Study on Engineering Barrier Role in Nuclear Waste Disposal

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ABSTRACT

This paper studies the leaching behaviors of pyrochlore-rich synroc incorporated 46.8 wt% simulated actinides waste under the five simulated geological disposal media, which included the bentonite, granite, granite + ferroferric oxide, granite + cement, bentonite + ferroferric oxide, respectively. The mass loss rates reached to equilibrium after 182 day and was 10-7 g/cm2·d. That suggests the mass loss rate of pyrochlore-rich synroc, loaded 46.8wt% actinides waste, was very low. The surfaces of the leached specimens were analyzed by XRD, SEM/EDS. The experimental results show that the pyrochlore-rich synroc samples in the systems, which contained bentonite and cement, have two new phases formed on the leached specimens surface at 90 ℃ for 728d; The bentonite and cement can retard the elements leaching; Fe3O4 can speed the elements leaching; Expect for Ti ion depleted on the sample surface, other ion, such as U, Zr, Al, Ca, were in equable states and Ba ion was enriched during test time, which indicated the simulated disposal media have good ability to retard the leaching behavior of the pyrochlore-rich synroc.

INTRODUCTION

Safe disposal of HLW in the geologic formation to keep them far from human being for ten thousand years is hopeful. Since synroc has good durability and can highly incorporate actinide waste, synroc is expected as an ideal carrier of HLW [1]. To assess high level waste disposed into geologic condition, some study on immobilized waste material had developed in many literatures[2]~[4]. This experiment fabricated pyrochlore-rich synroc loaded 46.8 wt% simulated actiniums waste to study the leaching behaviors of the pyrochlore-rich synroc under simulated disposal conditions.

EXPERLMENTAL

Specimens fabrication and equipments

The pyrochlore-rich synroc was fabricated as the steps described in reference [5]. Nd and U were used as the simulacrum for trivalent and tetravalent actinides, respectively.

Before starting the experiment, cut the synroc samples into 10×10×2 mm specimens. The oxide compositions [6] of the specimens are listed in the Table 1.The experimental containers were made of PTFE and washed according to reference [7] before used in the experiments.

Table1. The oxide compositions of the synroc samples(waste loading 46.8%)

composition	Al_2O_3	BaO	CaO	TiO_2	ZrO_2	Nd_2O_3	UO_2
wt%	0.7	1.0	9.4	42.2	2.7	4.9	39.1

The leachates were analysed by AAS and ICP-MS (VG Plasma Quad Ⅱ Plus). The phase development was examined by X-ray diffraction using Rigaku D/MAX-RB instrument (Cukα radiation,λ= 1.54 Å). And the phase surfaces were determumed by SEM/EDS using a Britain Cambriage S-250 MK3 instrument.

Simulated disposal medium

The experiments designed five experimental systems to simulate the disposal condition. The mediums of five systems are as follow: P1, granite (100%); P2, granite (80%) + cement (20%); P3, granite (80%) + Fe3O4 (20%); P4, bentonite (100%); P5, bentonite (80%) + Fe3O4 (20%). The granite was came from Beishan site (a candidate disposal site in Gansu Province, China) and the composition of granite is shown in the Table 2; The bentonite was gathered from Weifang, Shangdong Province, and the composition is listed in the Table 3.

Table2. The oxide compositions of granite

Oxide	SiO_2	TiO_2	Al_2O_3	Fe_2O_3	FeO	MnO
wt%	67.2	0.61	15.98	0.71	2.48	0.04
Oxide	CaO	Na_2O	K_2O	P_2O_5	MgO	
wt%	3.51	3.76	3.36	0.13	0.04	

Table3. The oxide compositions of bentonite

Oxide	SiO_2	TiO_2	Al_2O_3	Fe_2O_3	FeO	MnO	MgO	CaO	Na_2O	K_2O
wt%	71.12	0.13	14.16	1.77	0.39	0.03	2.63	1.51	2.21	1.37

Experimental

Three specimens as a group were put into a container. The deionized-water was used as leachant, media/leachant=1:1, SA/V=20m-1. The leaching container set is same as that in reference[5].The experimental sets were put into the oven at 90 ℃; and lasted 91 d, 182 d, 364 d and 728 d, respectively.

RESULTS AND DISCUSSION

Chemical durability

When the designed experiment time reached, the containers were taken out from the oven, weighted the mass weight of the dried specimens. The mass loss rate was calculated according to the following equation:

$$LR_m = \frac{m_0 - m_t}{SA \cdot t}$$

LRm: the mass loss rate (g/(cm2·d)); mo: the mass of specimens before leach test(g); mt: the mass of specimens after leach test (g); SA: the surface area of the specimens(cm2); t: the leaching time(d). Figure 2 shows relation between the mass loss rate and the leaching time.

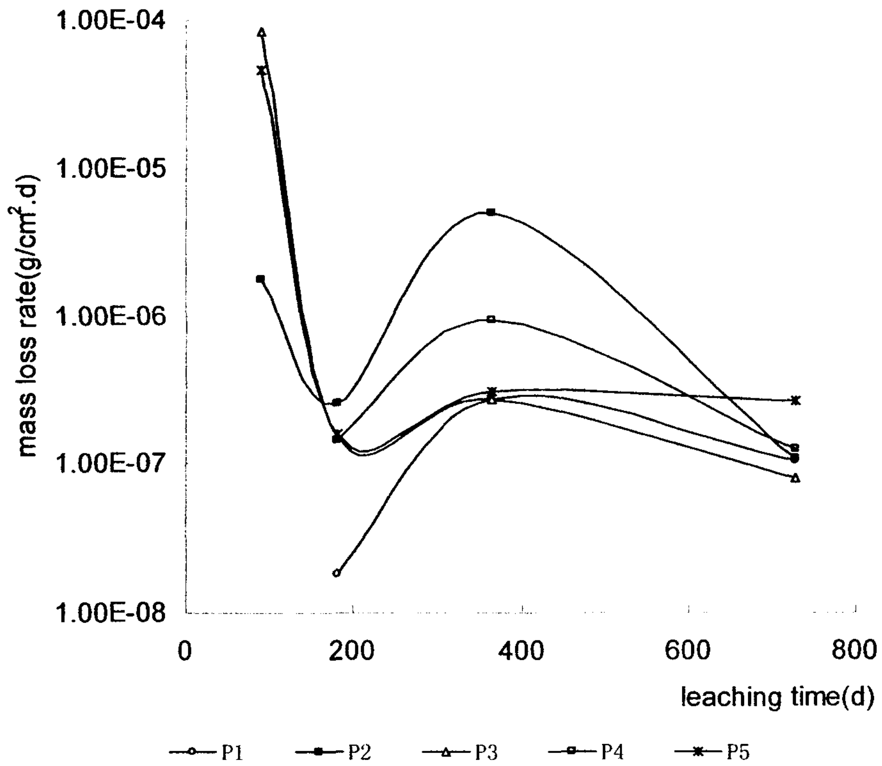

Fig.2. The dissolution rate of specimens at 90 ℃ for 728d

The Figure 2 shows (1) The specimens reached the leaching equilibrium after 182 days with 10-7 (g/cm2·d) of the mass loss rate, which suggests pyrochlore-rich synroc has a good chemical duriability; (2) The mass loss rate of system (P3, P5) with Fe_3O_4 were higher than other systems before 182 days, which indicates that Fe_3O_4 is disadvantage to durability of pyrochlore-rich synroc and causes the mass loss rate increasing.

XRD analyses of leached surfaces

The results show in fig.3~fig.7.

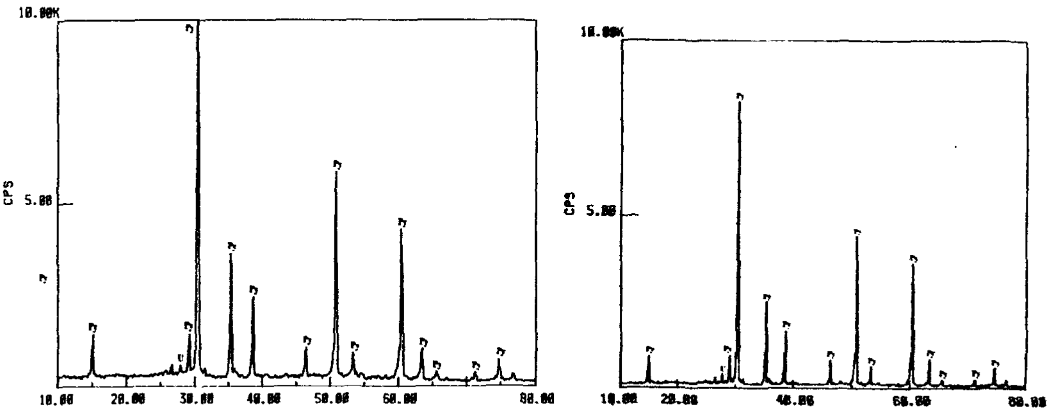

fig.3 Granite /(left:91d; right:728d)

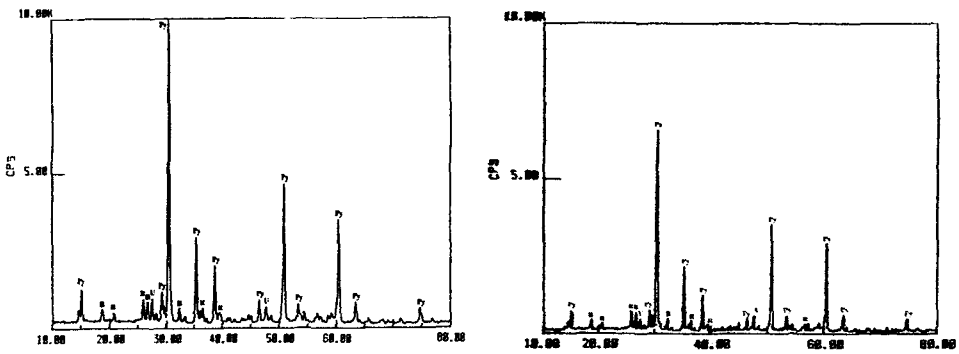

fig.4 Granite80%+cement20%/(left:91d ;right:728d)

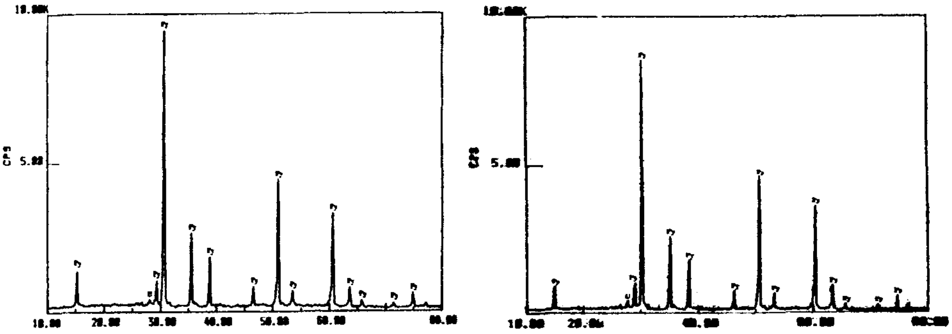

fig.5 Granite80%+cement20%/(left: 91d ; right:728d)

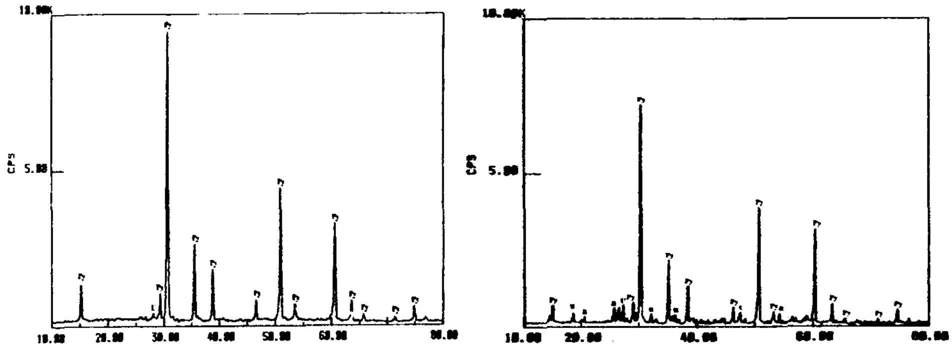

fig.6 Bentanite / (left:91d; right:728d)

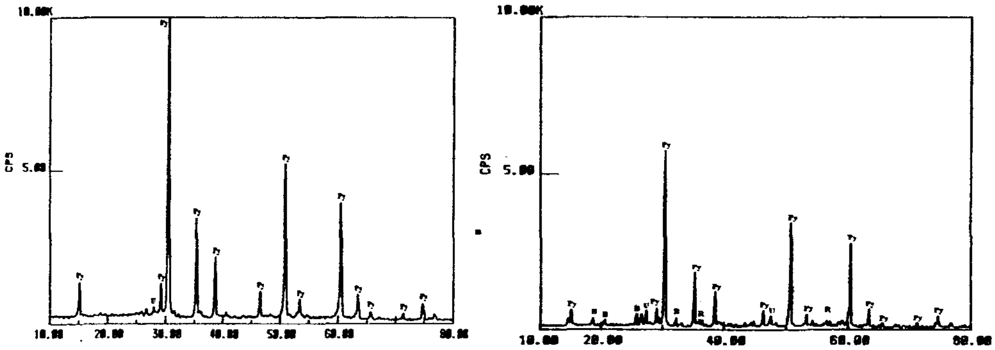

fig.7 bentanite 80%+ Fe3O420%/ (left:91d; right:728d)

(Py: pyrochlore synroc, U: crystal uranium mineral, B: betafite, R: rutile)

(1) The specimens (bentonite system P4, P5) had two new phase peaks (B, R) on the XRD pattern, and the intensity of new phase peaks were increased with test time, which may be

caused by some sediment formed on the sample surface;

(2) The specimens in P2 had the two new phase peaks on the pattern at the 91st day, but the intensity of the new phase peaks were not increased at the 728th day;

(3) The peak intensity of pyrochlore in P5 (bentonite80%+ Fe3O420%) is lower than that in P4 (bentonite100%), which suggests that Fe3O4 can promote the leaching behavior of synroc.

EDS analyses of leached surface analyses

The equipment used is the same as SEM/EDS analysis. The determined element (in atom %) on the specimen surface is shown in Fig.8. It indicted:

(1) U ion were enriched in the five systems during the test times, which is consisting with the crystal uranium mineral formed on the specimen surfaces found in the XRD pattern;

(2) Ti ion was depleted on the surfaces after leaching 728 days; the atom percent of Ti ion in P4, P5 and P2 systems were lower than that in P1 and P3 systems. Ti ion is most depleted in granite system (P1) among the five systems. The system with cement and bentonite can retard Ti ion leaching, and Fe3O4 had no an effect on Ti ion leaching;

(3) Ba ion was enriched on the surfaces after leaching 182 days, and there is an enrichment order P3>P1>P5>P4>P2 at leaching 728 days; Fe3O4 can enrich Ba ion on the specimen surfaces;

(4) Ca ion was enriched in bentnoite system P4, and most depleted in granite system P1 than other systems;

(5) The Zr ion was enriched on the specimen surfaces and had an enrichment order P3>P5>P4>P2>P1; Zr ion has an obviously enrichment in the systems with Fe3O4; bentonite and cement can speed the enrichment;

(6) Al ion was enriched in P1 and P2 systems at 728 days, and slightly depleted in P3, P4 and P5 systems; the media with bentonite and Fe_3O_4, can speed the depleted trend.

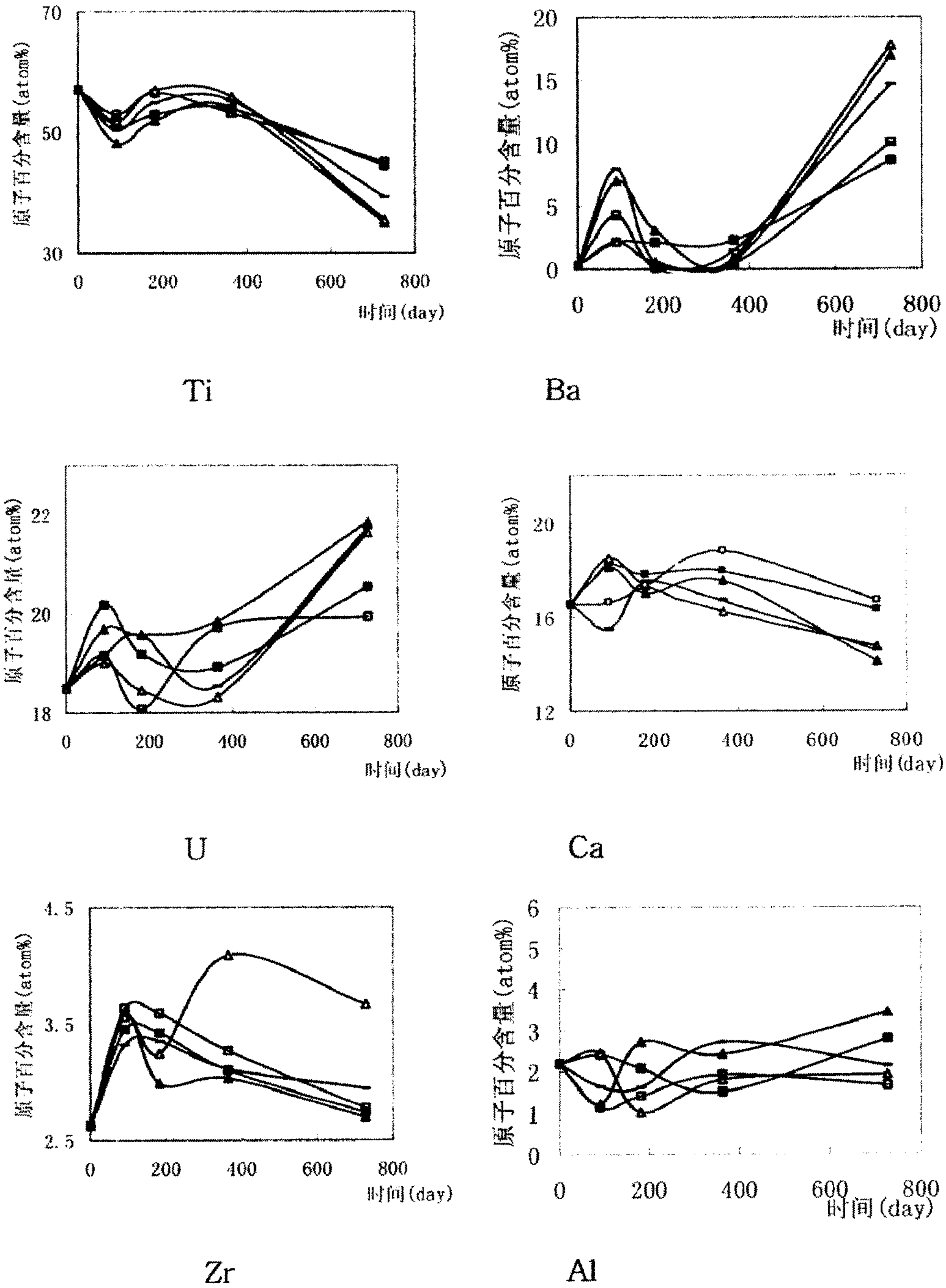

Fig.8. The EDS pattern of the element concentration on the leached surfaces at 90℃ for 728d

(P1▲— ,P2■— , P3△— ,P4□— ,P5—)

CONCLUSION

The pyrochlore-rich synroc loaded with 46.8% actinides waste were leached in simulated disposal condition at 90℃ for 728d, the results can be summarized in following:

The mass loss rate reached the leaching equilibrium after 182days with 10-7 g/cm2·d of the mass loss rate, the mass loss rate is veıy low and indicates that pyrochlore-rich synroc has good resistibility to leach;

The pattern of XRD showed two new peaks appeared on the leached surface in the system with bentonite and cement (P2, P4 and P5 systems).

The analytical results of EDS suggest that the system with bentonite and cement can speed the enrichment of Zr ion, and slow down the leaching of Ti ion. And in the five simulated disposal media, expect that Ti ion was depleted on the surface, U, Zr, Al and Ca ion were in equable states and Ba was enriched, which indicated the simulated disposal media can effectively retard the leaching behavior.

ACKNOWLEDGEMENT

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