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Reduction of Nuclide Leaching from Paraffin Waste Form by Addition of Low Density PolyEthylene

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ABSTRACT

Low-level liquid concentrate wastes have been immobilized with paraffin wax using concentrate waste drying system in Korean nuclear power plants. Small amount of low density polyethylene(LDPE) was added to increase the leaching resistance of the existing paraffin waste form and the influence of LDPE on the leaching behavior of waste form was investigated. It was observed that the leaching of nuclides immobilized within paraffin waste form remarkably reduced as the content of LDPE increased. The acceptance criteria of paraffin waste form associated with leachability index and compressive strength after the leaching test were met with the help of LDPE.

1. INTRODUCTION

Low-level liquid borate wastes arising from the operation of Korean nuclear power plants have been evaporated, concentrated and immobilized with paraffin wax using concentrate waste drying system(CWDS) since 1995. CWDS has the advantages of high volume reduction(about 1/8 compared with cement waste form), decrease in radiation expose dose to worker(1/9), low cost and simple manufacturing process although paraffin waste form can be classified as class A unstable. Paraffin waste form should satisfy various acceptance criteria of radioactive waste form in order to guarantee the long-term safety performance of the planned radioactive waste repository in Korea. Among these, leaching resistance and compressive strength after the leaching test were mainly considered in this study.

Previous leaching test of paraffin waste form showed that 55~65% of boric acid and 63~70% of initial mass in case of cobalt, strontium and cesium were released during ninety days. It was observed that leachability indexes(LIs) of boric acid and cobalt were 5.8 and 7.2, respectively.[1] LI was used to define the performance criteria of waste form and U. S. NRC[2] recommended that

LI of waste form should be greater than 6. Low density polyethylene (LDPE) was chosen as an additive to solidifying agent for the purpose of improving the leaching resistance of paraffin waste form. It was shown that the leaching of nuclides from paraffin waste form remarkably decreased in spite of a little addition of LDPE.

2. PREPARATION OF WASTE FORM

The CWDS in Korean nuclear power plant has produced the paraffin waste form with the mixing ratio of 78:22 between boric acid and paraffin. Paraffin played a binding role within waste form and the concentrate waste including boric acid was encapsulated by paraffin. The mixing ratio of boric acid to paraffin was very important in order to make a homogeneous waste form because it made a difference between boric acid(1.44) and paraffin(0.933) in specific gravity.

Small amount of LDPE was added to waste form in order to increase the leaching resistance of paraffin waste form and the influence of LDPE on the leaching behavior of waste form was investigated. The density of LDPE(0.925 g/cm^3) was similar to that of paraffin(0.933 g/cm^3) so that the stratification could be avoided during the preparation of waste form. The LDPE also had the melting temperature(120℃) similar to operating temperature of CWDS. If the LDPE were used in small quantities, the existing facilities of CWDS would be enough to operate without supplementary equipment such as extruder.

Paraffin waste forms with various mixing ratio of paraffin to boric acid were prepared with the change of LDPE as shown in Table 1. The maximum content of LDPE in Table 1 was limited by the workability during the manufacturing process. As the content of LDPE increased, the mixture grew more viscous and the more paraffin should be included to prevent a low fluidity of mixture.

Table 1. Results of Leaching Test

Waste Form	CASE 1		CASE 2		CASE 3		CASE 4	
Constituents, Boric Acid/Paraffin/LDPE(wt%)	78.0/22.0/0.0		78.0/20.9/1.1		70.0/28.5/1.5		60.0/37.0/3.0	
	Boric Acid	Cobalt	Boric Acid	Cobalt	Boric Acid	Cobalt	Boric Acid	Cobalt
Overall Reaction Rate Constant K_D(day^{-1})	8.95 $\times10^{-4}$	9.22 $\times10^{-4}$	1.39 $\times10^{-4}$	1.70 $\times10^{-4}$	1.42 $\times10^{-5}$	2.73 $\times10^{-5}$	1.00 $\times10^{-5}$	1.90 $\times10^{-5}$
Effective Diffusion Coefficient D_e(cm^2/sec)	1.59 $\times10^{-6}$	5.79 $\times10^{-8}$	2.39 $\times10^{-7}$	1.07 $\times10^{-8}$	2.11 $\times10^{-8}$	1.71 $\times10^{-9}$	1.19 $\times10^{-8}$	1.20 $\times10^{-9}$

Leaching Index LI	5.8	7.2	6.6	8.0	7.7	8.8	7.9	8.9
Compressive Strength after the Leaching Test of 90 days psi (MPa)	203 (1.38)		598 (4.07)		681 (4.63)		936 (6.37)	

3. RESULTS AND DISCUSSIONS

It was observed that the leaching of nuclides immobilized within paraffin waste form reduced remarkably as the content of LDPE increased. After the leaching test of 260 days according to ANSI/ANS-16.1 procedure[3], the paraffin waste form with no additive had the cumulative fractions leached(CFLs) of 0.95 and 0.96 for boric acid and cobalt, respectively, as shown in Fig. 1. To the contrary, the paraffin waste form whose constituents were boric acid of 70%, paraffin of 28.5% and LDPE of 1.5%, had the CFLs of 0.17 and 0.24 for boric acid and cobalt, respectively. In this case, LIs of boric acid and cobalt were 7.7 and 8.8, respectively, which satisfied the U. S. NRC recommended standard of > 6.

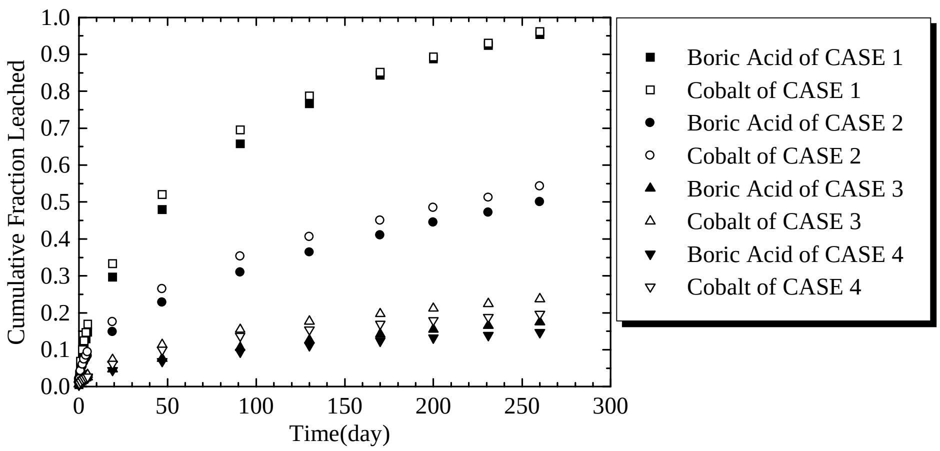

Fig. 1. CFLs of Boric Acid and Cobalt with the Change of LDPE during 260 days

LIs of nuclides were calculated from the developed leaching mechanism of paraffin waste form, shrinking core model based on the diffusion-controlled dissolution kinetics.[1]

$$1-\frac{2}{3}CFL-(1-CFL)^{2/3}=K_D\,t=2D_e\,\frac{C_o}{q_o}\frac{t}{r_o^2} \qquad (1)$$

where, K_D was overall reaction rate constant(day^{-1}), t time(day), D_e effective diffusion coefficient(cm^2/sec), C_o solubility of nuclide(g/cm^3), q_o concentration of undissolved nuclide within waste form(g/cm^3), and r_o initial radius of waste form(cm). This shrinking core model assumed that the reaction between solid and fluid was noncatalytic and solid particle was spherical with equivalent surface area. Although the paraffin waste form was finite cylinder, the above shrinking core model could be reasonably applied to the leaching analysis of paraffin waste form because the leaching rates were almost constant along all the surfaces, i.e., top, bottom and lateral sides, of the cylindrical waste form. The overall reaction rate constants and effective diffusion coefficients of nuclides were obtained using Eq. (1) as shown in Fig. 2.

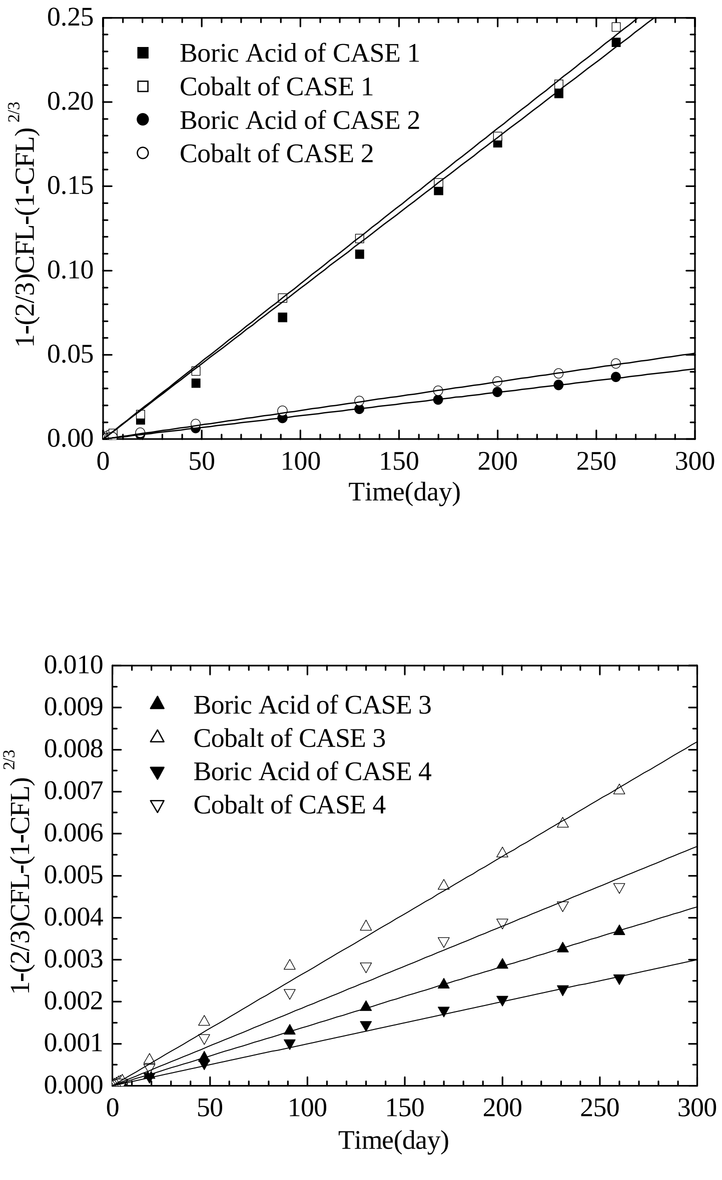

Fig. 2. The Relation between Shrinking Core Model Predictions and Test Data for Overall Reaction Rate Constants and Effective Diffusion Coefficients

It seemed that the reasonable content of LDPE was 1.5% in consideration of LI and volume reduction of concentrate waste. There was little difference between CASE 3 and CASE 4 in Table 1 although the content of LDPE in CASE 4 was twice as much as that in CASE 3.

The compressive strength test whose purpose was to confirm the integrity of produced waste form was carried out after 90 days of the leaching test. The test was performed according to ASTM C39-86.[4] The criterion of compressive strength for paraffin waste form has not been determined yet. Accordingly, the recommended standard by the U. S. NRC of > 60 psi (408 kPa) for bitumen and > 500 psi (3.4 MPa) for cement waste form, respectively, was applied instead after the leaching test. The paraffin waste form with no LDPE met the recommendation for bitumen, but not for cement waste form. In the case where the LDPE was added to waste form, CASE 2 ~ CASE 4 in Table 1, all the above recommended standards was fully satisfied.

4. CONCLUSIONS

The leaching test of paraffin waste form using of LDPE as an additive to solidifying agent was performed. The leaching resistance of paraffin waste form was remarkably improved despite of a small amount of LDPE. The acceptance criteria of paraffin waste form associated with leachability index and compressive strength after the leaching test were satisfied with the help of LDPE so that the long-term safety performance of the planned radioactive waste repository in Korea could be more surely guaranteed.

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