⁵ investigated the formation of p-type zeolite in the mixture of FA and 2–5 M NaOH solutions. The XRD peaks of p-type zeolite at a certain time increased with increasing NaOH concentrations, but the peaks were not observed in the sample treated with the 5 M NaOH solution. The magnitude of the XRD peaks increased with time and reached a constant after a few days.

The crystal structures of chabazite and p-type zeolite are reported by Yakubovich et al.³⁶ and Chen et al.³⁷. Yang et al.³⁸ investigated the K_d of Sr and Cs on the chabazite in the solutions containing K, Cs, Ca, and Sr with different ratios. When K and Ca did not present in the solution, the K_d was on the order of 10^6 for Cs and 10^5 for Sr. The K_d of both Sr and Cs decreased with increasing concentrations of K and Ca because of interference. In addition, Mimura and Akiba³⁹ investigated the K_d on p-type zeolite varied by Na concentration and pH in solution. The K_d of Sr and Cs showed approximately 10^4 in solution with Na concentration lower than 10^{-3} M, but decreased with increasing Na concentrations in the solutions.

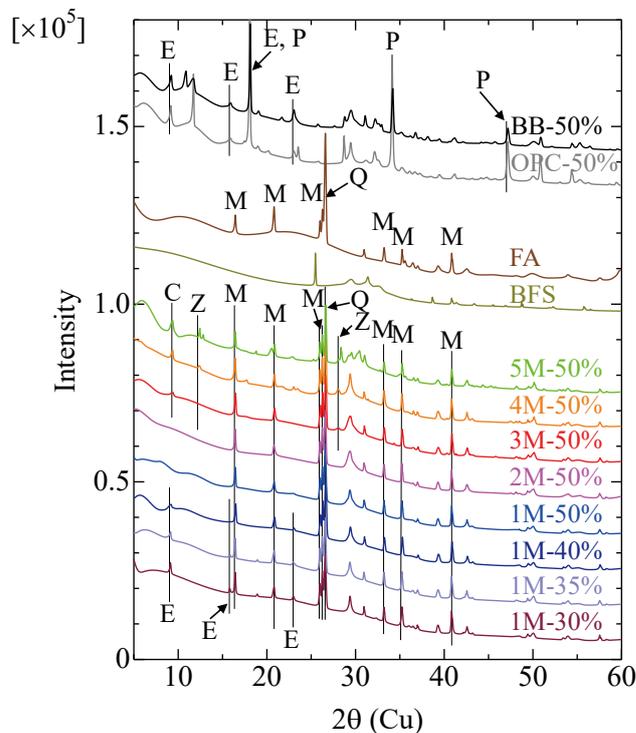


Figure 5. XRD spectra of the geopolymers, ingredients, and cements. C: chabazite, E: ettringite, M: mullite, P: portlandite, Q: quartz, Z: p-type zeolite.

The K_d for both elements were larger in an alkaline solution than in an acid solution. The chabazite and p-type zeolite are related to the adsorption of Sr and Cs on the geopolymer. The poor adsorption on the cements results from less formation of these products, as seen in the XRD spectra. In the present work, Sr showed the K_d 1–2 orders of magnitude greater than Cs in the geopolymer samples, in contrast to these investigations. The magnitude of K_d is affected by the concentrations of elements eluted from the geopolymers that interfere with the adsorption of Sr and Cs.

3.3. Possibilities of further improvement for actual use.

A geopolymer with a larger flow is preferable because void spaces between wastes can be easily filled due to higher flowability, when used for the immobilization of large-volume radioactive wastes as a filler. The geopolymer prepared using more volume of 1 M NaOH solution resulted in improvement for the flow but degradation for the strength. The same trend should be observed in the geopolymers prepared using 2–5 M NaOH solutions according to basic concept in the concrete⁴⁰. A mortar prepared with a weight ratio of cement : fine aggregate : water for 10 : 30 : 5, which is the same composition as the present work, showed a compressive strength of 22 MPa at an age of 28 days⁴⁰. The strengths of geopolymers in the present work are lower than this mortar by more than 30%. The mortars prepared using more water resulted in degradation for the strength as well as the geopolymers. Wastes which sludge and resin, etc. are immobilized with cement showed the strengths equal or less than the cement mortars⁴¹. The strength would be degraded for the geopolymers which these wastes are immobilized as well as the cementation.

On the other hand, the geopolymers prepared using NaOH solution with a higher concentration would be suitable for immobilizing radioactive wastes to suppress leaching. However, the geopolymer prepared using 1 M NaOH solution, which is not classified as a deleterious substance, provided better adsorption than cement. For actual use as a filler, fine aggregates are mixed to increase the volume of a geopolymer at low cost. The K_d of aggregates is reported to be $2.0\text{--}1.0 \times 10^5$

(mean: 640) for Co, 0.4–2400 (mean: 22) for Sr, and $9.6\text{--}3.5\times 10^4$ (mean: 530) for Cs⁴². In particular, the adsorption of Cs can be expected to be improved by mixing in fine aggregates. Co and Sr would adsorb to the geopolymer part rather than the aggregates based on the K_d .

We reported the physical and adsorption properties of the geopolymer prepared using NaOH solutions with different concentrations in the present paper. Use of lower concentrations and less amounts of NaOH solution enables to decrease the cost for the geopolymers. Adsorption performance for Cs would be greater than the data in the present work for actual use as fine aggregates are mixed in the geopolymers. Flowability and strength are affected by the amount of NaOH solution. In contrast, adsorption performance is not varied regardless of the amount of the solution. Concrete admixtures are frequently used to improve flowability and delay time to set. All the commercial admixtures are not necessarily effective with geopolymers. If suitable admixtures are mixed in the geopolymer, the flowability of the one prepared using higher concentration of NaOH solution would be improved⁴³. In addition, a certain flowability would be maintained even though the amount of NaOH solution is reduced. As mentioned above, the strength can be improved by using less amount of NaOH solution. Both strength and adsorption of the geopolymers using 1 M NaOH may surpass those of the cements by further investigation. The present work will serve as basic data for further improvement in workability and strength while maintaining a certain adsorption performance depending on the concentration of NaOH solution.

4. Conclusion

The temporal variations of the compressive strengths and slump-flows, and distribution coefficients for fly ash-based geopolymers were investigated for possible use as filler for the immobilization of large-volume radioactive wastes. All the geopolymers prepared using different amounts of 1 M NaOH solution achieved strength of approximately 20 MPa at 1 year. The geopolymers using more NaOH solution showed greater flowability. When 2–5 M NaOH solutions were used, the flows decreased as the concentration of NaOH solution increased, but the strengths were comparable with each other. The strength and temporal change of the flow were explained by the amount of heat generated during an early stage of curation. On the other hand, distribution coefficients, K_d , of Sr and Cs, one of the indicators to evaluate the adsorption behaviors of these elements, improved with increase in concentrations of NaOH solution. Geopolymers showed greater distribution coefficients than cement by a factor of more than 10. Co showed comparable K_d regardless of the concentration and composition of the ingredients. The trends of Sr and Cs were related to the amount of p-type zeolite and chabazite observed in XRD spectra. Adsorption performance for Cs would be greater than data in the present work for actual use as fine aggregates are mixed. The present work will serve as basic data to improve the strength and flowability using concrete admixtures. We need further investigation to immobilize radioactive wastes by the geopolymer prepared using lower concentration and less amount of NaOH solution.

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