

SORPTION AND DISPERSION OF STRONTIUM RADIONUCLIDE IN THE BENTONITE-QUARTZ-CLAY AS BACKFILL MATERIAL CANDIDATE ON RADIOACTIVE WASTE REPOSITORY

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ABSTRACT

The experiment of sorption and dispersion characteristics of strontium in the mixture of bentonite-quartz, clay-quartz, bentonite-clay-quartz as candidate of raw material for backfill material in the radioactive waste repository has been performed. The objective of this research is to know the grain size effect of bentonite, clay, and quartz on the weight percent ratio of bentonite to quartz, clay to quartz, bentonite to clay to quartz can be gives physical characteristics of best such as bulk density (ρ_b), effective porosity (ε), permeability (K), best sorption characteristic such as distribution coefficient (K_d), and best dispersion characteristics such as dispersivity (α) and effective dispersion coefficient (D_e) of strontium in the backfill material candidate. The experiment was carried out in the column filled by the mixture of bentonite-quartz, clay-quartz, bentonite-clay-quartz with the weight percent ratio of bentonite to quartz, clay to quartz, bentonite to clay to quartz of 100/0, 80/20, 60/40, 40/60, 20/80, 0/100 respectively at saturated condition of water, then flowed 0.1 N $\text{Sr}(\text{NO}_3)_2$ as buffer solution with tracer of 0.05 Ci/cm^3 ^{90}Sr as strontium radionuclide simulation was leached from immobilized radioactive waste in the radioactive waste repository. The concentration of ^{90}Sr in the effluents represented as C_t were analyzed by Ortec β counter every 30 min, then by using profile concentration of C_o and C_t , values of K_d , α and D_e of ^{90}Sr in the backfill material was determined. The experiment data showed that the best results were -80+120 mesh grain size of bentonite, clay, quartz respectively on the weight percent ratio of bentonite to clay to quartz of 70/10/20 with physical characteristics of $\rho_b = 0.658 \text{ g/cm}^3$, $\varepsilon = 0.666 \text{ cm}^3/\text{cm}^3$, and $K = 1.680 \times 10^{-2} \text{ cm/sec}$, sorption characteristic of $K_d = 46.108 \text{ cm}^3/\text{g}$, dispersion characteristics of $\alpha = 5.443 \text{ cm}$, and $D_e = 1.808 \times 10^{-03} \text{ cm}^2/\text{sec}$ can be proposed as candidate of raw material of backfill material in the radioactive waste repository.

Keywords: sorption, dispersion, strontium, backfill material

INTRODUCTION

A final repository for small and medium activities of the radioactive waste recommended by International Atomic Energy Agency can be done by near surface disposal and shallow land burial in the fine repository building at the best location [1]. Final repository by near surface disposal method must have multi-barrier system consisting of engineered barriers and natural barriers such as shown in Fig. 1 [2].

Backfill material is a segment of engineered barriers; it is a porous solid material from natural minerals that can be used to retard the radionuclides migration from the immobilized radioactive waste in the repository by which the environmental exposure on the soils as natural barriers in the surrounding repository can be reduced [1].

As backfill materials usually used natural minerals with the condition as follows: very low pore to minimize groundwater flow and ionic diffusion, good sorption properties to retard the movement of radionuclides in

case the container is broken, sufficient high thermal conductivity to provide adequate heat dissipation to the surrounding rock, sufficient load-bearing capacity to be supported as the container but sufficient plastic to dissipate high localized stresses, long-term chemical and physical stability under the temperatures, pressures and groundwater conditions of the vault [1,3]. The required properties of the buffer and backfill material are low permeability, high radionuclide retardation capacity, high swelling potential and good thermal and mechanical properties.

Bentonite has been considered as a candidate backfill material. As the high density bentonite has a low permeability, molecular diffusion will be the principle mechanism by which radionuclides will migrate through the backfill material, and the release of radionuclides will be limited due to their low ionic diffusion coefficient. However, as large quantities of bentonite are required to be use as a backfill material for a radioactive waste repository, bentonite-sand or bentonite-crushed rock mixture instead of pure bentonite

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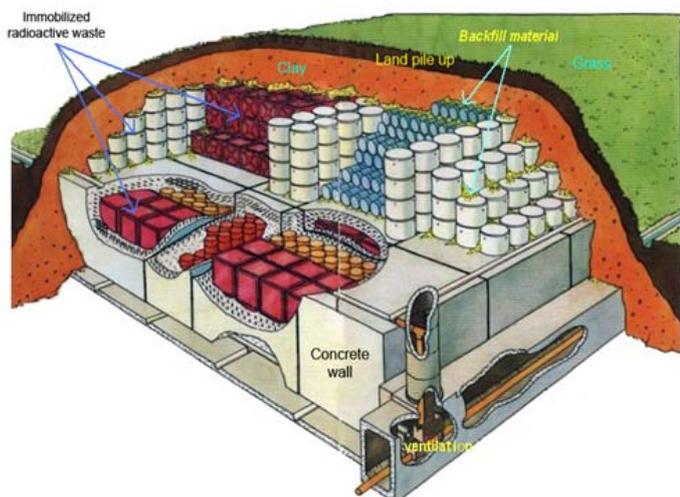


Fig 1. Concept of radioactive waste repository for near surface disposal system

has been considered as a backfill material from the viewpoint of the availability of the material and the economy. The hydraulic conductivities in the bentonite-sand mixtures with dry densities of 1.6 g/cm^3 and 1.8 g/cm^3 were measured within a sand content of 0 to 90 wt.%, and those in bentonites with a dry density of 1.4 g/cm^3 were also measured [4].

Scientist such as Zhijian [5] has already conducted the experiment on engineered barriers using local mineral e.g. bentonite, clay and quartz as backfill material and buffer material as part of engineered barriers system in the radioactive waste repository.

Particle size of minerals affect the physical characteristics i.e. bulk density (ρ_b), effective porosity (ε), and permeability (K). Physical characteristics of mineral, chemical-physical incident such as adsorption and ion exchange in the backfill material influence on the importance of the retardation of radionuclides migration [1,6].

In this paper, sorption and dispersion characteristics of the mixture of bentonite-quartz, clay-quartz, and bentonite-clay-quartz as candidate of raw material of backfill material in the radioactive waste repository are reported.

EXPERIMENTAL SECTION

Materials

The materials used in this experiment were bentonite has chemical composition in the weight % as follows: H_2O (1.80%), pH at $25 \text{ }^\circ\text{C}$ with 10% solid (8.05%), result of dry sample at $105 \text{ }^\circ\text{C}$: SiO_2 (64.73%), Al_2O_3 (13.14%), Fe_2O_3 (2.92%), CaO (4.13%), MgO (1.40%), TiO_2 (0.36%), K_2O (1.64%), Na_2O (1.97%),

H_2O^+ (6.03%) [7]. Whereas, quartz used in this experiment from accumulating basin of the Sermo Kulon Progo with SiO_2 content of 77.89 wt.%, and clay from the Kasongan Bantul. Whereas as simulation of radionuclide in the radioactive waste will be dispersed is tracer of ^{90}Sr . The simulation of radionuclide is ^{90}Sr tracer with concentration of 0.01 Ci/cm^3 in the liquid of $\text{Sr}(\text{NO}_3)_2$ 0.1 N as buffer.

Instrumentation

The equipments utilized in this experiment were sieving pans of ASTM ISO 585-R20 standard, oven Sybron, analytic balance Sartorius, β counter Ortec, glass column, glass apparatus.

Procedure

Preparation of Local Minerals (Bentonite, Quartz, Clay) Powders

Gravels of bentonite from Nanggulan Kulon Progo were dried in a oven until its constant weight, then it was crushed to powders. The bentonite powders were poured in the sieving pan of ASTM ISO 585-R20 standard with the sieve from upper to bottom of 16 mesh and 30 mesh, and then were sieved. The bentonite powder grain size of -16+30 mesh was kept on the sieving pan of 30 mesh. The same method was used for the sieve from upper to bottom of 30 and 40 mesh, 40 and 50 mesh, 50 and 60 mesh, 60 and 70 mesh, 80 and 120 mesh respectively until the grains size bentonite of -30+40 mesh, -40+50 mesh, -50+60 mesh, -60+70 mesh, -80+120 mesh respectively were obtained.

Preparation of quartz with SiO_2 content of 77.89 wt.% from accumulating basin of Sermo Kulon Progo and clay powders from Kasongan Bantul was carried out by the same method as that of bentonite powders.

Determination of Bulk Density of Bentonite, Quartz, and Clay Powders by ASTM D1895B A21.A Method

The bentonite powders were poured excessively into the bowl of cylindrical glass with known volume (V) and weight (M_1). The excess of bentonite powders was scraped horizontal by using thin blade precise at upper segment of the bowl. The bowl was filled with bentonite powders weighed as M_2 . Bulk density of bentonite (ρ_b) is determined by equation [8]:

$$\rho_b = \frac{M_2 - M_1}{V} \quad (2)$$

Determination of bulk density of quartz and clay powders was carried out by the same method as that of bentonite powders.

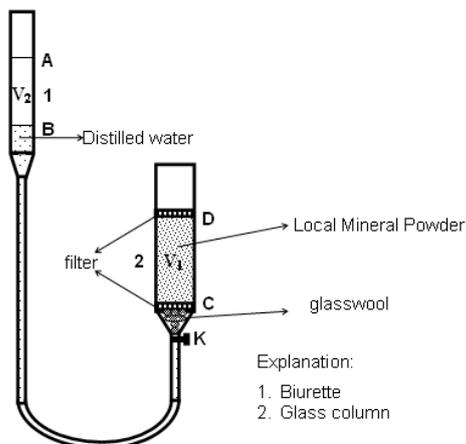


Fig 2. Apparatus for determination of effective porosity

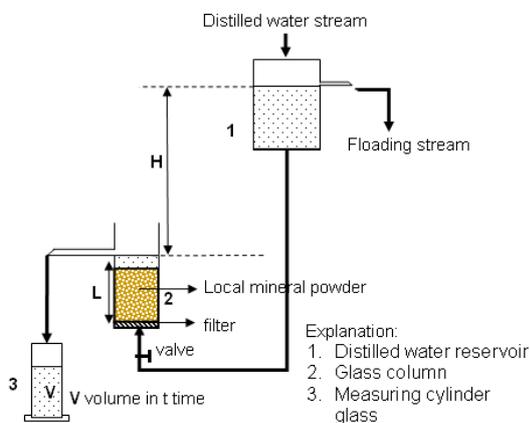


Fig 3. Apparatus for determination of permeability by constant head method

Determination of Effective Porosity of Bentonite, Quartz, and Clay Bed

The porosity of samples was determined by an apparatus as shown in Fig. 2. The 2 glass column filled with the powder of single local mineral (bentonite or quartz or clay) or the mixture of (bentonite-quartz or clay-quartz or bentonite-clay-quartz) of V_1 bed volume was flowed with distilled water from burette 1 through bottom of the 2 glass column. When the distilled water stream is precisely at bed bottom of local mineral powder at C boundary on the 2 glass column, then valve K on burette 1 was closed. Noted volume at A boundary, then distilled water was flowed to the 2 glass column filled with the local mineral powder by opening K valve. After all of the local mineral powder in the 2 glass column was flowed with distilled water to D boundary, and then K valve was closed. The decrease of distilled water volume in the burette from A to B boundary as was noted V_2 that represents particle pores and cavities between particles of local mineral powder in the 2 glass

column. Effective porosity (ε) can be calculated by equation as follows [4,8]:

$$\varepsilon = V_2/V_1 \quad (3)$$

Determination of effective porosity of quartz and clay powders was carried out by the same method as that of bentonite powders.

Determination of Permeability (K) of Bentonite, Quartz, and Clay Bed by Constant Head Method

The permeability of samples was determined as shown in Fig. 3. The 2 vertical glass column with inner diameter of $d = 1.75$ cm was filled with local mineral powder with height of $L = 10$ cm. Feeder glass column of 1 was filled with distilled water on the head loss of $H = 50$ cm. Distilled water from 1 reservoir was flowed to the 2 column through the bottom section. As the steady condition was achieved i.e. stream of inlet = stream of outlet 2 column, the stream of distilled water was stopped by closing the outer valve of 1 reservoir. The determination of time of distilled water stream (t) was began until the effluent volume of $V = 5$ cm^3 by opening the outer valve of 1 reservoir. Permeability can be determined by constant head method by equation as follows [9]:

$$K = \frac{V.L}{A.t.H} \quad (4)$$

with V = volume of effluent in t time (cm^3), L = height of local mineral bed in the glass column (cm), A = cross area of glass column (cm^2), t = time of V volume (sec), H = head loss (cm).

Determination of Sorption and Dispersion Characteristics of Strontium in the Mixture of Bentonite-Quartz, Clay-Quartz, and Bentonite-Clay-Quartz as Backfill Materials

Determination of sorption and dispersion of ^{90}Sr in the local mineral mixture as backfill material candidate were done by using ^{90}Sr tracer with concentration of $0.01 \mu\text{Ci}/\text{cm}^3$ in the buffer liquid of $\text{Sr}(\text{NO}_3)_2$ 0.1 N that flowed through local mineral mixture. The stream of buffer liquid of $\text{Sr}(\text{NO}_3)_2$ bringing ^{90}Sr tracer as simulation of ^{90}Sr was leached from packet of radioactive waste were immobilized on a radioactive waste repository. This radioactive waste has been immobilized as simulation of immobilization of evaporator concentrate from type of PWR 400 MWe reactor according to *Standard Reference Waste Form No.2/2* that transmitted radionuclides of β and γ with activity of $0.5\text{--}50 \mu\text{Ci}/\text{cm}^3$ [10].

The experiment was conducted by flow simulation of leaching ^{90}Sr pass a glass column loaded grains of local mineral mixture in gravitation according to data of vertical linear velocity = 10.3 m/year at shallow land in

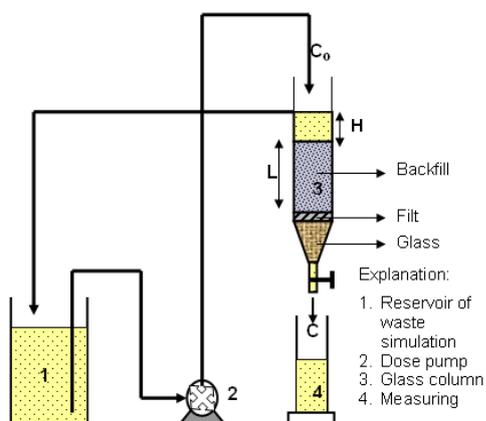


Fig 4. Apparatus for determination of ^{90}Sr sorption and dispersion in the backfill material

the area of Muria-Jepara peninsula as location candidate of nuclear power plant (NPP) and radioactive waste repository.

The determination of sorption and dispersion characteristics were carried out using a column filled with samples of bentonite-quartz mixture as simulates of backfill materials saturated with distilled water. Distilled water under samples of bentonite-quartz inside the column will exit when the distilled water was opened. As the distilled water surface is precisely reaching precision the surface of bentonite-quartz samples inside the column, influent from reservoir of 1 (as simulated the strontium leached due to the infiltration of surface water which pass the radioactive waste repository) flows through the column at an apparatus shown in Fig. 4.

The time by which the influent flows was recorded. The effluent from the column was collected in the graduated cylinder glass every 30 min, volume of effluent was measured and 0.1 cm^3 effluent entered into planchets and was dried by the lamp drier. Dry sample in the every planchet was analysis with β counter Ortec to know the activity or the concentration of ^{90}Sr in the effluent. Intake sample of the effluent discontinued after reached the concentration of ^{90}Sr in effluent that equal to concentration of ^{90}Sr in influent.

Interaction between nuclides and absorbent might be caused by chemical and physical interaction. One is interrelated to the other and it could not be defined separately. This basic phenomenon is unknown by specific model. Those interactions could empirically described by distribution coefficient (K_d). According to Poernomo [9], distribution coefficient is a constant value to express the ratio of total of chemical species adsorbed in the solid to concentration of chemical species in the solution contacted with solid at steady state. Distribution coefficient at batch process is determined by equation as follows [11-13]:

$$K_d = \frac{(C_o - C_e) / w}{C_e / V_e} \quad (5)$$

with K_d = distribution coefficient (cm^3/g), C_o = concentration of solute in the influent (g/cm^3), C_e = concentration of solute in the effluent (g/cm^3), w = weight of absorbent (g), V_e = volume of effluent (cm^3).

The determination of distribution coefficient at continuous flow process in the column filled with adsorbent of the porous solid medium is expressed by equation as follows [11- 13]:

$$K_d = \frac{V_T \cdot C_o - \sum V_t \cdot C_t}{\sum V_t \cdot C_t} \cdot \frac{V_T}{w} \quad (6)$$

with V_T = volume at the time where $C_t = C_o$ (cm^3), C_o = concentration of strontium in the influent ($\mu\text{Ci}/\text{cm}^3$), $\sum V_t \cdot C_t$ = accumulation of strontium concentration in the effluent where $C_t = C_o$ (μCi), w = weight of absorbent bed in the column as backfill material (g).

Weight of absorbent bed in the column is determined by equation as follows:

$$w = \rho_b \cdot v_b \quad (7)$$

with ρ_b = bulk density of absorbent bed (g/cm^3), v_b = volume of absorbent bed (cm^3) expressed by equation $v_b = (\pi/4) \cdot d^2 \cdot L$, which d = inside diameter of column (cm), L = height of absorbent bed in the column (cm).

Dispersion characteristics can be expressed with dispersivity (α) and effective dispersion coefficient (D_e). Dispersivity can be determined by equation as follows [11]:

$$\alpha = \frac{3 \cdot L}{16 \cdot \pi} \cdot \frac{(t_{1,0} - t_{0,0})^2}{(t_{0,5})^2} \quad (8)$$

with L = height of absorbent bed in the column (cm), $t_{1,0}$ = time at condition solute concentration in the effluent $C_o = C_t$ (min), $t_{0,5}$ = time at condition $C_t/C_o = 0.5$ (min), $t_{0,0}$ = residence time of solution in the absorbent bed in the column (min), and $t_{0,0}$ is calculated by equation as follows [11- 13]:

$$t_{0,0} = \frac{V}{Q/\varepsilon} = \frac{A \cdot L}{Q/\varepsilon} = \frac{\pi \cdot d^2 \cdot L \cdot \varepsilon}{4 \cdot Q} \quad (9)$$

with L = height of absorbent bed in the column (cm), A = cross area of glass column (cm^2), Q = volumetric velocity of influent (cm^3/min), ε = effective porosity of absorbent bed in the column (cm^3/cm^3), d = inner diameter of glass column (cm).

Effective dispersion coefficient are total of molecular dispersion coefficient cause gradient concentration of solute and hydrodynamic dispersion coefficient cause by solvent flow can be determined by equation as follows [12]:

$$D_e = \frac{\alpha \cdot V_w}{1 - R} \quad (10)$$

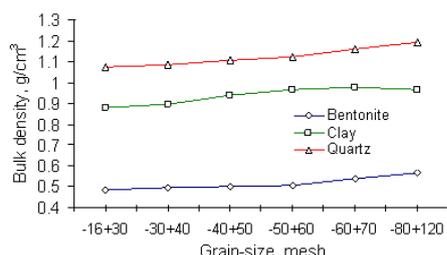


Fig 5. Effect of grain-size of local minerals to bulk density

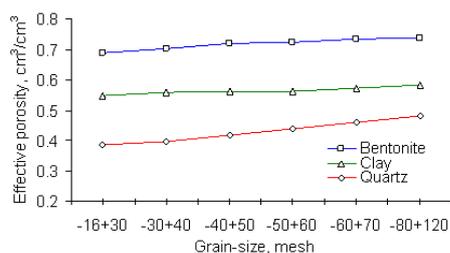


Fig 6. Effect of grain-size of local minerals to effective porosity

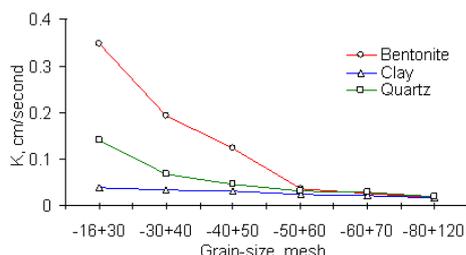


Fig 7. Effect of grain-size of local minerals to permeability

with V_w = linear speed of solvent stream in the porous solid medium, (cm/sec), R = retardation factor.

Determination V_w can be come near with the linear speed of ground water according to the following Darcy equation as follows [14]:

$$Q = K.A \frac{\Delta h}{\Delta L} \rightarrow \frac{Q}{A} = K. \frac{\Delta h}{\Delta L} \rightarrow V_w = K. \frac{\Delta h}{\Delta L} \quad (11)$$

with K = permeability or hydraulic conductivity (cm/sec), $\Delta h/\Delta L$ = gradient hydraulic, Δh = head loss (cm), ΔL = height of absorbent bed in the column (cm).

Determination of retardation factor (R) by using the following equation [15]:

$$R = 1 + \frac{\rho_b \cdot K_d}{\varepsilon} \quad (12)$$

RESULT AND DISCUSSION

Grain Size Effect on Physical Characteristics of Bentonite, Quartz, Clay Bed

The grain size affects the physical characteristics of bentonite, quartz, and clay. Therefore by knowing the

grain size, the best physical properties could be determined. Physical characteristics e.g. Bulk density (ρ_b), effective porosity (ε), and permeability (k) were showed in the Fig. 5, 6, and 7 respectively.

Fig. 5 shows that the smaller the grain sizes of the samples, the higher its density. This case is caused at constant volume; the smaller the grain size, the greater total particles to fill its volume. With total of particles increases, weight of the samples will increase so that the greater of the bulk density such as determined by using equation (2).

Fig. 6 shows that the smaller of grain size of samples, the greater the porosity (ε). This case is caused by grain size smaller in the constant bed volume; hence total of particles will be larger. Each particle has internal pore, and then particles to fill bed volume to form external pore between particles. Powders of the backfill material with smaller grain size in the constant bed volume cause internal and external pores greater until total of pore volumes in the bed or usually expressed by effective porosity greater. Effective porosity result of mineral with variation of grain size according to experiment result expressed by Poernomo [11-12].

Fig. 5-7 showed that bentonite, quartz and clay with grain size of -80+120 mesh gave the bigger bulk density, the bigger effective porosity, and the smallest permeability. According to this result, hence bentonite, quartz, and clay with grain size of -80+120 mesh respectively are used as the mixture of local mineral to determine physical characteristics in the backfill material mixture of bentonite-quartz, clay-quartz, and bentonite-clay-quartz. Fig. 5-7 showed that at grain size of -80+120 mesh, bulk density of quartz > clay > bentonite, effective porosity of quartz > clay > bentonite, and permeability of quartz \approx clay \approx bentonite.

Weight Ratio Effect of Mineral Local Mixture on Physical Characteristics

The effect of bentonite-to-quartz, clay-to-quartz, and bentonite-to-clay-to-quartz weight ratio on physical characteristics was showed on the Fig. 8, 9, 10 respectively.

Table 1 shows that mixture of bentonite-quartz with the composition bentonite of 80 wt.% and quartz of 20 wt.% give best of physical characteristics (ρ_b , ε , K). The mixture of clay-quartz with the composition clay of 60 wt.% and quartz of 40 wt.% gave best of physical characteristics. The mixture of bentonite-clay-quartz with the composition bentonite of 70 wt.%, clay of 10 wt.%, and quartz of 20 wt.% is the best of physical characteristics.

The biggest bulk density of mineral local mixture can increase the mechanical strength in holding canister

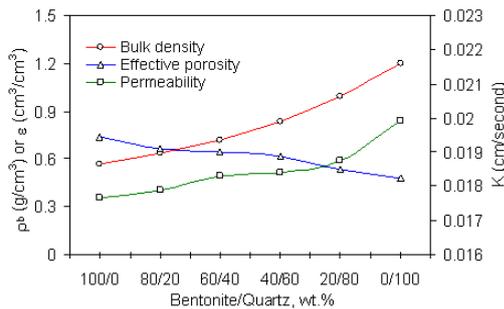


Fig 8. Effect of bentonite-to-quartz weight ratio to ρ_b , ϵ , K

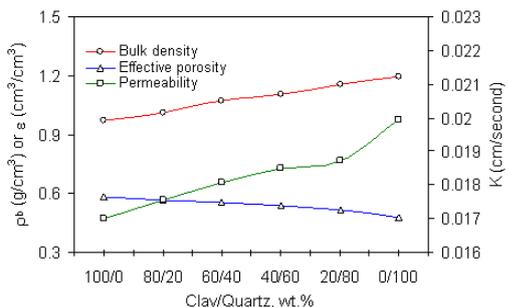


Fig 9. Effect of clay-to-quartz weight ratio to ρ_b , ϵ , K

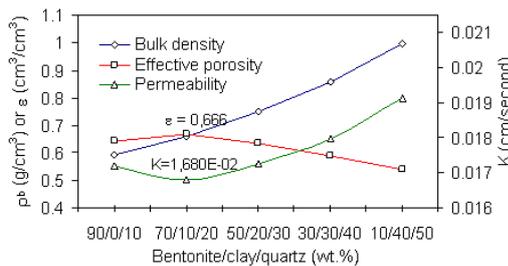


Fig 10. Effect of bentonite-to-clay-to-quartz weight ratio to ρ_b , ϵ , K

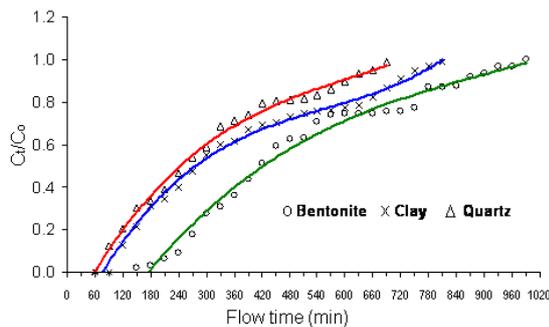


Fig 11. Concentration profile of ^{90}Sr in the effluent from bed of bentonite, clay, quartz in the glass column

of the radioactive waste in the repository such as certain by Nuclear Energy Agency (NEA) [3].

The smaller grain size of adsorbent in the experimental column will increase surface area and effective porosity of adsorbent particles. The bigger effective porosity at local mineral mixture of bentonite-to-clay-to-quartz weight ratio of 70/10/20 wt.% was

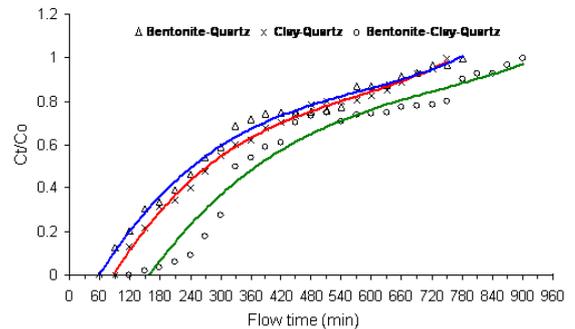


Fig 12. Concentration profile of ^{90}Sr in the effluent from bed of mixture of bentonite-quartz, clay-quartz, bentonite-clay-quartz in the glass column

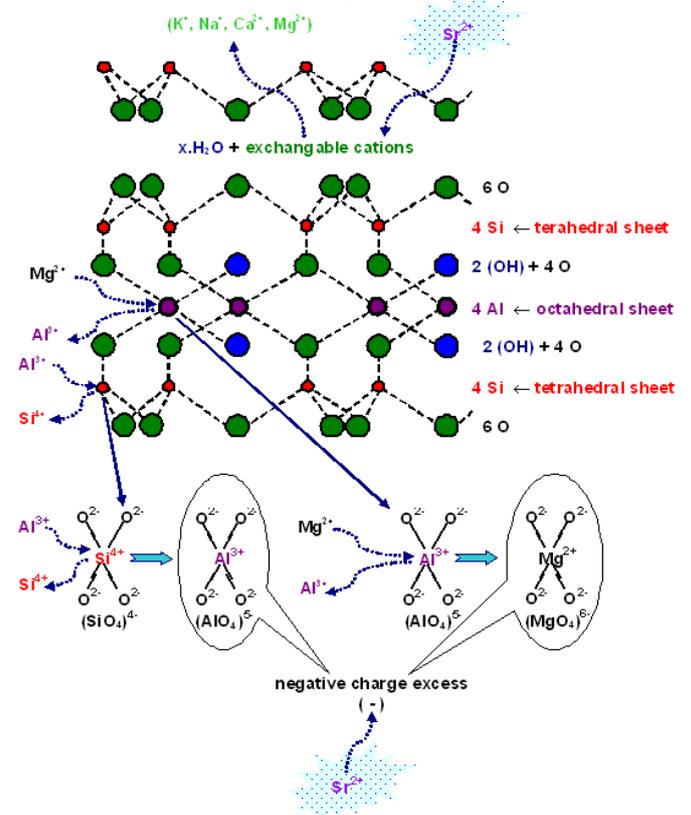


Fig 13. Replacement of exchangeable cations for Sr^{2+} and neutralization negative charge excess from isomorphous substitution of Al^{3+} for Si^{4+} in the tetrahedral sheets, and Mg^{2+} for Al^{3+} in the octahedral sheets of the bentonite mineral with Sr^{2+}

compared to other mixture will increase total of liquid filled internal pores of adsorbent bulk.

The smaller permeability at local mineral mixture of bentonite-to-clay-to-quartz weight ratio of 70/10/20 wt.% was compared to other mixture will the greater retardation of solution stream and the more extensive contact time of solution with adsorbent. Possibility of

Table 1. Effect of composition of local mineral mixture to sorption, dispersion characteristics

Composition of Local Minerals (wt.%)	Sorption characteristic	Dispersion characteristics	
	Kd (cm ³ /g)	α (cm)	De (cm ² /sec)
Bentonite (100 %)	59.723	4.224	1.324×10^{-03}
Clay (100 %)	22.557	6.127	2.866×10^{-03}
Quartz (100 %)	15.288	5.144	3.381×10^{-03}
Bentonite (80%) - Quartz (20%)	28.154	6.857	3.916×10^{-03}
Clay (60%) - Quartz (40%)	19.588	5.089	2.453×10^{-03}
Bentonite (70%) - Clay (10%) - Quartz (20%)	46.108	5.443	1.808×10^{-03}

contact between adsorbent grains with solution contain of strontium is bigger so that adsorption of strontium by bentonite become bigger.

Sorption and Dispersion of ⁹⁰Sr Characteristics on the Local Mineral

From the experiment by equipment such as Fig. 4 can be obtained concentration profile of ⁹⁰Sr in the effluent from glass column was filled single local mineral and local mineral mixture such as Fig. 11 and Fig. 12 respectively. With help Fig. 11 and Fig. 12 are used for determine characteristics of sorption of ⁹⁰Sr from equation of 6 and dispersion of ⁹⁰Sr from equations of 8 and 10.

Base on the result of analysis of ⁹⁰Sr activity in influent and effluent of artificial waste solution of strontium, the distribution coefficient can be determined, representing the amount of ⁹⁰Sr²⁺ in the solution adsorbed by backfill material, dispersivity i.e. represents ⁹⁰Sr²⁺ dispersion in the backfill material caused by the difference of ⁹⁰Sr²⁺ in the solution and in the particles of the backfill material, and convection that causes the flowing of fluid. The effect of backfill material formula on the strontium sorption characteristic was represented by strontium distribution coefficient (K_d) and strontium dispersion characteristics was represented by dispersivity (α) and the effective dispersion coefficient (D_e) shown in Table 1.

Result of the experiment in Table 1 showed that distribution coefficient of ⁹⁰Sr (K_d) in the bentonite mineral from Nanggulan Kulon Progo is 59.723 cm³/g only. This case caused by bentonite was used is natural bentonite from Nanggulan Kulon Progo contain of exchangeable cations such us K⁺, Na⁺, Ca²⁺, and Mg²⁺ to maintain the neutrality of the mineral charge. So the potential of ⁹⁰Sr²⁺ for the impurities cations in the natural bentonite will not be as good as that of active bentonite. This can be understood, because in the active acid used for an active agent, ⁹⁰Sr²⁺ cation only exchanged for H⁺ cation of the H-bentonite or Na⁺ cation of the Na-bentonite if NaCl used as active agent.

Because bentonite used is montmorillonite minerals, hence in bentonite exchange takes place at

three sites: on the flat surface, on the edge, and between the silica and alumina layers where ions are loosely held to neutralize deficiencies in these layers caused by isomorphous replacements. Isomorphous replacements are substitutions in the octahedral and tetrahedral units giving negative charge excess, unsatisfied valences on edges of unit. The excess negative charge on the mineral is neutralized by an equivalent number of positive ions. In natural situations the most common exchangeable cations are K⁺, Na⁺, Ca²⁺, and Mg²⁺.

If natural bentonite are annoyed with liquid contain of pollutant such as stream of the liquid of Sr(NO₃)₂ of 0.1 N with concentration of ⁹⁰Sr of 0.01 Ci/cm³. Element of ⁹⁰Sr as simulation of radionuclide was leached from radioactive waste was immobilized on a near surface disposal system such as Fig. 1, hence cation of Sr²⁺ replace exchangeable cations such as K⁺, Na⁺, Ca²⁺, and Mg²⁺ in bentonite mineral.

The isomorphous substitution of Al³⁺ for Si⁴⁺ in the tetrahedral sheets, and Mg²⁺ for Al³⁺ in the octahedral sheets. Result of isomorphous substitution of Al³⁺ for Si⁴⁺ and Mg²⁺ for Al³⁺ in the tetrahedral and octahedral layers giving negative charge excess respectively. Replacement of exchangeable cations for Sr²⁺ cation and neutralization negative charge excess from isomorphous substitution of Al³⁺ for Si⁴⁺ in the tetrahedral sheets, and Mg²⁺ for Al³⁺ in the octahedral sheets of the bentonite mineral with cation of Sr²⁺ can be illustrated in Fig. 13.

Quartz gives strontium distribution coefficient (K_d) of 15.288 cm³/g, this result is smaller than K_d of bentonite. This can be understood, because sorption of solute in the bentonite was caused by physical and chemical sorption, whereas in the quartz was due to physical sorption only. Physical sorption of strontium molecule will be netted in the cavity of among quartz particles or to stick on the quartz particles surface. Chemical sorption in the bentonite particles can be occurred because cations of K, Na, Ca, Mg in the bentonite particles on outer framework structure of alumina-silica will be exchanged by ⁹⁰Sr²⁺ cation. In addition, chemical sorption can occur because ⁹⁰Sr²⁺ cation to draw in the negative charge on the alumina-

silica crystal lattice. This because the isomorphous replacement of Al^{3+} for Si^{4+} and Mg^{2+} for Al^{3+} gives rise to a negative charge on the lattice. Such a replacement is common to all aluminosilicates, including clays, and the net negative charge must be compensated by cations to preserve the electroneutrality [11].

The addition of quartz to bentonite will decrease the adsorption potential of strontium by solid particles as represented by strontium distribution coefficient. The decrease of strontium distribution coefficient was caused by part of bentonite exchanged by quartz with the adsorption potential smaller than bentonite.

Backfill material candidate from the mixture of bentonite-clay-quartz with grain size of -80+120 mesh respectively with composition of bentonite-to-clay-to-quartz weight ratio of 70/10/20 wt.% gives the best physical characteristics (ρ_b , ϵ , K), sorption characteristic (K_d), and dispersion characteristics (α and D_e). At this composition, the three materials are interrelated/interdependent. Distribution coefficient and effective porosity of bentonite are higher than that of quartz, but dispersivity and bulk density of bentonite are lower than that of quartz. In the backfill material, bentonite functions as an ion exchanger, whereas clay and quartz function to strengthen in its physical conditions. Bentonite usually has bad physical properties such as low thermal conductivity, low resistance to radiation and react with acid.

On the other hand, quartz in the backfill material has good properties such as long-term chemical and physical stability under temperatures, pressures and condition of groundwater so that it can improve the physical properties of the backfill material due to the following reasons: SiO_2 content in the quartz higher than that of bentonite so that the physical strength of backfill material will increase and resistance to heat of radiation is occurred due to immobilized radioactive waste, bulk density of the clay and quartz higher than bentonite, so the potential of will be improved load-bearing capacity as a container of radioactive waste, grains of the quartz can increase the dissipation potential of high localized stresses by means of distribution the stress concentration at one point to surrounding rock evenly if the earth quake conditioned [3].

Backfill material by composition of the mixture bentonite-clay-quartz with grain size of -80+120 mesh respectively with zeolite-to-clay-to-quartz weight ratio of 70/10/20 wt.% gave physical characteristic represented by bulk density (ρ_b) of 0.65817 g/cm³, effective porosity (ϵ) of 0.66602 cm³/cm³, and permeability (K) of 1.680x10⁻² cm/sec, sorption characteristic represented by distribution coefficient (K_d) of 46.108 cm³/g, dispersion characteristics represented by strontium dispersivity (α) of 5.443 cm and effective dispersion coefficient (D_e) of 1.808x10⁻³ cm²/sec. This case can be

explained that space of strontium migration in the backfill material by composition of bentonite-clay-quartz mixture smaller if than that of the backfill material by composition of bentonite or clay or quartz only. The experiment result by Lopez [16] on the backfill material from a mixture of 19.5 mm diameter aggregate and 6 wt.% clay gave permeability (K) of the backfill material of 5.3x10⁻⁴ cm/second. Whereas experiments result by LeNeveu [17] gave K of the backfill material from phyllosilicate mineral of 2.2x10⁻⁶ cm/sec.

If K data from this experiment result was compared with K data from experiment result done by Lopez [16] and LeNeveu [17], accordingly the backfill material formula from a mixture of bentonite-clay-quartz with grain size -80+120 mesh respectively by bentonite-to-clay-to-quartz weight ratio of 70/10/20 wt.% can be proposed as candidate of raw material of backfill material on the radioactive waste repository.

CONCLUSION

From the experimental results it can be concluded that a mixture of bentonite-clay-quartz with grain size of -80+120 mesh respectively by bentonite-to-clay-to-quartz weight ratio of 70/10/20 wt.% with permeability of 1.680x10⁻² cm/sec, strontium distribution coefficient of 46.108 cm³/g, strontium dispersivity of 5.443 cm, strontium effective dispersion coefficient of 1.808x10⁻³ cm²/sec can be proposed as candidate of raw material of backfill material in the radioactive waste repository.

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REFERENCES

- Poernomo, H. and Kundari, N.A., 2006, *Indo. J. Chem.*, 6, 3, 261–267.
- Dixon, D.F., 1985, *A Program for Evaluation from Storage to Disposal of Radioactive Wastes at CRNL*, Atomic Energy of Canada Limited, AECL-7083.
- CECD/NEA, 2001, *Geological Disposal of Radioactive Waste Review of Development in the Last Decade [M]*, Atomic Energy Press, Beijing.
- Cho, J.W., Lee, J.O., and Kang, C.H., 2000, *J. Korean Chem. Soc.*, 32, 5, 495–503.
- Zhijian, W., 2006, *Chin. J. Rock Mech. Eng.*, 25, 4, 794–800.

6. Verstricht, J., Demarche, M., and Gatabin, C., 2001, *Development of Backfill Material within the Belgium Concept for Geological Disposal of High-Level Radioactive Waste: An Example of Successful International Co-operation*, Waste Management Conference, Tucson Arizona.
7. Munir, and Sedewo, B., 1981, *Pengaktifan Bentonit untuk Lumpur Bor dan Koagulan*, Laporan Lengkap Laboratorium Penelitian Teknologi Kimia, Departemen Teknologi Kimia, Fakultas Teknologi Industri, Institut Teknologi, Bandung.
8. Anonymous, *Physical Properties of Soil Bulk Density, Soil Porosity and Particle Density*, Modesto Junior College Agriculture and Environmental Sciences, <http://virtual.yosemite.cc.ca.us/agens/instruction/pollard/nr/resources/LAB%205%20Soils%20bulk%20density.doc>, January 7, 2010.
9. Tiwari, B., 2008, *Constant Head and Falling Head Permeability Test*, Civil and Environmental Engineering Department, Cal State Fullerton, http://faculty.fullerton.edu/btiwari/geotech_Lab/...files/.../Permeability.pdf, January 6, 2010.
10. Vejmelka, P., and Sambell, R.A.J., 1984, *Characteristic of Low and Medium-Level Radioactive Waste Form*, Commission on the European Communities, Luxembourg, 169.
11. Poernomo, H., 2004, *Difusi ⁹⁰Sr dalam Calon Bahan Urug Montmorilonit pada Penyimpanan Limbah Radioaktif*, Prosiding Pertemuan dan Presentasi Ilmian Penelitian Dasar Ilmu Pengetahuan dan Teknologi Nuklir, P3TM – BATAN, Yogyakarta.
12. Poernomo, H., Sardjono, and D., Budiyo, M.E., 2004, *Karakteristik Sorpsi dan Difusi ⁹⁰Sr dalam Abu Batubara sebagai Kandidat Bahan Urug pada Penyimpanan Limbah Radioaktif*, Prosiding Seminar Nasional Kimia XIV, FMIPA Kimia - Universitas Gadjah Mada, Yogyakarta.
13. Poernomo, H., Raharjo, and Suyatno, T., 2005, *Pasir Kuarsa sebagai Kandidat Backfill Material pada Penyimpanan Limbah Radioaktif*, Prosiding Seminar Nasional Kimia II, FMIPA–Kimia Universitas Islam Indonesia, Yogyakarta.
14. Darcy's Law for Modeling Groundwater Flow, <http://www.brighthub.com/engineering/civil/articles/58490.aspx>, January 6, 2010.
15. Eric Weber, L.G., and Kris Hendrickson, P.E., 2006, *Arsenic and Lead Mobility in Area-Wide Contamination-Impacted Soil*, Landau Associates, http://www.ecy.wa.gov/programs/TCP/SAB/SAB_mtg_info/mtg_061211/Addl%20As-Pd%20Mobility-TM%20Addend_TM.pdf, January 6, 2010.
16. Lopez, R.S., and Johnson, L.H., 1986, *Vault Sealing Research and Development for the Canadian Nuclear Fuel Waste Management*, Atomic Energy of Canada Limited - 9053.
17. LeNeveu, D.M., 1986, *Vault Submodel for the Second Interim Assessment of the Canadian Concept for Nuclear Fuel Waste Disposal: Post-Closure Phase*, Atomic Energy of Canada Limited - 8383.