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UTILIZATION OF TROPICAL MATERIALS FOR ENSURING THE SAFETY OF STRONTIUM WASTE IMMOBILIZATION

Susetyo Hario Putero, Kusnanto, Widya Rosita and Nunung Prabaningrum
Department of Nuclear Engineering and Engineering Physics, Universitas Gadjah Mada, Jl. Grafika, Yogyakarta, Indonesia
E-Mail: susetyo@ugm.ac.id

ABSTRACT

Radioactive waste has to be immobilized prior to storing it in a storage place. The sustainability and safety of an immobilized waste should be ensured. This paper will describe how several tropical materials can be used to achieve that objective. Indonesia has plenty of natural resources, and the ability of its materials to immobilize strontium waste has been studied. The ability of zeolite as an adsorber on cement mortar had been studied. For increasing its compressive strength, coconut (*cocosnucifera*) and bamboo (*bambusavulgaris*) fibers had been compared to increase mortar's strength and durability, as well as rice husk ash as pozzolanic materials in theprevious study. The mortars were tested to determine their compressive strength by using Universal Wood Testing after being cured for 28 days. Strontium leaching rate encapsulated on the blocks was analyzed by using Hitachi Zeeman 8000 Atomic Absorption Spectrophotometer for 21 days. This research showed several materials that can be found in Indonesia, such as zeolite, natural fibers, and rice husk, are able to be used for immobilizing strontium waste. Cement mortar containing these materials can meet IAEA standards for compressive strength and strontium leaching rate of cementation, and hence the safety of strontium waste immobilization can be ensured.

Keywords: tropical materials, strontium waste, safe immobilization.

INTRODUCTION

Nuclear technology has been studied in Indonesia since 1954 when a national committee studied fallout produced by several nuclear weapon tests in the Pacific Ocean. Since 1965, Indonesia had operated a research nuclear reactor in Bandung. Recently, Indonesia was intensively studying various aspects, including radioactive waste management, for establishing first nuclear power reactor due to the global energy crisis.

Strontium, which has a long half-life (28 years), is one of the major fission products in nuclear spent fuel, radioactive waste associated with reactor operation and reprocessing [1]. Cementation is one of the several methods to immobilize radioactive waste prior to storing it in a storage place for minimizing radionuclide release into the environment. Cementation technique has to have the proper compressive strength and leaching rate for ensuring environmental safety. This paper will describe the study on the utilization of tropical materials, especially zeolite, coconut (*cocosnucifera*) fiber, and bamboo (*bambusavulgaris*) fiberas well as rice husk ash (RHA) as published by Putero, *etal.* (2012) [2], for immobilizing strontium waste that had been resulted.

THEORY

Cementation is a solidification process of radioactive waste using cement matrix. In general, cement has cheap, simple, strong mechanical properties and also a strong binding strength with radionuclide. There are several types of cement, such as Portland cement, white cement, and pozzolanic cement. Pozzolanic cement can reduce setting time and increase sulfate resistance. Pozzolanic material is composed of 45%-72% SiO_2, 10%-18% Al_2O_3, 1%-6% Fe_2O_3, 0.5%-3% MgO and 0.3%-1.6% SO_3. To form pozzolanic cement, 15%-40% of pozzolanic materials should be added to a Portland cement.

One of the natural pozzolanic materials is RHA obtained by burning rice husk due to its high content of silica (85~90%) as shown in Table-1. The moisture content of rice husk is relatively small because the husk is a residual of dry milling process. Commonly, RHA is only used as a primary or additional fuel in the bricks industry, decorating materials, or even discarded in animal cages [2].

Table-1. Composition of RHA [2].

Mineral compound	Composition (%)
CaO	0.28
SiO_2	85.94
Fe_2O_3	0.26
MgO	0.61
K_2O	1.36
Na_2O	0.33

The objective of radioactive waste cementation is to immobilize waste and to retain it on a concrete, and hence there is no radiation risk to humans and the environment. Based on the objective, IAEA requires the standard for radioactive waste cementation as follows [3]: (1) Density: 1.70-2.50 $g.cm^{-3}$, (2) Compressive strength: 20-50 $N.mm^{-2}$, (3) Compressive strength after waste loading: 2.5 N/mm^2, (4) Leaching rate (Rn): 1.70 x 10^{-1}-2.50 x 10^{-4} $g.cm^{-2}.days^{-1}$, (5) Dose rate in contact surface: 200 $mrem.hours^{-1}$, (6) Dose rate in 1 m from contact surface < 10 $mrem.hours^{-1}$, (7) Dose rate outside of interim storage 0.50 $mrem.hours^{-1}$.

To meet the requirement, physical properties of the cement can be improved by inserting fiber into it. Fiber can be classified intometallic fiber, polymeric fiber, mineral fiber and natural fiber [4]. Natural fiber, such as coconut and bamboo, is easier to be found and is cheaper due to its abundance in the tropical area, especially in Indonesia. Table-2 shows coconut plantation area and its productivity in Indonesia within the last 5 years [5-7]. Approximately 2 million Ha bamboo can be found in Indonesia, whichranks the third in Asia after India (almost 11.4 million Ha) and China (over 5.4 million Ha) [8].

Table-2. Coconut plantation in Indonesia from 2011 up to 2015 [5-7].

Years	**Area** (Ha)	**Production** (Ton)	**Productivity** (kg/Ha)
2011	3,767,704	3,174,379	1,158
2012	3,781,649	3,189,895	1.157
2013	3,654,478	3,051,585	1,130
2014	3,609,812	3,005,916	1,136
2015	3,571,376	2,960,851	1,131

A large number of zeolite in Indonesia was found in 1985 by BATAN and was spread in Sumatra and Java Island as shown in Figure-1. There are 46 locations of zeolite field, but intensive mining is carried out only in a few locations, such as in Bayah, Banten, Cikalong, Tasikmalaya, Cikembar, Sukabumi, Nanggung, Bogor, and Lampung [9]. The major component of Indonesian's zeolite isclinoptilolite and mordenite.Zeolite can be used as an ion exchanger and adsorber because it has an active side that can bind elements on a solution. Zeolite is also composed of silicate and alumina that will react with $Ca(OH)_2$ to form calcium silicate hydrate (CSH) named tobermorite. It improves cement's microstructure, hardening, and impermeability.

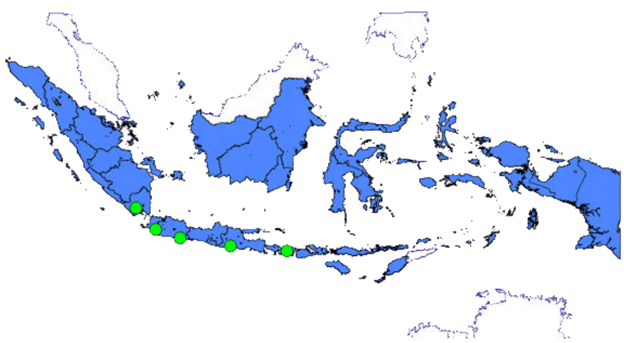

Figure-1. Zeolite resources in Indonesia [10].

METHODOLOGY

A. Zeolite and natural fiber effect

Activated natural zeolite was used to adsorbed Sr^{2+} on 200 ppm $Sr(NO_3)_2$ liquid. Zeolite adsorbing Sr ion (13%) was mixed with 3% water, type I Portland cement (SNI 15-2049-2004) and 0.50 $^{v}/_{o}$ natural fiber (coconut or bamboo). Cement used in this research contained SiO_2(20.8%), Al_2O_3 (6.9%), Fe_2O_3 (3.0%), CaO (63.7%), MgO (2.0%), SO_3 (1.6%), loss on ignition (1.5%) and insoluble residue (0.5%). The mixture was cast into a cylinder having a diameter of 2.2 cm and height of 4.4 cm. The cement reinforced mortars were heated at room temperatures of 50^0, 100^0, 150^0, 200^0 and 250^0C.

B. Reliability test

The cement mortars were tested to determine their compressive strength by using Universal Wood Testing after being cured for 28 days. The compressive strength, σ_t ($N.mm^{-2}$) was calculated by dividing force, F (N) by the surface area of cement mortar that the force applied, A (mm^2) as shown in Equation (1).

$$\sigma_t = \frac{F}{A} \quad (1)$$

The strontium leaching rate, R ($g.cm^{-2}.days^{-1}$), that wasencapsulated on the block,was tested for2, 4, 6, 9, 12, 15, 18, 21 days and was analyzed using a Hitachi Zeeman AAS type 8000. Data were calculated using Equation (2) and Equation (3) as described below.

$$R = \frac{A_t}{A_0} x \frac{W_0}{Sxt} \quad (2)$$

$$S = [0.5\pi D^2] + [\pi D h] \quad (3)$$

where A_t is activity of strontium at time t (Ci), A_0 is initial activity of strontium at time 0 (Ci), W_0 represents cement mortar weight (g), S represents surface area of cement mortar (cm^2), t is leaching time (days), D represents cement mortar diameter (cm) and h is cement mortar height (cm).

RESULTS AND DISCUSSIONS

Table-3 shows that bamboo fiber is more feasible than coconut fiber for reinforcing cement mortar. Becausethe modulus of elasticity of bamboo fiber (33~40 GPa) is greater than that of the coconut fiber (19~26 GPa) [4], bamboo fiber is better than coconut fiber in resisting force. Coconut fiber also absorbs more water than bamboo fiber due to its hygroscopic properties.

Table-3. Compressive strength of cement mortar reinforced by coconut fiber and bamboo fiber.

Temperature (^{0}C)	**Compressive strength** (N/mm^2)	
	Coconut	**Bamboo**
27	27.62	25.65
50	45.57	14.56
100	17.91	30.87
150	14.4	35.34
200	12.8	36.89
250	10.31	13.33

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When cement mortar is heated, the water remained on the mortar will evaporate. Then, thewater-cement ratio (w/c) becomes precisely 25%. The overheating will burn the fiber on themortar and enlarge the cavity in between cement matrix and fiber [11]. The strain of natural fiber also decreases along with temperature change. The decrease in fiber's elasticity reduces the fiber's ability in transferring the load on the matrix to the fibers [12]. At high temperatures, water on the fiber-matrix cavity will be evaporated which led to the emergence of the air cavity. All causes described above result in the change of mortar's compressive strength.

As an inorganic material, bamboo and coconut fiber are not durable enough under alkali condition inalong time period. In this research, alkali condition occurs due to the presence of $Ca(OH)_2$ as a product of hydration reaction. This condition is reduced by the reaction between zeolite and $Ca(OH)_2$, which forms CSH or tobermorite as described above.

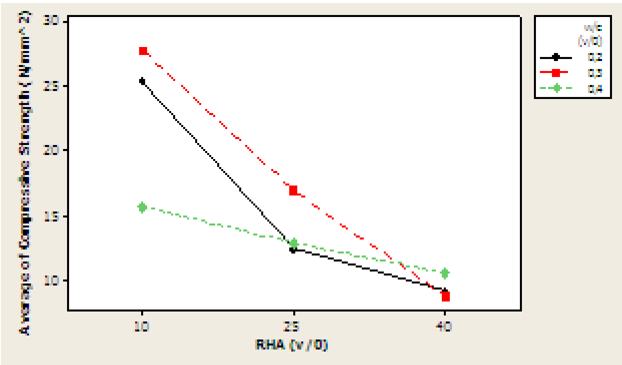
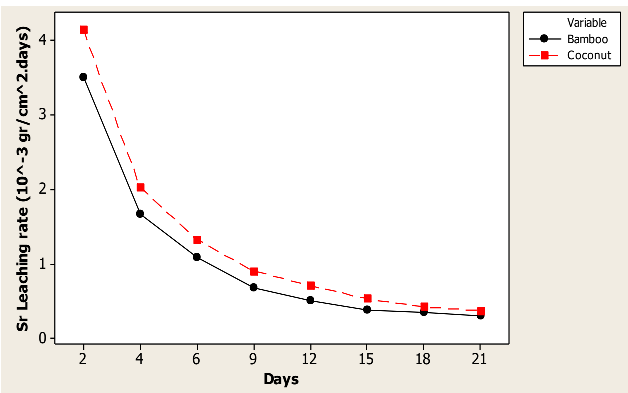
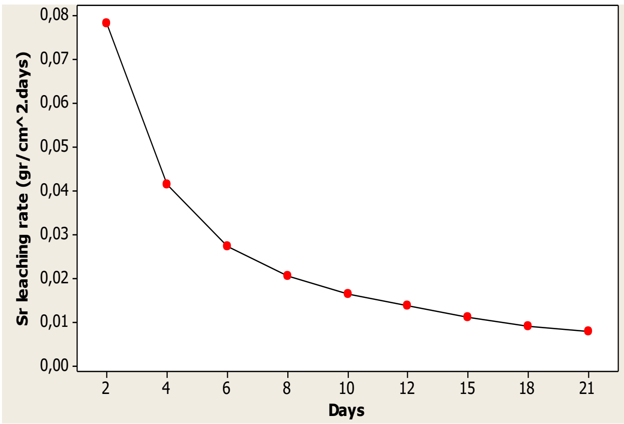


Figure-2. Effect of the addition RHA on compressive strength at various % of w/c [2].

According to Figure-2 above, the RHA addition will decrease mortar's compressive strength. But, whensuch strength is compared to the strength without the addition of RHA (16.645 ± 4.241 N/mm^2), the addition of RHA of up to 10 $^v/_o$results inimproving the compressive strength [2]. The compressive strength values shown in Table-3and Figure-2 are over 2.5 N/mm^2, that is minimum compressive strength for cementation [3], as well as Russian Federation standard (5 N/mm^2) [1].



(a)



(b)

Figure-3. Leaching rate of strontium in (a) cement mortars reinforced by coconut fiber and bamboo fiber, (b) RHA-cement mortar [2]

Figure-3(a) shows that the leaching rate of strontium on mortar reinforced by the bamboo fiber is lower than the one reinforced by coconut fiber. Natural fiber fills the cavities in the mortar so that the pores in the cement become smaller. It results in stronger containment of elements on cement mortar. It is also the result of zeolite presence on the mortar.

Strontium ion adsorbed in zeolitereplaces $M_{x/n}$(exchangeable cation) of zeolite structureor trapped in cavities. Strontium also has the isomorph replacement property with Ca and hence they can replace each other. Strontium will replace calcium inthe cement, and consequently, strontiumcontained in themortarwill be stronger. In this case, theeffect of zeolite is stronger than the effect of pore decrease. Therefore, the differences between bamboo and coconut fibers can be negligible.

The leaching rate of strontium in mortar reinforced by the natural fiber is lower than the one reinforced by RHA-cement shown in Figure-3(b). In Figure-3(b), sand was used as filler material, but in Figure-3(a) zeolite was used to replace sand. As described earlier, zeolite has anability to adsorb and toexchange ion, therefore strontium ion will be bound stronger on the mortar. However, the 91st leaching rates shown in Figure-3 are still under maximum IAEA standard (10^{-2} g/cm^2.days). Therefore, cement mortars enriched by tropical materials are able to immobilize strontium waste.

CONCLUSIONS

This research shows several tropical materials that can be found in atropical area, especially Indonesia, such as zeolite, natural fibers, and rice husk, that are able to be used for immobilizing strontium waste. Cement mortars containing these materials can meet IAEA standards for compressive strength and strontium leaching rate of cementation, and hence the safety of waste immobilization can be ensured.

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