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PRELIMINARY STUDY OF THE UTILIZATION OF THE FLY ASH FROM COAL-FIRED POWER PLANT FOR IMMOBILIZATION OF RADIOACTIVE WASTE

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ABSTRACT

Preliminary study of the utilization of the fly ash from coal-fired power plant for immobilizing simulated radioactive waste has been done. The objective of this research was to study characteristics of pozzolanic material of the fly ash from coal-fired power plant as substitute of compactor material for immobilizing simulated radioactive waste. The experiment was carried out by mixing of the compactor materials such as (cement + lime), (cement + fly ash), (cement + fly ash + lime), (fly ash + lime) with Na_2SO_4 225 g/L and KCl 4.6 g/L as simulation of evaporator concentrate according to reference waste form no. 1 on characterization of low and medium-level radioactive waste forms in the EUR 9423-EN. Each mixture of compactor materials solidified for 14 days, 21 days, and 28 days. Solidified result was monolith, and then its compressive strength, water absorption, and porosity were tested. The experiment result showed that the best of the compactor materials on the immobilizing simulated radioactive waste was cement of 30% (wt), fly ash of 20% (wt), and lime of 20% (wt) with compressive strength of monolith of 1512.7 N/cm^2. The condenser substance on the weight ratio of fly ash/lime of 20/50 – 60/10 % (wt) as pozzolanic substance could be used for immobilizing simulated radioactive waste by compressive strength of monoliths of 345 – 610.4 N/cm^2. Minimum compressive strength of monolith from radioactive waste cementation according to IAEA is 320 N/cm^2, hence compressive strength of monoliths from this experiment can be expressed enough well.

Keywords: *fly ash, coal-fired power plant, immobilization, radioactive waste*

INTRODUCTION

Fly ash is the solid waste originated from the ash that is carried by the exhaust gases from coal combustion used to the coal-fired power plant, the clinker combustion process in a rotary kiln of cement plants, coal gasification, and coal liquefaction. The exhaust gases contain highly soft fly ash particles. The fly ash will be retained on electrostatic settling before run out with flue gas.

Fly ash generated by the coal steam power plant in Indonesia has granulometry analysis about 85% (wt) with size of grains from 0.5 to 50 µm [1]. Association of the fly ash development in Australia has performed a granulometry analysis of the fly ash with the results 86.7% % (wt) with size of grain about 45 µm [2].

According to ASTM C 618-91, Pozzolan divided into several classes: N Class: Pozzolan derived from natural materials such as trass, diatomaceous, clay, kaolin, and bentonite. F&C Class: Artificial Pozzolan or man-made. Included in this type is furnace slag, fly ash from the coal combustion [3-4].

Characteristics of some Pozzolan classes can be seen in Table 1 [3-5]. Based on the type of coal used as fuel, fly ash is divided into two classes (ASTM C 618–94a (in Husin, 1998)), namely [5]:

F Class: fly ash generated from coal combustion the type of anthrasite or bituminous.
C Class: fly ash produced from coal combustion the type of lignite or sub-bituminous.

Cement that can be used as substitutes of Portland cement is 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 [5].

In the Pozzolan cement reactions between Pozzolan, lime and water are described as follows [6]:

$$Ca(OH_2) + SiO_2 + H_2O \rightarrow CaO.SiO_2.2H_2O \quad (1)$$

$$Ca(OH)_2 + Al_2O_3 + 5H_2O \rightarrow CaO.Al_2O_3.6H_2O \quad (2)$$

$$2Ca(OH)_2 + Al_2O_3 + Fe_2O_3 + 5H_2O \rightarrow 2CaO.Al_2O_3.Fe_2O_3.7H_2O \quad (3)$$

$$4Ca(OH)_2 + Al_2O_3 + 27H_2O + 3SO_3 \rightarrow CaO.Al_2O_3.3CaSO_4.31H_2O \quad (4)$$

On the Pozzolan cement, formation of calcium hydroxide heat of hydration was slow that can prevent cracks in concrete.

Minerals derived from rocks and soils that exist in the earth layers contain most of the uranium radionuclide series (^{238}U) and thorium (^{232}Th), and a radioactive isotope of potassium (^{40}K). In uranium series,

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Table 1. Pozzolan classification according to ASTM C 618-91

Description		**N Class**	F Class	C Class
SiO_2	min (%)		54.90	39.90
$SiO_2 + Al_2O_3 + Fe_2O_3$	min (%)	70.0	70.0	50.0
SO_3	max (%)	4.0	5.0	5.0
Water content	max (%)	3.0	3.0	3.0
Incandescent lost	max (%)	10.0	12.0	6.0
Alkali as Na_2O	max (%)	1.5	1.5	1.5
Pozzolan activity with 7 days lime:	min (psi)	800	800	-
	min (kg/cm^2)	56.25	56.25	-

(^{226}Ra) is the most important as disintegration chain segment in radiological, hence radium is chosen instead of ^{238}U [7]. Based on this, the ^{238}U, ^{232}Th, and ^{40}K contained in solid waste fly ash from coal-fired power plant is classified as TENORM (Technologically Enhanced Naturally Occurred Radioactive Materials).

Results of radioactivity concentration measurement in fly ash TENORM that generated from coal-fired power plant in Paiton Probolinggo East Java showed the concentration of natural radioactivity of uranium series (^{238}U) is 170 Bq/kg, thorium series (^{232}Th) is to 87 Bq/kg, and potassium (^{40}K) is 105 Bq/kg [8]. The value of radioactivity concentration in the fly ash is lower than the clearance level provisions required by the IAEA-TECDOC -855 (1996) and Nuclear Energy Agency (2004) that is 300 Bq/kg for single radionuclide ^{238}U, ^{226}Ra and ^{232}Th [9, 10], while the clearance levels for the ^{40}K is 300,000 Bq/kg [11].

From TENORM analysis result primarily for the series of uranium and thorium and also potassium in the fly ash from coal-fired power plant in Paiton, it can be concluded that based on the exemption level of TENORM issued by the International Atomic Energy Agency (IAEA) regulations, the fly ash generated from coal-fired power plant in Indonesia is predicted to provide the radioactivity concentrations in the range which is almost equal to the value of the fly ash radioactivity concentration in Paiton. Thus the fly ash from coal-fired power plant in Indonesia can be classified as TENORM which could be released.

Thus, the fly ash waste from coal-fired power plant in Tanjung Jati B Jepara is possible to be used as the condensed material in the immobilization of radioactive waste. Result of immobilizing radioactive waste was called monolith. Physical characteristics such as compressive strength, porosity and water absorption of the monolith were then evaluated.

This research has been conducted on the influence of fly ash as an ingredient on the immobilization of radioactive waste compactor simulation based on an evaluation of physical characteristics (compressive strength, porosity and water absorption) of the monolith.

EXPERIMENTAL SECTION

Materials

The materials were used in this research include: waste simulation in accordance with reference waste form no.1 in the characterization of low and medium-level radioactive waste forms in the EUR 9423-EN with non-active component consist of Na_2SO_4 225 g/L and 4.6 g KCl/L [12].

Fly ash from electrostatic precipitator (ESP) of coal-fired power plant in Tanjung Jati B Jepara with chemical composition in % weight as follows: 53.626% SiO_2, 21.611% Al_2O_3, 11.159% Fe_2O_3, 5.457% CaO, 0.882% MgO, 2.260% Na_2O, 1.697% K_2O, 3.308% H_2O [13].

Gresik Portland cement type 1 obtained from building material stores in Yogyakarta with the chemical composition in weight % as follows: 65.29% CaO, 21.30% SiO_2, 5.41% Al_2O_3, 3.53% Fe_2O_3, 0.89% MgO, 2.25% SO_3, 1.20% free lime, 0.20% alkali (Na_2O + 0.658 K_2O), and 1.63% incandescent lost [14].

Limestone obtained from the building materials store in Yogyakarta.

Instrumentation

The equipment used are a set of glassware, Sybron furnace, Sartorius analytical balance, sieve Tyler 400 mesh, magnetic stirrer, mixer, cylindrical monolithic printing container covered with diameter 4 cm and 4 cm in tall, cylindrical container covered with a volume > 10 x volume of cylindrical monolith, Paul Weber measuring tools of compressive strength.

Procedure

Preparation of Fly Ash Mineral

Fly ash from coal-fired power plant in Tanjung Jati B Jepara was heated in oven at 110 °C to obtain a constant weight, cooled in excicator, sieved using a Tyler sieve mesh of size 400, and stored in tightly closed containers.

Table 2. Monolith material test composition (composite of the immobilization result of RAW simulation)

No.	Sample Code	Composition, % weight			
		RAW Simulation	Cement	Lime	Fly Ash
1	K1	30	70	-	-
2	K2	30	60	10	-
3	K3	30	50	20	-
4	K4	30	40	30	-
5	K5	30	30	40	-
6	K6	30	20	50	-
7	K7	30	10	60	-
K1 – K7 as control					
8	L1	30	50	10	10
9	L2	30	40	10	20
10	L3	30	30	10	30
11	L4	30	20	10	40
12	L5	30	10	10	50
13	M1	30	40	20	10
14	M2	30	30	20	20
15	M3	30	20	20	30
16	M4	30	10	20	40
17	N1	30	30	30	10
18	N2	30	20	30	20
19	N3	30	10	30	30
20	O1	30	20	40	10
21	O2	30	10	40	20
22	P1	30	10	60	10
23	Q1	30	-	-	70
24	Q2	30	-	10	60
25	Q3	30	-	20	50
26	Q4	30	-	30	40
27	Q5	30	-	40	30
28	Q6	30	-	50	20
29	Q7	30	-	60	10

Preparation of Radioactive Waste (RAW) Simulation

Na_2SO_4 of 225 g and KCl of 4.5 g were inserted into the measurement gourd that filled with demineralized water in 1000 mL, then stirred with a magnetic stirrer until the Na_2SO_4 and KCl was dissolved. The solution in the measurement gourd represents RAW simulation with levels of Na_2SO_4 225 g/L, and KCl 4.5 g/L in accordance with reference waste form no.1 [12].

Preparation of Monolith (Solid Composite of Immobilization Results of RAW Simulation)

Fly ash pass siever of 400 mesh, cement, lime and RAW simulation at a certain ratio were mixed until homogeneous. RAW simulation was solidified with concentrate/cement ratio of 0.37 to 0.52 [12]. The role of cement as a condensed material will be replaced by a condensed material which includes a mixture of materials (cement + lime), blended (cement + lime + fly ash), and Pozzolan mixtures (lime + fly ash). The monolith establishment from immobilization results of RAW simulation was made from a mixture (RAW simulation + compactor material) on the ratio of RAW simulation/compactor material = 30/70 = 0.428 with a composition as presented in Table 2.

The mixtures with compositions such as in the Table 2 was stirred until homogeneous, and then each mixture according to the sample code was entered in 3 pieces mold in cylindrical shape, then was cured for 14 days, 21 days, and 28 days. Monolith in the mold on the day of curing of 14^{th} , 21^{st}, and 28^{th} out from the mold, and then pressing test was done using a measuring tool of compressive strength of Paul Weber.

Test of Water Absorption and Monolith Porosity Determination

Test of water absorption and monolith porosity determination was conducted on monolith with monolith-forming composition that gives the best compressive strength at 28 days of curing monolith. The monolith was then weighed, and inserted into the test container with monolith position adhere horizontally at the bottom of the container. Container was filled with monolith, then filled with demineralized water as much as 10 times the volume of monolith, then was sealed for 7 days in water bath to reduce temperature changes. Monolith was removed from the container after submerged by demineralized water for 7 days, then placed in a pan with horizontal position monolith.

Table 3. Some minerals that are formed in Portland cement type 1 hydration process

Minerals name	Formula name	Composition, % weight
Portlandite	$Ca(OH)_2$	29.2
C-S-H	$Ca_3Si_2O_3(OH)_8$	50.9
Hydrogarnet	$Ca_3Al_2(OH)_{12}$	6.8
Fe oxides	Fe_2O_3	3.0
Monosulfate	$[Ca_2Al(OH)_6.2H_2O]_2SO_4.2H_2O$	1.8
Ettringite	$[Ca_3Al(OH)_6.12H_2O]_2(SO4)_3.2H_2O$	8.1

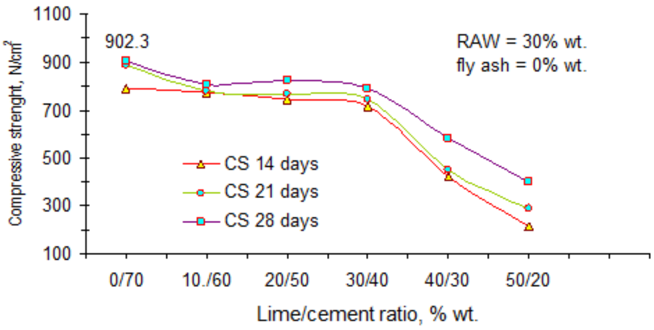

Fig 1. Effect of lime / cement ratio with 0% weight fly ash addition against compressive strength of immobilization results of RAW simulation

Monolith was aerated until the monolith surface exactly dry, and then was determined water absorption (%) and monoliths porosity using the equation:

$$WA = \frac{W_i - W_o}{W_o} \times 100\% \quad (5)$$

when, WA = water absorption (%), W_i = the final weight of monolith after soaking in the demineralized water (g), W_o = initial weight of monolith after heated at temperature 110 °C for 24 h (g).

$$\varepsilon = \frac{\frac{W_i - W_o}{\rho_w}}{V_t} \quad (6)$$

when, ε = monolith porosity, ρ_w = density of demineralized water at a temperature of water bath (g/cm^3), V_t = volume of monolith (cm^3).

RESULT AND DISCUSSION

Analysis of Chemical Composition of Fly Ash as Pozzolanic Terms

Content of SiO_2 in fly ash from coal-fired power plant in Tanjung Jati B Jepara is 53.626% by weight. According to ASTM C 618-91, the fly ash from Tanjung Jati B including pozzolan in C class with minimum content of SiO_2 39.90% weight. Total chemical composition of SiO_2 (53.63%), Al_2O_3 (21.6%), Fe_2O_3 (11.16%) in the fly ash from coal-fired power plant in Tanjung Jati B Jepara is 86.40% by weight. It have fulfilled the conditions of pozzolan according to ASTM C 618-91 with total chemical composition of (SiO_2, Al_2O_3, Fe_2O_3) for fly ash pozzolan of F and C class is 70% and 50% by weight respectively. Components of SiO_2, Al_2O_3, Fe_2O_3 in fly ash is an important element of pozzolan composer which by water and lime will be forming compounds of calcium silicate hydrate ($CaO.SiO_2.2H_2O$), aluminate calcium hydrate ($CaO.Al_2O_3.6H_2O$), and calcium aluminate ferrite hydrate ($2CaO.Al_2O_3.Fe_2O_3.\ 7H_2O$) as an addition to the concrete monolith compiler framework [4-6].

Effect of Lime/Cement Ratio against Immobilization RAW Simulation Results without Pozzolan Materials of Fly Ash

Immobilization process of RAW simulation using cement, lime, and fly ash is the nipping process or molecules immobilization of RAW simulation by tubermorite gel $3CaO.SiO_2.4H_2O$, $3CaO.2SiO_3.4H_2O$, $3CaO.Al_2O_3$. $Ca(OH)_2.12H_2O$, and $6CaO.Al_2O_3.Fe_2O_3.12H_2O$ that will be hard caused by process of hardening tobermorite gel into compact, dense and hard monolith. Immobilization of RAW was intended so that radionuclide molecules can be jam in well in monolith so that it will be difficult to slip off at the time performing of repository. Effect of lime and cement to the monolithic of immobilized RAW simulation results without the addition of fly ash Pozzolan was shown in the Fig. 1.

Fig. 1 shows that the addition of more lime on the immobilization of RAW simulations has been gave compressive strength monolith from results of curing time during 14 days, 21 days and 28 days smaller. This is understandable because the formation of free lime from the cement can not be avoided, because base material of cement itself contains limestone. Free Lime $Ca(OH)_2$ is the air mortar and is the weakest crystal in concrete. The higher the amount of free lime was added to the mixture of concrete monolithic, hence the compressive strength of concrete monoliths was decreased [15].

Number of free lime (Portlandite) in the concrete monolith from the cement hydration process is shown in Table 3 [16]:

The forming of some mineral in Table 3 were came from hydration reaction of the Portland cement type 1 which usually can be expressed with the equation (7) up to (12) or (13) up to (16) as follows [16-17]:

$$\underset{alite}{2Ca_3SiO_5} + 7H_2O \rightarrow \underset{C\text{-}S\text{-}H}{Ca_3Si_2O_3(OH)_8} + \underset{portlandite}{3Ca(OH)_2} \quad (7)$$

$$\underset{belite}{2Ca_2SiO_4} + 5H_2O \rightarrow \underset{C-S-H}{Ca_3Si_2O_3(OH)_8} + \underset{portlandite}{Ca(OH)_2} \quad (8)$$

$$\underset{ferrite}{Ca_4Al_2Fe_2O_{10}} + 7H_2O \rightarrow Fe_2O_3 + \underset{hydrogarnet}{Ca_3Al_2(OH)_{12}} + \underset{portlandite}{Ca(OH)_2} \quad (9)$$

$$CaO + H_2O \rightarrow \underset{portlandite}{Ca(OH)_2} \quad (10)$$

$$\underset{aluminate}{Ca_3Al_2O_6} + \underset{gypsum}{3CaSO_4.2H_2O} + 26H_2O \rightarrow \underset{ettringite}{[Ca_3Al(OH)_6.12H_2O]_2(SO_4)_3.2H_2O} \quad (11)$$

$$\underset{ettringite}{[Ca_3Al(OH)_6.12H_2O]_2(SO4)_3.2H_2O} + \underset{aluminate}{Ca_3Al_2O_6} + 4H_2O \rightarrow \underset{monosulfate}{3[Ca_2Al(OH)_6.2H_2O]_2SO_4.2H_2O} \quad (12)$$

The other form of hydration reaction of cement as follows [17]:

$$\underset{Tricalsium\ Silicate\ (C_3S)}{2(3CaO.SiO_2)} + 7H_2O \rightarrow \underset{Calsium\ Silicate\ Hydrate\ (C-S-H)}{3CaO.SiO_2.4H_2O} + 3Ca(OH)_2 \quad (13)$$

$$\underset{Dicalsium\ Silicate\ (C_2S)}{2(2CaO.SiO_2)} + 5H_2O \rightarrow \underset{C-S-H}{3CaO.2SiO_3.4H_2O} + Ca(OH)_2 \quad (14)$$

$$\underset{Tricalsium\ Aluminate\ (C_3A)}{3CaO.Al_2O_3} + 2H_2O + Ca(OH)_2 \rightarrow \underset{C-(A,C)-H}{3CaO.Al_2O_3.Ca(OH)_2.12H_2O} \quad (15)$$

$$\underset{Tetracalsium\ Aluminate\ Ferrite\ (C_4AF)}{4CaO.Al_2O_3.Fe_2O_3} + 10H_2O + 2Ca(OH)_2 \rightarrow \underset{C-(A,F)-H}{6CaO.Al_2O_3.Fe_2O_3.12H_2O} \quad (16)$$

The reaction (7) and (8) or (13) and (14) arising of $C_3S_2H_3$ ($3CaO.2SiO_2.3H_2O$) or calcium silicate hydrate (C-S-H) or tubermorite gel and calcium hydroxide ($Ca(OH)_2$). Excess $Ca(OH)_2$ and water from the cement hydration reaction is then reacted with calcium aluminate ($CaO.Al_2O_3$) and calcium aluminate-ferrite ($CaO.Al_2O_3.Fe_2O_3$) had been forming crystals of $CaO.Al_2O_3$. $Ca(OH)_2.12H_2O$ or C-(A, C)-H and $CaO.Al_2O_3$. $Fe_2O_3.12H_2O$ or C-(A, F)-H [18].

In concrete technology, result of reaction of concrete forming generally was explained with hydration reaction like equation reaction (7) up to (12) or (13) up to (16). While time influence forming of crystals of constructor of concrete monolith that usually of explainable with Fig. 2 [19].

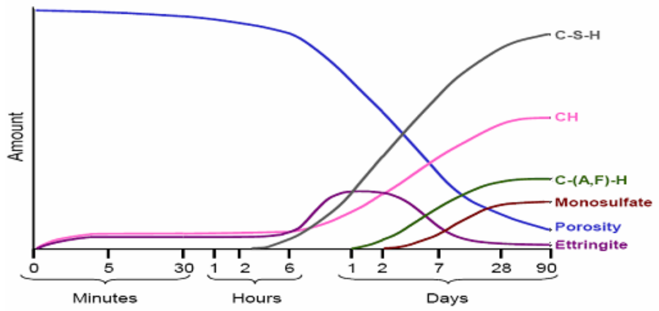

Fig 2. Curing time effects against amount of hydrate molecules and monolith porosity of the cement hydration results

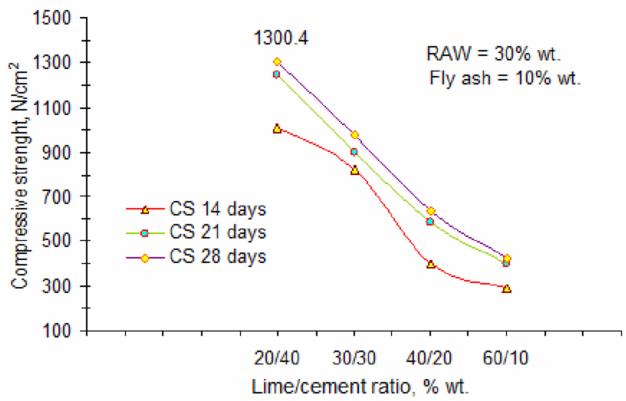

Fig 3. Effect of lime / cement ratio with 10% weight fly ash addition against compressive strength of immobilization results of RAW simulation

If the ratio of water and cement is proper, the cement hydration reaction through the phase of setting process, curing, and hardening of the gel tubermorite can take place perfectly (providing greater compressive strength) [20].

Effect of Limestone/Cement Ratio against the Immobilization RAW Simulations with Material Pozzolan of Fly Ash

Effect of lime and cement to the monolithic immobilized RAW simulation results with the addition of 10% weight fly ash is shown in the Fig. 3.

Fig. 3 shows that amount of lime that greater on the fixed amount of fly ash as much as 10% weight in the dough (waste + lime + cement) provide smaller compressive strength. This can be understood because the amount of lime that greater, hence the amount of cement in dough smaller. Number of cement that reduced in the dough causing the smaller amount of C_3S and C_2S were required for the formation of C-S-H crystals from cement hydration reaction in equation (13) and (14). The smaller amount of cement because of the increasing lime in the dough, hence smaller amount of C_3A and C_4AF were required for the formation of crystals of C-(A,C)-H and C-(A,F)-H of the cement hydration reaction in equation (15) and (16).

Important role of the hydrate compounds of C-S-H, C-(A,C)-H and C-(A,F)-H is as crystal molecules former hard monolith, hence the decreasing number of crystals of C-S-H, C-(A,C)-H and C-(A,F)-H by increasing the amount of lime in the dough causing compressive strength of the monolithic of dough at curing results for 14, 21, and 28 days became lower.

Effect of Fly Ash/Cement Ratio against Immobilization Results of RAW Simulation with Addition of Lime

Effect of fly ash/cement ratio against monolithic immobilization results of RAW simulation with addition of lime is shown in the Fig. 4, 5, and 6.

Fig. 4, 5, and 6 shows that with addition of fly ash and lime were gave a greater compressive strength monolith. This can be understood because the fly ash has a finer grain than granulated cement and has hydraulic properties like Pozzolan. By the nature of Pozzolan in fly ash, it can change the free lime Ca $(OH)_2$ (portlandite) as air mortar into hydraulic mortar as reaction (1), (2), and (3). Fly ash is expected not only to increase the compactness and density of concrete, but also can add strength. It is quite reasonable, because the fly ash mechanically will fill the empty space (cavity) between the grains of cement and a chemical will provide the hydraulic properties of the free lime (portlandite) generated from the hydration reaction in the cementation process, where this hydraulic mortar will be stronger than the air mortar (free lime + water).

In accordance with lime hardening theory which states that the ability of lime to harden due to its hydraulic forces, namely a comparison between CaO with the amount of (SiO_2 + Al_2O_3 + Fe_2O_3), this ratio is called the hydraulic modulus. The smaller hydraulic modulus shall increase ability of lime to harden in water.

Since the number of (SiO_2 + Al_2O_3 + Fe_2O_3) in lime only 2.64% [17], then with the addition of fly ash from coal-fired power plant in Tanjung Jati B that have a content (SiO_2 + Al_2O_3 + Fe_2O_3) 86.396% by weight, it will reduce the number of hydraulic modulus, so it can be understood that amount fly ash content greater, hence the hardness of monolith more was increasing [20].

From the constituent components, cement and fly ash have similar components. If the composition gives the best characteristics of monolith, it is supposed because of compositions of fly ash dominated by higher compound of alumina-silica when compared with cement and lime. The condition makes composition with larger fly ash component produce better monolithic characteristics, because the alumina-silica compounds in the fly ash will provide additional formation of several

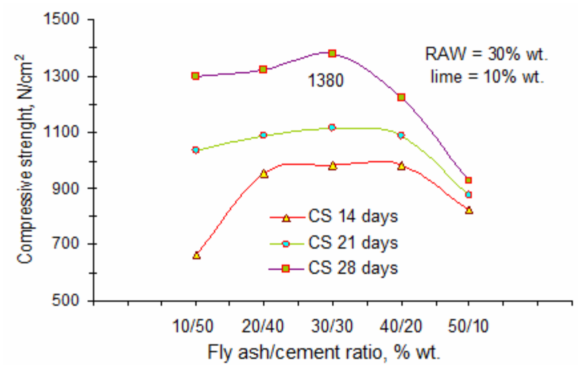

Fig 4. Effect of lime / cement ratio with 10% weight lime addition against compressive strength of immobilization results of RAW simulation

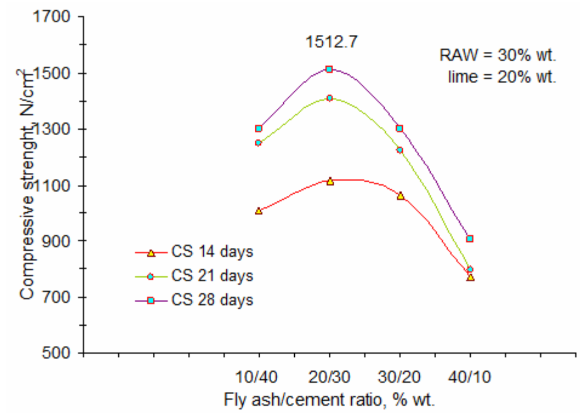

Fig 5. Effect of lime/cement ratio with20 % weight lime addition against compressive strength of immobilization results of RAW simulation

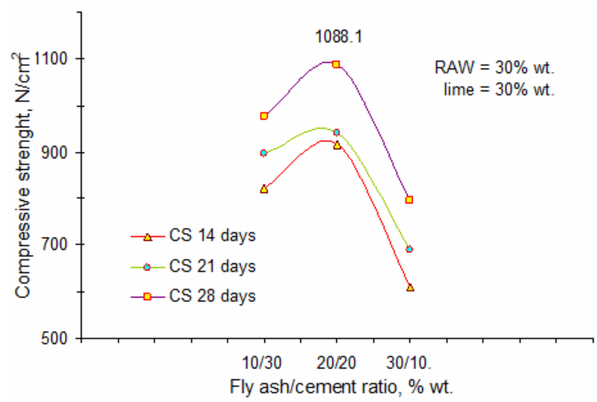

Fig 6. Effect of lime/cement ratio with 30% weight lime addition against compressive strength of immobilization results of RAW simulation

hydrate compounds of $CaO.SiO_2.2H_2O$, $CaO.Al_2O_3.6H_2O$, and $2CaO.Al_2O_3.Fe_2O_3.7H_2O$ such as in equation (1), (2), and (3). Hydrate compounds as additive formers of monolith initially will gave initial compressive strength lower than concrete of Portland cement type 1 that caused by the formation of ettringite

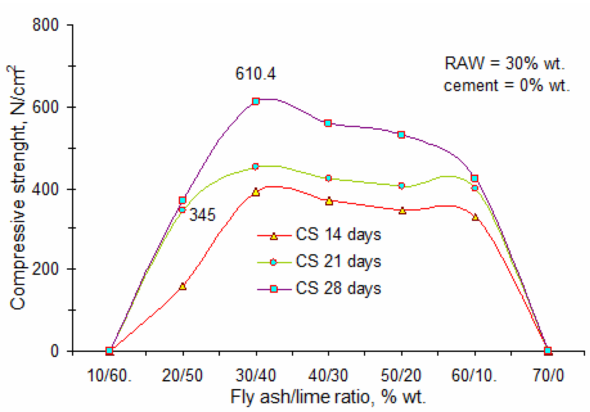

Fig 7. Effect of fly ash / lime ratio against immobilization results of RAW simulation without cement addition

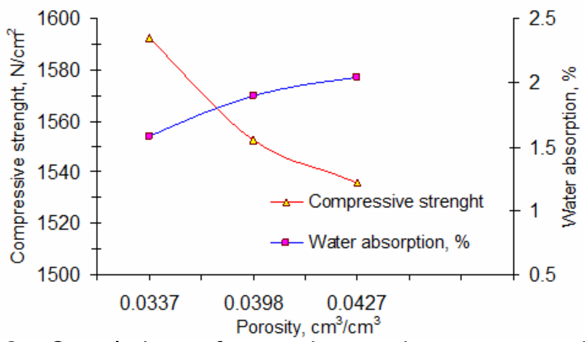

Fig 8. Correlation of porosity against compressive strength and water absorption of monolith on the best composition

($3CaO.Al_2O_3.3CaSO_4.31H_2O$), but the final compressive strength obtained on the monolith will be higher than the concrete of Portland cement type 1 [4,18].

Effect of Fly Ash/Lime Ratio against Immobilization Results of RAW Simulation without Cement Addition

Effects of fly ash/lime ratio against monolith of LRA immobilization simulation results without the cements addition are shown in the Fig. 7.

Fig. 7 shows that on the range ratio of fly ash/lime 20/50 – 60/10 (in % weight) gives the solid simulation of RAW immobilization results of monolith. This matter as evidence of pozzolanic character of fly ash from coal-fired power plant in Tanjung Jati B. Time of hardening monolith of Pozzolan usually slower than the monolith of Portland cement type 1, so the initial compressive strength lower than the compressive strength of monolith of Portland cement type 1, but the final compressive strength of monolith of Pozzolan will be higher than the monolith of Portland cement type 1.

Monolith from immobilization results of RAW simulation in curing > 28 days was not done in this study. However the compressive strength of monolithic immobilization results of RAW simulation in curing of 14, 21, and 28 days can be compared with the compressive strength monolith of immobilization results required by the International Atomic Energy Agency (IAEA). According to the IAEA Technical Report Series No. 222, monolithic quality standards of radioactive waste immobilization results after attain of age 28 days give limits monolithic compressive strength of 0.32 to 7 kN/cm^2 or 320 – 7000 N/cm^2.

Comparison between compressive strength the immobilization results of RAW simulation existing in Fig. 7 with the IAEA requirements result shows that the range ratio of fly ash/lime: 20/50 – 60/10 (in % weight) has been entered in the range of compressive strength as required by the IAEA by providing a minimum compressive strength of 345 N/cm^2 from the monolith curing results of 21 days and the highest compressive strength of 610.4 N/cm^2 from the monolith curing results of 28 days.

In general, Fig. 5 shows that the mineral composition forming the best monolith is which has composition (30% simulation radioactive waste, 30% cement, 20% lime, and 20% weight fly ash with the highest compressive strength of 1512.73 N/cm^2 compared with other mineral composition monoliths shaper. Beside that, the monolith compositions at the same curing time give different values of compressive strength. Furthermore, for the same monolith mineral composition, any change in curing time gives different compressive strength too. This case shows that the mineral composition had an effect on the compressive strength value of the monolith was resulted. Likewise, the curing time also had an effect on the compressive strength value of the monolith was resulted.

Absorption Characteristics of Water and Monolith Porosity on the Best Composition

Water absorption test and determination of monolith porosity were done as a simulation of the process of absorption of water as a discharger medium into the monolith through the pores of monolith because of the hydrostatic pressure gradient in water that is greater than the pressure inside the pores of monolith. The amount of water absorbed by the monolith pores will affect to the amount of radioactive substance that will slip off by the water out of the monolith pores in molecular diffusion. The occurrence of molecular diffusion in the monolith was caused water and substance was leached in the monolith has a higher concentration than the water outside the walls of monolith. The monolith porosity smaller usually was followed by the increasingly monolith quality compact and solid that can be identified by the greater compressive strength. The monoliths porosity are smaller, then the amount of water absorbed by the monoliths smaller so that radioactive substance are clamped by several hydrate compounds in the monolith to slip off by water in the monolith pores are smaller.

Correlation of monolith compressive strength of water absorption and monolith porosity characteristics on the best composition (30% RAW simulation, 30% cement, 20% lime, 20% fly ash) with curing time 28 is shown in the Fig. 8.

Fig. 8 shows that there is a correlation of water absorption, porosity and monolith compressive strength. The smaller porosity in the monolith resulted in more compact and dense monolith so that the water absorption by monoliths was smaller.

Since the limit value for water absorption of radioactive waste immobilization is not required by the IAEA, the data of water absorption by the monolith of 28 days curing in Fig. 8 can be compared with water absorption of hollow concrete bricks from a mixture of fly ash, lime and sand with water absorption value on the wane from 12% to 7.5% in weight addition of pasta in a mixture of 455.5 kg/m^3 to 723 kg m^3 [21].

If real radioactive substance was found in RAW simulation, the porosity results in Fig. 7 can be correlated with the speed of radionuclide slip of the porous monolith of immobilization results of RAW simulation. In the porous solid medium, the correlation between effective diffusion coefficient (D_e) and molecular diffusion (D_m) radionuclide with the porous solid media porosity (ε) and porous media tortuous (τ) are as follows [22]:

$$D_e = D_m \frac{\varepsilon}{\tau^2} \quad (17)$$

Whereas the correlation of effective diffusion coefficient (D_e) with fraction of radionuclides are released from the monolith of immobilization results of radioactive waste (FR) as follows [23]:

$$FR = 2\frac{A}{V}\left(\frac{D_e.t}{\pi}\right)^{1/2} \quad (18)$$

when, FR = fraction of radionuclides are released (g.cm^{-2}.day^{-1}), A = surface area of monolith (cm^2), V = volume of monolith (cm^3), D_e = effective diffusion coefficient (cm^2/sec), t = time (sec).

From equation (17), if the monolith porosity is smaller so effective diffusion coefficient (D_e) is smaller too. Then because D_e is smaller, hence from equation (18) will be obtained fraction of radionuclides are released (FR) became smaller.

CONCLUSION

The composition of the materials that make up the best monolith was 30% (wt) RAW simulation, 30% (wt) cement, 20% (wt) fly ash, and 20% (wt) lime with the monolith compressive strength of 1512.73 N/cm^2.

Using of fly ash and lime without cement with ratio composition of fly ash/lime 20/50 to 60/10 (in % (wt)) and RAW simulation of 30% weight can give the monoliths at curing time of 21 days with a minimum compressive strength of 345 N/cm^2 and at curing time of 28 days with the highest compressive strength of 610.4 N/cm^2. The monolith compressive strength result according to monolith compressive strength requirements of RAW immobilization results in accordance with the IAEA (320 – 7000 N/cm^2).

Water absorption test and monolith porosity determination of immobilization results of RAW simulation on monoliths forming composition can be represent the proportional correlation with the slip off speed of radionuclide at the monolith. Thus the smaller effective porosity at the monolith, will give smaller slip off velocity.

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