

The Use of Pozzolanic Material for Improving Quality of Strontium Liquid Waste Cementation in Saline Environment during Nuclear Waste Immobilization Process

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Abstract-This research was conducted to study the use of artificial pozzolanic material (portlandpozzolana cement or PPC) and natural pozzolanic material (fly ash) to improve quality of strontium liquid waste cementation in saline environment during nuclear waste immobilization process. The portland cement was mixed with 10 v/0 strontium liquid waste (water containing 65 ppm Sr(II)), 0.3 water-cement ratios (w/c), 20 v/0 fly ash, -40 mesh to -60 mesh sand. Similar process was performed using commercial portlandpozzolana cement (PPC). Compressive strength test after cured the block cements for 28 days was performed using pressing machine. To study leaching rate test, 10 ml contaminated saline water was sampled from immersion of cement block with 1 liter saline water for 21 days. Hitachi Zeeman 8000 Atomic Absorption Spectrophotometer was used for analyzing leaching rate test. All cement blocks showed the results above than 2.5 N/mm² of IAEA standard during strength test. The fly ash showed 103-105% higher strength test than commercial PPC in all grain size. But the fly ash showed 4-5% less durable during leaching rate than commercial PPC during 21 days immersion. All the cement blocks showed the results above than IAEA standard during leaching rate test. The addition of natural pozzolanic material (fly ash) could improve the strength of strontium liquid waste cementation. In other hand, the artificial pozzolanic material could improve the durability from leaching rate of strontium liquid waste cementation..

Keywords-Cementation, fly ash, leaching rate, Portland pozzolana cement, pozzolanic material, strength test.

I. INTRODUCTION

Radioactive waste generally was sorted based on the physical form and level of activity. In Chernobyl and Fukushima accidents, large amounts of volatile radionuclides were released into the environment, one of them was strontium-90 [1,2]. Strontium-90 was one of the major of radionuclide in spent nuclear fuel (SNF), high-level radioactive

wastes (HLW) resulting from SNF reprocessing and radioactive wastes associated with the operation of reactors and fuel reprocessing plants [3]. It is considered among the potentially most hazardous of the products of nuclear fission because of its moderately long half-life (29.1 years) and its relatively high yield (6%) in the fission process.

Besides, it is also highly water-soluble [2].

Therefore, immobilization of the strontium liquid waste is important because it protects the waste from migration and dispersion to the environment. Cementation is one of the immobilization process that has met acceptance in many countries and has been considered, because of its low cost, high density, durability and amenability to simple processing techniques [4]. Some requirements are needed for concrete resulted from the cementation process based on IAEA [4]: density: 1.70~2.50 g/cm³, compressive strength: 20~50 N/mm², compressive strength after waste loading: 2.5 N/mm², leaching rate (Rn): 1.70 x 10⁻¹~2.50 x 10⁻⁴ g/cm².days, dose rate in contact surface: 2 mSv/hr, dose rate at 1 meter from contact surface: < 0.10 mSv/hr, dose rate outside of interim storage: 0.005 mSv/hr.

In addition, the Russian Federation standard requires higher compressive strength than IAEA standard (5 N/mm²) [3].

Cement could be for substituting Portland cement was the Pozzolan cement (PC). Usually the presence of pozzolan on the Portland cement will give low initial compressive strength. The power will eventually exceed the concrete compressive strength of Portland cement type 1 as Portland pozzolanic cement (PPC) [6]. Besides of PPC, many other materials had potential pozzolanic activities, one of them was fly ash. Generally, fly ash produced from combustion of pulverized coal. Fly ash was blended with cement had an appropriate granulometry, were free from deleterious impurities and sufficiently reactive with cement to reach minimum standards of strength gain. However, fly ashes met these criteria embraced a wide range of compositions and crystallinity and, at long ages, after reaction, may confer a wide range of properties on the resulting cement matrix, especially when used at high replacement levels [7].

This research was conducted to study the use of natural pozzolanic material (fly ash) and artificial pozzolanic material (portlandpozzolan cement or PPC) to improve quality of strontium liquid waste cementation in saline environment during nuclear waste immobilization process.

II. METHODS

Natural pozzolanic material in 20v/0 fly ash was mixed with portland cement and artificial pozzolanic material was performed using portlandpozzolana cement (PPC). Each cement was mixed with 10 v/0 strontium liquid waste (water containing 65 ppm Sr(II)), 0.3 water-cement ratios (w/c), sand with grain size of -40 mesh, -50 mesh and -60 mesh. And all cement blocks were cured for 28 days. Compressive strength after cured the cement blocks for 28 days was performed using pressing machine which had capacity up to 200 kN.

To study leaching rate test, cement blocks were immersed in saline water from mixing 1 liter of distilled water with 35 grams of NaCl (represented as simulation sea water with 3.5 % salinity [8]). 10 ml contaminated water was sampled from the immersion at day 2,3,4,5,6,12,18,21. Hitachi Zeeman 8000 Atomic Absorption Spectrophotometer was used for analyzing the strontium contents in contaminated water during leaching rate test. The leaching rate of strontium was calculated using Eq. 1.

$$R = \frac{A_t}{A_0} \times \frac{W_0}{S \times t} \left(\frac{\text{gram}}{\text{cm}^2 \cdot \text{day}} \right) \quad (1)$$

where R was leaching rate (g.cm⁻².day⁻¹), A_t was leached activity at t time (Ci), A_0 was initial activity (Ci), W_0 was mass of cement block (gram), S was surface are of cement block (cm²), and t was leaching time.

III. RESULTS AND DISCUSSIONS

The result of compressive strength test from different type of pozzolanic materials was shown in Fig. 1.

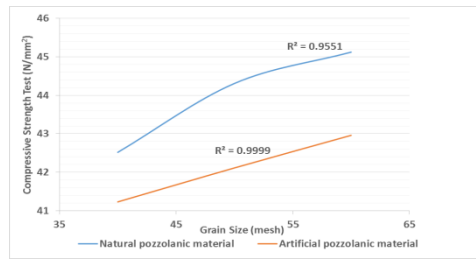
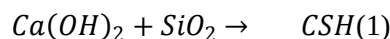


Fig. 17 Compressive strength test between natural pozzolanic material and artificial pozzolanic material

All the results showed above than 2.5 N/mm² of IAEA standard during test. Linearity showed during increasing of grain size. Because the grain size was played role for cement matrix workability and quality of hardening itself. When the grain size became smaller, the sand became rougher and made workability became harder that could decrease the compressive strength of cement block. By increasing the grain size, could increase the compressive strength of cement block. Because increasing the grain size will enlarge the interaction area of sand and cement adhesive. Cement adhesive will fill the airspaces uniformly and bonded the grains perfectly. Thus will increase the compressive strength of cement block [9].

The addition of pozzolanic material through cement will increase bonding time and decrease the strength of cement matrix during early process as follow reaction:



The reaction of SiO₂ from pozzolanic material with Ca(OH)₂ formed CSH (tobermorite) for hardening cement matrix. But in the end, after became cement block, the strength of cement block will increase significantly and higher than normal cement block [10,11,12]. No difference significant (3-5%) was shown between adding natural pozzolanic material and artificial pozzolanic material. Although natural pozzolanic material contained more than artificial pozzolanic material that be

shown in table 1, the duration for hardening required 90 days to be completely homogeneous [11]. So during 28 days, the compressive strength test obtained similar result between each other. Although incompletely hardening, the results still higher than standard IAEA.

TABLE IV
MINERAL COMPOUNDS IN DIFFERENCE
POZZOLANIC MATERIALS

Mineral Compounds	Content (%)	
	Natural pozzolanic material	Artificial pozzolanic material
CaO	2.23	58.35
SiO ₂	56.96	24.60
Al ₂ O ₃	17.27	9.45
Fe ₂ O ₃	12.37	4.40
Na ₂ O	4.05	1.05
SO ₃	1.05	2.15

Leaching rate test was conducted to evaluate the safety of liquid waste cementation in saline environment during nuclear waste immobilization process. Natural pozzolanic material showed higher than artificial pozzolanic material in Fig 2.

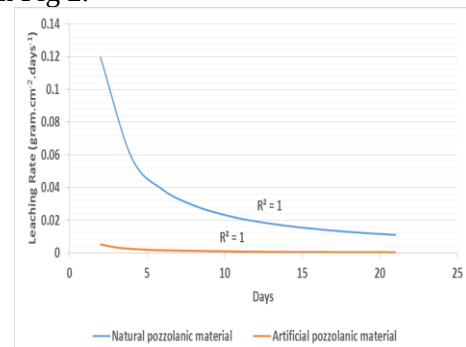
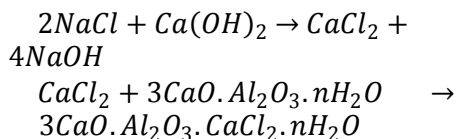


Fig. 2 Leaching test rate between natural pozzolanic material and artificial pozzolanic material

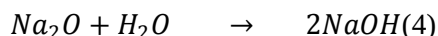
There are two reasons behind this. First, in saline environment, mineral compound will be more aggressive and reactive to cement matrix compounds. So it will trigger the reaction between both and create corrosion for long time.

Corrosion due to penetration of sodium chloride through the airspace of cement matrix followed reaction [13]:



Then $CaCl_2$ reacted with $3CaO \cdot Al_2O_3 \cdot nH_2O$ and obtained $3CaO \cdot Al_2O_3 \cdot CaCl_2 \cdot nH_2O$ (Chloro aluminate). Chloro aluminate will expand cement block until exceed the origin form. The expansion will create crack then create porous through inside of cement block. Accumulate porous will make the cement block became fragile. When it was fragile, the strontium will easily release and mix with the saline water.

Second, the content of Na_2O in difference pozzolanic materials as described in Table 1 above. Na_2O reacted with water followed:



Strontium is easily soluble into NaOH and KOH [14]. Therefore, the increasing of Na_2O affected rapid dissolving of strontium through the leachate liquid. Although natural pozzolanic material higher than artificial pozzolanic material, after 21 days, both strontium leaching rates were met the IAEA standard ($10^{-2} \sim 10^{-3}$ g/cm².days).

IV. CONCLUSIONS

All cement blocks showed the results above than 2.5 N/mm² of IAEA standard during strength test. The natural pozzolanic material showed 103-105% higher strength test than artificial pozzolanic material in all grain size. But the natural pozzolanic material showed 4-5% less durable during leaching rate than artificial pozzolanic material during 21 days immersion. All the cement blocks showed the results above than IAEA standard during leaching rate test.

Both pozzolanic materials could be applied as alternative methods for liquid strontium cementation in saline environment.

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