Colloid-facilitated effects on migration of radionuclides in fractured rock with a kinetic solubility-limited dissolution model

Chun-ping JEN^{1,*} Neng-chuan TIEN²

¹Department of Mechanical Engineering, Chung Cheng University, Taiwan 62102 ²Energy and Environment Research Laboratories, Industrial Technology Research Institute, Taiwan 31040

Abstract Nuclides can move with groundwater either as solutes or colloids, where the latter mechanism generally results in much shorter traveling time as the nuclides interact strongly with solid phases, such as actinides. In the performance assessment, it is therefore essential to assess the relative importance of these two transport mechanisms for different nuclides. The relative importance of colloids depends on the nature and concentration of the colloids in groundwater. Plutonium (Pu), neptunium (Np), uranium (U) and americium (Am) are four nuclides of concern for the long-term emplacement of nuclear wastes at potential repository sites. These four actinides have a high potential for migrating if attached to iron oxide, clay or silica colloids in the groundwater. Strong sorption of the actinides by colloids in the groundwater may facilitate the transport of these nuclides along potential flow paths. The solubility-limited dissolution model can be used to assess the safety of the release of nuclear waste in geological disposal sites. Usually, it has been assumed that the solubility of the waste form is constant. If a nuclide reaches its solubility limit at an inner location near the waste form, it is unlikely that the same nuclide will reach its solubility limit at an outer location unless this nuclide has a parent nuclide. It is unlikely that the daughter nuclides will exceed their solubility limit due to decay of their parent nuclide. The present study investigates the effect of colloids on the transport of solubility-limited nuclides under the kinetic solubility-limited dissolution (KSLD) boundary condition in fractured media. The release rate of the nuclides is proportional to the difference between the saturation concentration and the inlet aqueous concentration of the nuclides. The presence of colloids decreases the aqueous concentration of nuclides and, thus, increases the release flux of nuclides from the waste form.

Key words Radionuclides, Colloid, Kinetic solubility-limited dissolution.

1 Introduction

Colloids are fine particles in sizes ranging from 1 nm to 1 μ m, which can be suspended in groundwater for long periods of time due to their small size. Colloids are of interest when considering the safety requirements of repositories for spent nuclear fuel because of their potential to transport nuclides from a defective waste canister to the biosphere. The traveling time for a specific nuclide is an important characteristic in the overall evaluation of the long-term safety for such repositories. Nuclides transport with groundwater either as solutes or bound with suspended

colloids, where the latter mechanism generally results in the unexpectedly rapid transport of the nuclides interacting strongly with solid phases, such as actinides. For a performance assessment (PA), it is therefore essential to assess the role of colloids in facilitating the transport of radionuclides. Plutonium (Pu), neptunium (Np), uranium (U) and americium (Am) are four nuclides of concern for the long-term emplacement of nuclear wastes at potential repository sites. The four actinides may have high mobility when they attach to the colloids in groundwater, such as iron oxide, clay or silica. Strong sorption of the actinides by the colloids in groundwater facilitates the transport

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^{*} Corresponding author. E-mail address: imecpj@ccu.edu.tw

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of nuclides along potential flow paths. The transport of actinides is governed by their solubility and speciation in groundwater; their tendency to be sorbed onto minerals, colloids or other matrices; and their propensity to form mobile species, such as dissolved cations, anions and colloids. Formation of nuclide-bearing colloids occurs due to sorption of nuclides onto colloids existing naturally in groundwater. The chemical composition and concentration of colloids may differ from site to site^[1-3]. The transport of actinides with low solubility, which is modeled without taking the existence of colloids into consideration, can be underestimated when the effect of colloids is practically significant.

In many countries, the processes of colloidal formation and transport are included in the FEPs (features, events, and processes) list of their PA programs for high-level radioactive waste (HLW)/spent nuclear fuel disposal. Although some PAs defer the consideration of colloid-nuclide transport until detailed evaluations are performed, other PAs, for instance, H12 of Japan^[4] and the Yucca Mountain Project of USA^[5], have considered the effects of the presence of colloids. The experimental results indicated that the engineered barrier designed with a bentonite buffer density of over 800 kg \cdot m⁻³ might be capable of to eliminating the colloid transport through the engineered barrier. However, failure of the engineered barriers and bentonite buffer to filter colloids should be considered as an alternative scenario^[6], especially according to the colloid and nuclide retardation experiments^[7,8] in 2003 -2004 carried out in situ at the Grimsel test site in Switzerland. Pioneering field studies^[9] have provided strong evidence that colloids played a role in the transport of low solubility radionuclides in a saturated, fracture flow aquifer. Moreover, observations at the Nevada Test Site (NTS), USA demonstrated that migration of plutonium occurred at 1.3 km from the site of the BENHAM underground nuclear test; and it was suggested that the migration might have occurred as a result of the attachment of plutonium to colloids [10]

The solubility-limited dissolution model can be used to assess the safety of the release of nuclear waste in geological disposal sites. It has been assumed that the solubility of the waste form is constant. However, when a nuclide reaches its solubility limit at an inner location near the waste form, it is unlikely that the same nuclide will reach its solubility limit at an outer location unless this nuclide has a parent nuclide. Ahn and Suzuki^[11] studied the effect of neptunium precipitation in the engineered barrier of a geologic repository and the surface of the waste form, and concluded that the location of the neptunium precipitation would be affected by the behavior of its parent, ²⁴¹Am, as well as the chemical form of the neptunium itself in the pore water. Garisto and Garisto^[12] developed a kinetic solubility-limited dissolution (KSLD) model to investigate the precipitation of the waste form. In their model, the release rate of nuclides from the waste form depended on the concentration difference at the waste form/buffer boundary. Li and Chiou^[13] successfully solved the analytical solution using the KSLD boundary condition for an open fracture, and found that colloids enhanced or retarded the transport of nuclides through the geosphere^[14].

The effective concentration of a nuclide includes the amount of nuclide associated with suspended colloids; thus, the nuclide may exceed its solubility limit. The existence of mobile colloids facilitates the radionuclide transport. In fact, for low solubility nuclides, the quantity of nuclide associated with colloids can be the most important contributor to an effective concentration. In this work, we investigated the effects of colloids on the transport of solubility-limited nuclides under the KSLD boundary condition in fractured media. The release rate of nuclides was proportional to the difference between the saturation concentration and the inlet aqueous concentration of nuclides. The presence of colloids decreased the aqueous concentration of nuclides and, thus, increased the release flux of nuclides from the waste form. Moreover, a mathematical model for evaluating the release rates of nuclides with kinetic solubility has been presented and demonstrated in the present study.

2 Conceptual model

The "far-field" (geosphere) is a physical region representing the natural barrier in the concept of the

geological disposal of nuclear waste. In the concept of deep geological disposal of nuclear waste, the natural barrier is the geological material hosting the repository. In determining the medium type of the far-field, any potential pathways must be identified. Usually, tight formations are considered as ideal candidates for hosting repositories. Due to various reasons, such as stresses and defects, the geological materials with heterogeneity are seldom perfect. The most common defects are fractures. Most rock formations considered for the geological disposal of nuclear waste contain fractures, which have greater void spaces than rock matrixes. Therefore, they are considered in our PA modeling as the fast pathway of the geological medium. Radionuclide transport in fractured rock is characterized by advection and diffusion/dispersion attenuated by sorption and matrix diffusion, and these aspects of the transport process must be considered in the PA modeling. Plutonium, uranium, americium and neptunium, which are of concern for long-term safety in HLW/spent fuel repositories, have low solubility. These nuclides may be absorbed on the suspended colloids, hence a higher effective solubility due to the presence of colloids and their easier transport facilitated by the colloids. Colloids are not influenced by matrix diffusion because of their charge and size, and the radionuclides attached to colloids may, therefore, not be subjected to the matrix diffusion processes; colloids tend to stay centre-stream, where the velocity is higher due to hydrodynamic forces during flow through fractures. It this study, a kinetic solubility-limited dissolution model was applied as the inlet boundary condition, and the flux of the radionuclides at the inlet of the fracture was proportional to the difference between the saturation

concentration and the inlet aqueous concentration. The proportionality constant is called the dissolution-rate constant. A thin rigid fracture slab in a saturated porous rock was considered herein. The following assumptions were made regarding the properties of the groundwater colloids and the system:

(1) The fracture was open and, hence, had unit porosity;

(2) In the fracture, groundwater flowed in one direction. Radionuclides in the aqueous phase, both dissolved and sorbed on the mobile colloids, transported advectively and dispersed along the fracture;

(3) The transverse dispersion efficiently ensured the mixing of radionuclide concentrations across the fracture slab in the direction perpendicular to the flow;(4) In the matrix, the groundwater was at rest and radionuclide diffusion only occurred in the direction perpendicular to the fracture;

(5) Sorption of colloids to fracture surfaces was conservatively ignored;

(6) Diffusion of colloids into the rock matrix was conservatively ignored because the diameter of colloids was relatively large;

(7) Sorption of nuclides to colloids was linear, instantaneous and reversible;

(8) The colloid concentration was constant in time and space owing to the dynamic equilibrium between colloid generation and sedimentation; and

(9) The velocity of the colloids was taken to be the same as that of the groundwater flow.

3 Mathematical model

Accordingly, the equation describing radionuclide transport with colloids in the fracture is:

$$R_{\rm f}^* \frac{\partial N_{\rm f}}{\partial t} + v_{\rm f}^* \frac{\partial N_{\rm f}}{\partial x} - D_{\rm f}^* \frac{\partial^2 N_{\rm f}}{\partial x^2} - \frac{\theta_{\rm m} D_{\rm m}}{b} \frac{\partial N_{\rm m}}{\partial z} \bigg|_{z=b} + \lambda R_{\rm f}^* N_{\rm f} = 0, x \ge 0, b \ge z > 0, t \ge 0$$
$$R_{\rm f}^* = 1 + \frac{K d_{\rm m} \rho_{\rm m} \delta}{b} + K d_{\rm c} C_0, \quad v_{\rm f}^* = v_{\rm f} + v_{\rm c} K d_{\rm c} C_0, \quad D_{\rm f}^* = D_{\rm f} + D_{\rm c} K d_{\rm c} C_0.$$
(1)

Similarly, the transport equation for radionuclides in the rock matrix is:

$$R_{\rm m} \frac{\partial N_{\rm m}}{\partial t} - D_{\rm m} \frac{\partial^2 N_{\rm m}}{\partial z^2} + \lambda R_{\rm m} N_{\rm m} = 0, \ x \ge 0, \ z \ge b, \ t \ge 0$$
$$R_{\rm m} = 1 + \rho_{\rm m} K d_{\rm m} / \theta_{\rm m}$$
(2)

where, $N_{\rm f/m}$ is the aqueous concentration (kg·m⁻³), $V_{\rm f/c}$ is the groundwater/colloid velocity in the fracture (m/yr), $D_{\rm f/c}$ is the longitudinal dispersion coefficient of radionuclides/colloids in the fracture (m⁻²·yr⁻¹), $\theta_{\rm m}$ is the rock matrix porosity, $D_{\rm m}$ is the diffusion coefficient of radionuclides in the rock matrix pores (m⁻²·yr⁻¹), λ

is the decay constant (yr⁻¹), $Kd_{m/c}$ is the sorption (distribution) coefficient for radionuclides on rock/colloid (kg·m⁻³), ρ_m is the bulk density of the rock (kg·m⁻³), δ is the sorption length for radionuclides on the fracture surface (m), *b* is half of the fracture aperture (m), and C_0 is the colloidal concentration in the fracture (kg·m⁻³). Subscripts "f" and "m" refer to the fracture and rock matrix, respectively.

To solve the analytical solution of radionuclide concentration in the fracture, initial and boundary conditions for the transport equations, Eq.(1) coupled with Eq.(2) are given as:

$$-D_{\rm f}^* \frac{\partial N_{\rm f}(0,t)}{\partial x} + v_{\rm f}^* N_{\rm f}(0,t) = k \left[N_0 - N_{\rm f}(0,t) \right] \qquad (3a)$$

$$N_{\rm f}\left(x=\infty,t\right)=0\tag{3b}$$

$$N_{\rm f}(x,t=0) = 0$$
 (3c)

$$N_{\rm m}(x,z=b,t) = N_{\rm f}(x,t)$$
(3d)

$$N_{\rm m}\left(x,z=\infty,t\right)=0\tag{3e}$$

$$N_{\rm m}(x,z \ge b,t=0) = 0$$
 (3f)

where, *k* is the dissolution rate constant $(m \cdot yr^{-1})$, and N_0 is the saturation concentration of the radionuclides $(kg \cdot m^{-3})$. The boundary condition of Eq.(3a) is, thus, a Cauchy-type boundary condition. For single-radionuclide modeling, the analytical solution of Eq.(1), subjected to the conditions of Eqs.(3a–3f), was obtained and the total mobile concentration of radionuclides (N_T), in the fracture could then be given as:

$$N_{T}(x,t) = N_{f}(x,t) \cdot (1 + Kd_{c}C_{0})$$

$$= \frac{kN_{0}(1 + Kd_{c}C_{0})}{v^{*}\beta} \int_{0}^{t} \left(\frac{1}{\sqrt{\pi\xi^{\xi}}} \cdot \exp\left[\alpha x - \frac{(\alpha\beta x)^{2}}{4\xi} - \left(\frac{1}{\beta^{2}} + \lambda\right) \cdot \xi\right] - \frac{2k + v^{*}}{v^{*}\beta}$$

$$\times \exp\left\{\alpha x \frac{2k + v^{*}}{v^{*}} + \left[\left(\frac{2k + v^{*}}{v^{*}\beta}\right)^{2} - \left(\frac{1}{\beta^{2}} + \lambda\right)\right] \cdot \xi\right\} \cdot \operatorname{erfc}\left(\frac{\alpha\beta x}{2\sqrt{\xi}} + \frac{2k + v^{*}}{v^{*}\beta}\sqrt{\xi}\right)\right) \qquad (4)$$

$$\times \left(\exp\left(-\frac{\sqrt{\lambda\xi}}{A}\right) \cdot \operatorname{erfc}\left\{\frac{\xi}{2A(t - \xi)^{\frac{1}{2}}} - \left[\lambda(t - \xi)\right]^{\frac{1}{2}}\right\}$$

$$+ \exp\left(\frac{\sqrt{\lambda\xi}}{A}\right) \cdot \operatorname{erfc}\left\{\frac{\xi}{2A(t - \xi)^{\frac{1}{2}}} + \left[\lambda(t - \xi)\right]^{\frac{1}{2}}\right\}\right) d\xi$$

where, $\alpha = v_{\rm f}^{*} / (2D_{\rm f}^{*})$,

$$\beta^2 = 4 D_f^* R_f^* / (v_f^*)^2$$
 and
 $A = b R_f^* / [\theta_m (D_m R_m)^{1/2}]$

The mass flux can be defined as:

$$J(x,t) = -D_{\rm f}^* \frac{\partial N_{\rm f}(x,t)}{\partial x} + v_{\rm f}^* N_{\rm f}(x,t)$$

and the exact analytical solution of the mass flux can be thus expressed as:

$$J(x,t) = k \left\{ \frac{N_0 (1 + Kd_c C_0)}{\sqrt{\pi}} \int_{\frac{\eta}{2\sqrt{t}}}^{\infty} \exp\left(\alpha x - \xi^2 - \frac{(\alpha x)^2}{4\xi^2} - \lambda YA\right) \times \left[\exp\left(-\sqrt{\lambda}Y\right) \cdot \operatorname{erfc}\left(\frac{Y}{2\sqrt{T}} - \sqrt{\lambda}T\right) + \exp\left(\sqrt{\lambda}Y\right) \cdot \operatorname{erfc}\left(\frac{Y}{2\sqrt{T}} + \sqrt{\lambda}T\right) \right] d\xi \right\} - kN_T(x,t)$$
(5)
where, $\eta = \sqrt{\frac{R_f^*}{D_f^*}} \cdot x$ and $Y = \frac{(\alpha\beta x)^2}{4A\xi^2}$.

4 Numerical illustrations

The analytical solution of Eq.(5) can be integrated by the Gauss-Legendre quadrature. Numerical results performed herein were based on a mathematical model for evaluating the kinetic solubility mass fluxes of nuclides at the inlet of the fracture and at a farther location, 100 m away. Numerical demonstrations were conducted by employing the parameters listed in Table

Table 1 Transport parameters used in the present work.

1. Colloidal velocity was assumed to be equal to groundwater velocity. The dispersivity of the colloids was size-dependent, as suggested by Grindrod *et al.*^[15] For mono-dispersed colloids, the dispersivity of the colloids was assumed to be 30 times greater than that of the nuclides. The colloidal concentration in groundwater ranged from 10^{-4} (experimental data^[16]) to 10^{-1} (assumed value^[17]) kg·m⁻³.

| Parameter | Value |
|---|---------------------------|
| Fracture velocity, $v / m \cdot yr^{-1}$ | 10 |
| Longitudinal dispersivity, α_L/m | 10 |
| Sorption length, δ / m | 1×10^{-4} |
| Fracture aperture, $2b / m$ | 5×10^{-3} |
| Bulk density, $\rho_{\rm b}$ /kg·m ⁻³ | 2700 |
| Rock matrix porosity (θ) | 0.02 |
| Molecular diffusion coefficient, $D_{\rm m} / {\rm m}^2 \cdot {\rm yr}^{-1}$ | 7.88×10^{-4} |
| Distribution coefficient of radionuclide on rock, Kd_m / kg·m ⁻³ | 1 |
| Decay constant of ²³⁷ Np, λ / yr ⁻¹ | 3.24×10^{-7} |
| Dissolution limit, $N_0 / \text{kg·m}^{-3}$ | 1 |
| Dissolution rate, $k / \text{m·yr}^{-1}$ | 1 |
| Colloid concentration, $C_0 / \text{kg} \cdot \text{m}^{-3}$ | 0.1 |
| Distribution coefficient of radionuclide on colloid, $Kd_c / \text{kg} \cdot \text{m}^{-3}$ | 10 <i>Kd</i> _m |
| Colloid dispersivity in the fracture, α_c / m | $30 \alpha_L$ |
| Colloid velocity in the fracture, v_c / m | 10 |

Our previous study^[18] showed that the effect of the presence of colloids became significant for instantaneous equilibrium sorption when the colloidal concentration was greater than $10^{-4} \text{ kg} \cdot \text{m}^{-3}$. Thus, the colloidal concentration used in the present study was assumed as 10^{-1} kg·m⁻³, from a conservative point of view. The sorption coefficient for nuclides on colloids was not easy to identify; therefore, it was assumed to be 10 times greater than that for nuclides on rock. ²³⁷Np was the analyzed as an important nuclide for the long-term safety of a nuclear repository. The ²³⁷Np transport can be altered by its parent; however, it was assumed that the precursors of ²³⁷Np did not exist here. The role of colloids in radionuclide transport in a single-fractured porous medium was presented in this study. The dissolution-rate constant, and the sorption coefficients for nuclides on rock and on colloids as well, were treated as important factors affecting the release rates from the source and altering the transport in the fracture. Thus, uncertainty analyses were also performed for these parameters.

Fig.1 shows the mass fluxes of ²³⁷Np for various dissolution-rate constants at the inlet of the fracture and at 100 m away. The released mass fluxes of radionuclides, with or without colloids, increased with the dissolution-rate constant. This phenomenon occurred at both the inlet of the fracture and 100 m away. The results indicated that the colloids facilitated the transport of nuclides in the fracture for the same dissolution-rate constant. The ²³⁷Np flux with colloids was about one order of magnitude larger than that without colloids. Interestingly, it was observed that the flux in absence of colloids with a dissolution-rate of 100 m/yr at the fracture inlet decreased to a minimum at a certain time and then rose again. For a higher dissolution-rate, a greater amount of radionuclides escapes instantly from the source and the aqueous concentration reaches its saturation level, hence the flux decrease at first, but when the radionuclide escape makes the aqueous concentration decrease to below the saturation level, the concentration gradient increases and the flux rises again.



Fig.1 Mass fluxes of 237 Np for various dissolution-rate constants at the inlet (a) and 100 m from the radioactive source (b) with / without colloids.

The sorption reaction of radionuclides on rock is characterized by a sorption coefficient that expresses the ratio between the adsorbed mass onto the solid and the mass in the fluid. Many factors that affect radionuclide sorption cannot be accounted for in dynamic geologic environments. As pointed out by Moreno et al.^[19], sorption reactions come from various mechanisms and complex processes. Perhaps the most common approach used in the assessment of sorption interactions in the subsurface is the linear isotherm approach, based on results of batch sorption experiments. In the present study, both matrix sorption and fracture-surface sorption were included in the model. If colloids are absent, the retardation factor caused by matrix sorption is expressed by Eq.(3) and the retardation factor caused by fracture-surface sorption is expressed as^[20]:

$$R_{\rm f} = 1 + \frac{Kd_{\rm m}\rho_{\rm m}\delta}{b} \tag{6}$$

The value of the sorption coefficients, Kd_m , for ²³⁷Np can be found in many PA reports. The values reported in SITE-94^[21] were adopted herein. Three factors including the retardation factor of the rock matrix, the retardation factor of the fracture surface and the sorption coefficient for radionuclides on colloids, were hypothesized to depend on Kd_m . The sorption of colloids enhanced radionuclide transport. This mechanism competed against the retardation effects induced by matrix and fracture-surface sorption. Fig.2 shows the mass fluxes of radionuclide for various Kd_m at the inlet of the fracture and at 100 m. At the fracture inlet, the mass flux was nearly independent of the value of Kd_m when colloids were absent. However, Kdm with larger values showed a significant retardation effect on the mass flux in the absence of colloids at 100 m. When colloids were present in the groundwater system, the large adsorb -ability of nuclides with colloids, which provided an alternative mobile carrier for the nuclides, implied the reinforcement of the mobility of the radionuclides. In other words, the rapid transport of radionuclides occurred mainly because the radionuclides adsorbed on colloids were not subject to retardation by the sorption of the rock matrix and the fracture surface.



Fig.2 Mass fluxes of ²³⁷Np for various Kd_m at the inlet (a) and 100 m away from the radioactive source (b) with/without colloids.

Sorption depth was another parameter which determined fracture-surface retardation. A surfacesorption coefficient (m) was defined instead of the term $Kd_m\rho_m\delta$ for simplicity. A conservative value of 1×10^{-4} m was employed as a default in this study to take into account the lack of knowledge of sorption depth. It was suggested that a value of zero for sorption depth^[22], so as to ignore the surface retardation if matrix diffusion occurs. It was also reported that surface retardation was altered by the wetted area^[22]. Thus, the effect of sorption depth or surface retardation was further studied.



Fig.3 Simulation results at the inlet (a) and 100 m away from the radioactive source (b) at sorption depth of 0.05 m while other parameters remained fixed.

Fig.3 shows the simulation results by increasing the sorption depth to 0.05 m while the other parameters remained fixed. Generally, the break-through curves, as shown in Fig.3(b) and representing the flux at 100 m, were similar to those shown in Fig.3(a), with smaller surface retardation. The breakthrough curves with a higher value of sorption depth depicted in Fig.3 exhibited a greater sensitivity

to the change of Kd_m values than those shown in Fig.2. Moreover, Fig.4 illustrates the break- through curves with colloids for the nuclides based on the parameters listed in Table 1 for $Kd_c = Kd_m$, $Kd_c = 10Kd_m$ and $Kd_c =$ $100Kd_m$. The numerical results in this figure indicated that the enhancement of colloids was significant when increasing the nuclide-colloid sorption.



Fig.4 Breakthrough curves of 237 Np at the inlet (a) and 100 m away from the radioactive source (b) with colloids for various $Kd_{\rm c}$.

5 Conclusions

This work has numerically investigated the effect of colloids on the transport of solubility-limited nuclides under the KSLD boundary condition in fractured media. A mathematical model for evaluating the release rates of nuclides with kinetic solubility has been presented in this study. The radionuclide-colloid interactions were not easily identified, and the linear isotherm assumption was made for radionuclide sorption on colloids in this study. The presence of colloids decreased the aqueous concentration of nuclides and, thus, increased the release flux of nuclides from the waste form. The simulation results show the mass fluxes of radionuclides for various Kd_m

at the inlet of the fracture and at 100 m. At the fracture inlet, the mass flux was nearly independent of the value of Kd_m when colloids were absent. However, Kd_m with larger values showed a significant retardation effect on the mass flux in the absence of colloids at 100 m. The numerical results also indicated that the enhancement of colloids became significant when increasing the nuclide-colloid sorption. Consequently, it was determined that the role colloids play in the transport of the radionuclide decay chain should be assessed carefully for the disposal of radioactive waste.

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