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Original Article

Solidification of uranium tailings using alkali-activated slag mixed with natural zeolite

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ABSTRACT

Cemented uranium tailings backfill created from alkali-activated slag (CUTB) is an effective method of disposing of uranium tailings. Using some environmental functional minerals with ion exchange, adsorption, and solidification abilities as backfill modified materials may improve the leaching resistance of the CUTB. Natural zeolite, which has good ion exchange and adsorption characteristics, is selected as the backfill modified material, and it is added to the backfill materials with cementitious material proportions of 4%, 8%, 12%, and 16% to prepare CUTB mixtures with environmental functional minerals. After the addition of natural zeolite, the uniaxial compressive strength (UCS) of the CUTB decreases, but the leaching resistance of the CUTB increases. When the natural zeolite content is 12%, the UCS reaches the minimum value of 8.95 MPa, and the concentration of uranium in the leaching solution is 0.28–8.07 μg/L, the leaching rate R_{42} is 9.61×10^{-7} cm/d, and cumulative leaching fraction P_{42} is 8.53×10^{-4} cm, which shows that the alkali-activated slag cementitious material has a good curing effect on the CUTB, and the addition of environmental functional minerals helps to further improve the leaching resistance of the CUTB, but it reduces the UCS to an extent.

1. Introduction

Uranium tailings generally refer to the solid residue produced in the process of ore processing, hydrometallurgy, and heap leaching. They are a type of long-life and large volume low (extremely low) radioactive solid waste. Uranium tailings are mostly stacked by building reservoirs on the ground, which will inevitably cause certain environmental and safety problems [1,2]. However, the common dry-filling, hydraulic-filling, and even cemented filling methods do not fundamentally solve the problems of radioactive diffusion or the leaching of metal ions. Even so, techniques such as the cement solidification of medium and low-level radioactive wastes and the cemented backfilling of tailings containing heavy metals have revealed that stabilization and solidification techniques based on inorganic cementitious materials have a good effect on controlling the precipitation of radionuclides and heavy metals. If the problems of the poor mechanical properties and poor leaching resistance of cemented uranium tailings backfill in mines can be solved, underground backfill disposal will become the best method of the safe and efficient disposal of uranium tailings [3,4].

In the field of mine filling, the development of new cementitious materials based on solid waste is a hot issue. Numerous studies conducted by experts and scholars have shown that materials such as blast furnace slag, red mud, fly ash, and even acidic lead smelting slag can be prepared into cementitious materials. The preparation of cementitious materials from these mineral materials has a low cost, and these materials have a good water retention, corrosion resistance, and high later strength. In particular, alkali-activated binders have better performance than ordinary portland cement in immobilization of toxic, hazardous, or radioactive elements, which will greatly improve the application of cemented backfill in solid waste disposal. Zhang, Chen, and others have shown that alkali-activated slag can effectively solidify Cr (VI), and it can convert Cr(VI) into Cr(III) without any additional reductants [5,6]. Alonso showed that the fixation of alkali-excited materials to Pb exceeds 99.7%, and its immobilized performance exceeds that of OPC pastes [7], Abdel-Gawwad conducted the leaching experiment on the alkali-excited water cooled steel slug mixed with lead bearing slug, and the results showed that the lead concentration (mg/L) in the leaching solution was below the safe limit of toxicity characteristic leaching procedure (TCLP) [8]. Kiventerä showed that the alkali-activated GGBFS can effectively consolidate sulfites and

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heavy metals in gold mine tails [9]. Wang showed that alkali-activated slag can effectively reduce the leaching amount of heavy metals (Mn, Cr (VI) and Cr) and is lower than TCLP regulatory limit [10]. Jan, Yliniemi, Zhang, and others also have shown that alkali-activated binders are suitable materials for immobilization of heavy metals and have obvious advantages over ordinary portland cement [11,12]. Niels and Tian showed that alkali-activated cementitious materials have good curing effects on radionuclides such as Cs^+ and Sr^{2+} [13,14]. Komljenović also showed that alkali -activated blast furnace slag can not only be regarded as a potential effective substrate for caesium fixation, but also the addition of caesium can improve its strength [15]. Gijbels showed that alkali-activated materials based on ground blast furnace slag and phosphogypsum had good curing effects on ^{238}U, ^{226}Ra, ^{210}Pb and ^{228}Ra [16]. Zhou used alkali-activated coal gangue to immobilize uranium-contaminated soil, and showed that the maximum compressive strength of the solidified body reached 24.6 MPa, and the maximum fixation efficiency of uranium reached 77.44% [17]. Jiang showed that for the cement solidified form of uranium tailings, when slag powder is added, the leaching resistance of the solidified form is significantly improved. For the uranium tailings geopolymer with fiber, the solidification effect is optimal when 0.6% basalt fiber is added [18,19].

It can be seen that cemented uranium tailings backfill created from alkali-activated slag (CUTB) can theoretically reduce the adverse effects of the uranium tailings' characteristics on its formation and long-term stability. Different from the quality indexes of other metal tailings, the ultimate characteristic of the CUTB is its leaching resistance. Therefore, when considering adding certain modified materials to improve the fluidity of the filling slurry and mechanical properties of the filling body, we should consider whether these materials can improve the leaching resistance of the filling body to strengthen the ability of the filling body to retain uranium and other metal ions. Rudzionis showed that natural zeolite as an additive of portland cement can reduce the leaching of heavy metals, and its effect is better than that of synthetic zeolite with cement or cement alone [20,21]. Vyvail showed that Portland cement with natural zeolite has better curing effect on Ba, Cr, Cu, Ni and Pb [22]. Jimenez-Reyes reviewed the application of zeolites in radioactive waste and showed that zeolites have good adsorption performance for uranium [23]. If we choose environmental functional minerals such as natural zeolite as modifying materials, the leaching resistance of alkali activated slag and uranium tailings cemented backfill will be further improved.

To study the consolidation and immobilization effect of alkali-activated slag mixed with natural zeolite on uranium tailings, we prepared CUTB with specific mass concentration, specific cement to sand ratio, specific slag, specific alkali activation scheme and appropriate amount of natural zeolite. By testing the uniaxial compressive strength of CUTB and the concentration of uranium in the leaching solution, the influences of the characteristics of alkali-activated slag mixed with natural zeolite on the mechanical properties and leaching resistance of CUTB were analyzed.

2. Materials and methods

2.1. Experimental materials

The experimental materials used in this study mainly included uranium tailings, blast furnace slag, quicklime, liquid sodium silicate, sodium hydroxide, natural zeolite, and water.

The uranium tailings were obtained from a uranium tailings reservoir in China, with a water content of 10.91%, a density of 2.55 t/m^3, and a volume density of 1.375 t/m^3. The main chemical compositions of experimental materials obtained via X-ray fluorescence (XRF) analysis (PANalytical Axios) are presented in Table 1. The pH of the uranium tailings was 5.29 according to the *Soil Quality-Determination of pH* [24]. The particle size composition of the tailings was obtained using the screening method (Fig. 1). The particle size ranged from 45 to 425 μm, and the median particle size was 208 μm. The average particle size was close to the median particle size. The results show that the inhomogeneity coefficient was 2.31, the curvature coefficient was 0.93, the grain size distribution of the tailings was discontinuous and non-uniform, and the gradation was not good [3,25].

The blast furnace slag selected for the experiments was S95 slag micro powder, which was produced by a building materials enterprise, with a density of 2.86 g/cm^3 and a specific surface area of 431 m^2/kg. According to the chemical composition of the blast furnace slag, its chemical modulus was $K = 1.91$, its alkali modulus was $Mo = 0.97$, and its active coefficient was $Ma = 0.52$. The chemical modulus K of the slag was far greater than the index specified in GB/T 203-2008. The alkali modulus Mo was slightly less than the standard for neutral slag, which is acidic slag. The active coefficient Ma was much greater than the standard of 0.3, making it high activity slag [26–28]. The broad hump at $20°–35°(2\theta)$ of the XRD pattern of blast furnace slag indicates a typical amorphous structure (Fig. 2), and the crystalline form is mainly a small amount of calcite, dolomite and quartz, which indicates that blast furnace slag is mainly an amorphous glass body, which depolymerizes under the condition of alkali activator, which is also the reason for the high activity of blast furnace slag powder [29,30]. The quicklime was purchased from the market in the form of a white powder, with a specific surface area of 400 m^2/kg. According to the *Building Quicklime* (JC/T 479-2013), the $CaO + MgO$ content was 97.09% (i.e., >90.0%), and the MgO content was 2.31% (i.e., < 5.0%). SO_3 was not detected, which meets standard CL90-QP for powdery calcareous lime [31]. Liquid sodium silicate activator was prepared from liquid sodium silicate and sodium hydroxide purchased from the market. The natural zeolite was a 200-mesh micro powder purchased from the market, and its silicon aluminum ratio k was 5.22, which is in the medium silicalite range $k = 4–8$ [32]. According to the XRD analysis and comparison, it is found that the natural zeolite is mainly composed of clinoptilolite, and also contains quartz and other phases [33,34]. The water was urban tap water.

2.2. Experimental scheme

Non-equilibrium leaching is a common method in leaching resistance tests. It is generally realized via continuous flow of the leaching solution or the regular replacement of the leaching agent, which is also close to the environment of the underground filling body. According to the *Standard test method for the leachability of low and intermediate level solidified radioactive waste forms*(GB/T 7023–2011), non-equilibrium leaching is also used as the standard leaching method for solidified forms of low and medium level radioactive wastes. Referring to the two Chinese national standards (GB/T 7023–2011 and GB 14569.1–2011) [35,36], the tests mainly included the following steps:

1) **Preparation of CUTB.** First, according to the proportion determined in the test scheme (see Section 2.3) and referring to the *Standard testing method for the performance of building mortar* (JGJ/T70-2009), the filling materials were mechanically mixed using a JJ-5 cement mortar mixer. During the mixing, the material was mixed at low speed for 120 s, then suspended for 15 s, and then mixed at high-speed for 120 s to ensure that all of the materials were evenly mixed. Then, the prepared slurry was poured into $\Phi 50$ mm × 50 mm cylindrical molds, and the samples were vibrated manually to eliminate bubbles when

Table 1
Main chemical compositions of experimental materials.

	SiO_2	Al_2O_3	SO_3	K_2O	Fe_2O_3	Na_2O	CaO	TiO_2	P_2O_5	MnO	MgO	U
uranium tailings	86.06	6.04	2.43	2.37	1.55	0.51	0.38	0.21	0.13	——	0.10	0.04
blast furnace slag	31.86	16.53	0.04	0.54	0.43	0.33	39.81	1.23	——	0.18	6.89	——
natural zeolite	74.99	14.36	0.03	4.46	0.82	2.74	1.34	0.13	0.01	0.05	0.99	——

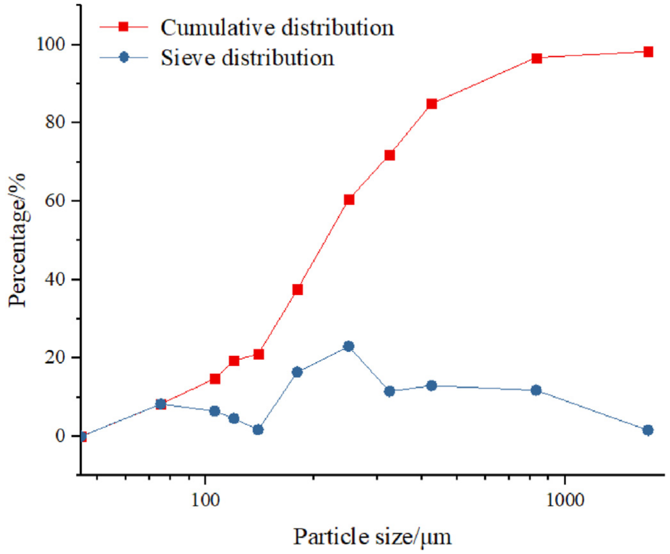


Fig. 1. Particle size accumulation curve for uranium tailings.

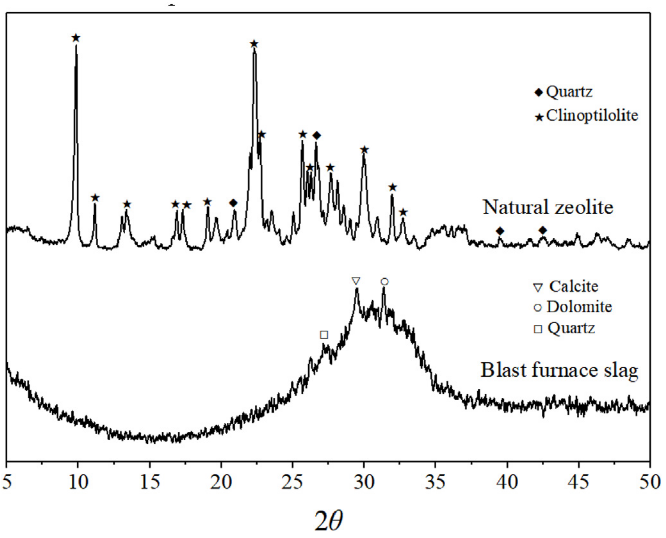


Fig. 2. XRD diagram of blast furnace slag and natural zeolite.

poured into the mold. Finally, the cast sample was placed in a curing box (25 ± 5°C, humidity $\geq$ 95%). It was removed from the mold after 24 h and allowed to continue curing for an additional 27 days (i.e., a total of 28 days of curing).

2) **Preparation for leaching.** First, the uniaxial compressive strength (UCS) test was carried out on the filling specimen cured for 28 days. If its strength was greater than or equal to 7 MPa, it was considered to meet the conditions for the leaching test, and its upper and lower end faces were polished. In addition, the height and diameter were measured to make sure its height to diameter ratio was $\geq$ 1 and its geometric surface area was 10–5000 cm^2. A polyethylene plastic wide mouth bottle with a cover was selected as the leaching container and was cleaned with deionized water for use after pickling. In addition, deionized water with a pH of 6.8–7.2 was selected as the leaching agent.
3) **Leaching test.** First, the prepared filling specimen was hung in the leaching container so that it was at least 1 cm from the container wall of the leaching container in all directions (Fig. 3). Second, according to the standard that the ratio of the volume of the leaching agent to the geometric surface area of the filling body test piece was 10, the geometric surface area of the filling body test piece was measured and a corresponding amount of deionized water (i.e., the leaching agent) was added. The container was covered and maintained at 25 ± 2°C for the static leaching test. The leaching agent was changed when the cumulative leaching time reached 24 h, 3 d, 7 d, 10 d, 14 d, 21 d, 28 d, 35 d, and 42 d, and an appropriate amount of leaching solution was taken out during the replacement process for testing. Finally, the concentration of uranium in the leaching solution was determined via inductively coupled plasma mass spectrometry (ICP-MS; 7700x), and the pH value of the leaching solution was determined using a pH meter.

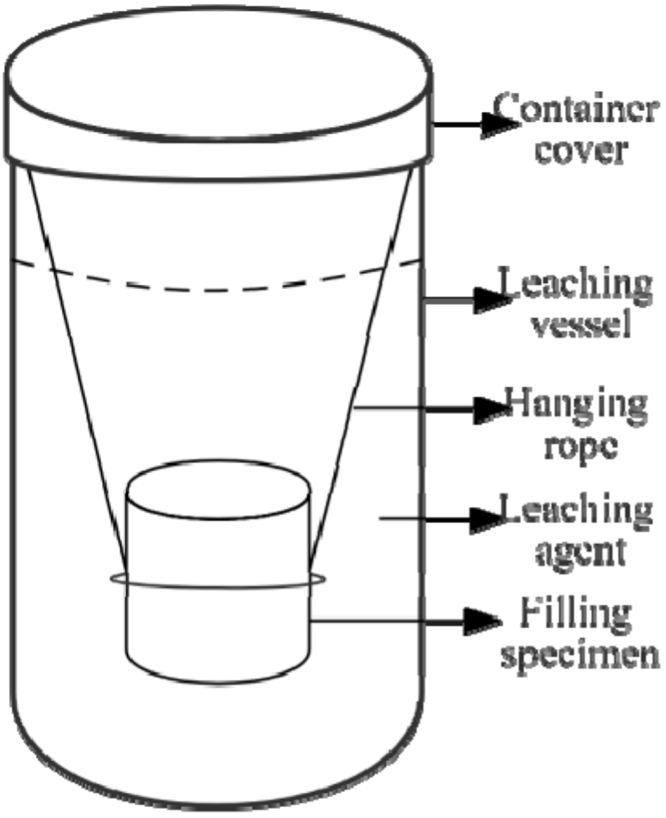


Fig. 3. Schematic diagram of the leaching experiment.

2.3. Test plan

To explore the influences of the characteristics of the natural zeolite on the leaching resistance of the CUTB, the other parameters were fixed, and only the types and additives of the environmental functional minerals were taken as variables. Considering the particularity of the use of uranium tailings as the filling aggregate, the water to solid ratio of the filling slurry was fixed at 0.25, and the cement to sand ratio was fixed at 1:4. Liquid sodium silicate was selected as the activator of the blast furnace slag. Its modulus was 1.2, the alkali equivalent of Na_2O accounted for 6% of the cementitious material, and 6% quicklime was added as the auxiliary material. The natural zeolite was mixed into the filler according to proportions of 4%, 8%, 12%, and 16% of cementitious materials for comprehensive testing. For comparison, a control group with no addition of functional minerals (i.e., 0%) was also set up.

3. Results and discussion

3.1. Mechanical properties of filling specimens to be leached

When the leaching resistance test was carried out on the cement solidified body of medium and low-level radioactive waste, the premise was that the UCS of the solidified body was greater than or equal to 7 MPa, and the strength range of the high-strength filling body in mine filling is only 4–5 MPa. For a filling body with a large amount of low or extremely low radioactive uranium tailings as the aggregate, the requirement of a UCS of greater than or equal to 7 MPa is inappropriate. Since there is no clear strength index requirement in the field of cemented filling of uranium tailings, the strength index before the leaching resistance test was temporarily set as 7 MPa.

According to the UCS tests (Fig. 4) on the CUTB after 28 days of curing, the strengths of the test groups and the control group were greater than 7 MPa, of which the UCS with 0% functional mineral was 13.45 MPa. The UCS of the CUTB with natural zeolite contents of 4–16% was 8.95–12.37 MPa, and the UCS initially decreased and then increased with increasing zeolite content. When the content was 12%, it reached the minimum UCS, and its strength was less than that of the control group, which indicates that the use of natural zeolite as a functional mineral has a weakening effect on the strength of the CUTB.

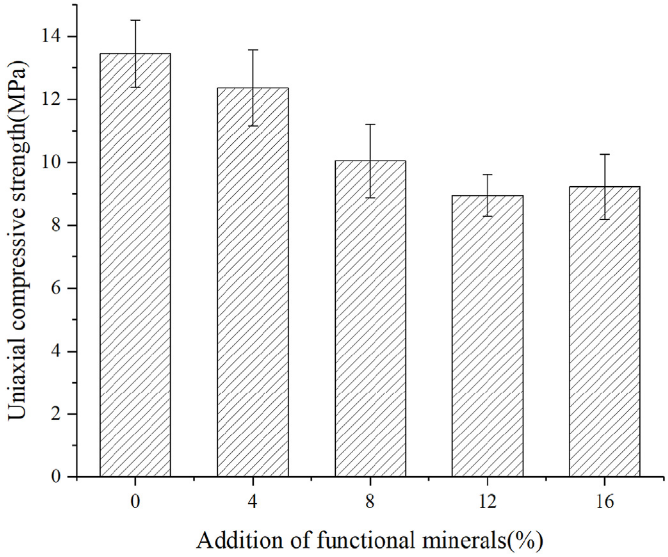


Fig. 4. USC of filling specimens mixed with natural zeolite (28 d).

The zeolite was mainly composed of SiO_2, Al_2O_3, CaO, and MgO, which is basically consistent with the chemical composition of blast furnace slag. Theoretically, the addition of zeolite should strengthen the performance of cementitious materials, but the weakening effect generally appears, which may be because the addition of these functional minerals improves the Q^3, Q^4, Q^4(1AL), and other high degree of polymerization products, but the content of low degree polymerization products is reduced and the micropores cannot be filled, which will make the structure of the filling body loose or cause the production of cracks, affecting the material's macro mechanical properties. Nevertheless, the strengths of the CUTB specimens were still much greater than the set standard of 7 MPa.

3.2. Effect of natural zeolite on uranium concentration in leaching solution

The concentration of uranium in the leaching solution is a direct index of the leaching resistance of the CUTB. If the concentration of uranium in the leaching solution is lower than the corresponding limit of the index, the water body similar to the leaching environment has reached the discharge standard. The non-equilibrium leaching method was adopted in this study, which is very similar to the flowing state of the groundwater environment in which the filling body would be located in mining applications. To more intuitively express the variation in the uranium concentration with increasing number of leaching cycles, we plotted the relationship in Fig. 5.

When the addition of zeolite was 0%, the uranium concentration of the leaching solution gradually decreased from 11.36 μg/L to 0.52 μg/L with leaching time. The relationship between these variables can be fitted and expressed using the Farazdaghi-Harris function (**Equation 1**) [37]. In addition, the curve can be divided into stages A, B, and C (Fig. 4), which are stages of rapid decrease, decelerating decrease, and stabilization of the uranium concentration, respectively, and the corresponding leaching times of the three stages are 1–14 d, 14–28 d, and 28–42 d. It can be seen that when the zeolite content was 4–16%, the uranium concentration of the leaching solution was lower than that of the control group

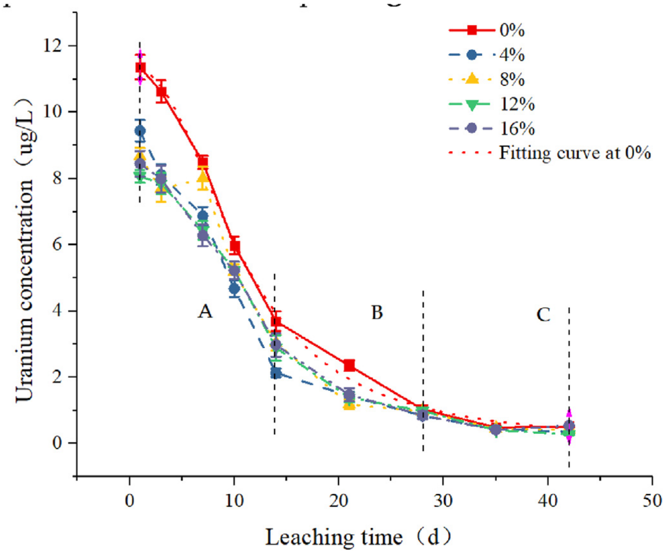


Fig. 5. Variation in the uranium concentration of the leaching solution with cumulative leaching time.

(content of 0%), especially in stage A (1–14 d). The uranium concentration of the leaching solution was much lower than that of the control group, but the difference decreased in stage C (28–42 d). Similarly, the variation in the uranium concentration of the leaching solution with increasing leaching time after the addition of zeolite can still be expressed using the Farazdaghi-Harris function, but after the addition of the functional minerals, the uranium concentration of the leaching solution decreased more significantly during 1–14 d, and the decreasing trend slowed down at 14–28 d. Generally, the influence of the zeolite content was not linear, that is, the uranium concentration of the leaching solution did not decrease linearly with increasing zeolite content.

$$Y = (a + b \times x^{c})^{-1}. \quad (1)$$

Where “Y” is the uranium concentration (μg/L), X is the leaching time (d), and a, b, c are constants. The values of a, b and c will change when the addition of natural zeolite are different.

The *Regulations for radiation protection and radiation environment protection in uranium mining and milling* (GB 23727–2020) stipulate that the uranium concentration at the wastewater discharge outlet shall not exceed 0.3 mg/L [38]. The uranium concentration of the uranium tailings filling body mixed with functional minerals was far lower than this limit within the tested number of leaching cycles, which indicates that alkali activated slag is an effective cementitious material for the creation of a solidified body of uranium tailings. The leaching resistance of the backfill was thus further improved.

3.3. Variations in the pH of the leaching solution with leaching time

The pH is the key factor affecting the leaching resistance of the CUTB, which directly affects the formation and stability of cementitious material products. Although the zeolite content of each test group was different, the activator modulus and alkali equivalent of each group of samples were consistent, and the pH values of the different groups of samples at different leaching times were relatively close. We used the average pH of the leaching solutions of the test groups with different contents to obtain the relationship between the average pH and the leaching time (Fig. 6).

It can be seen that the average pH value ranged from 10.70 to 11.14, and the pH of the leaching agent in each leaching cycle was about 7. This was due to the dissolution of the alkali activator in the filling system, and it changed slightly with increasing leaching time. The pH increased from 1 to 14 days, decreased from 14 to 28 days, and remained approximately stable from 28 to 42 days. This shows that the leaching test was always carried out in an alkaline environment. The alkaline environment of the leaching solution provides a good environment for the secondary polymerization of alkali-activated slag cementitious material, which results in the CUTB having good mechanical and anti-leaching properties.

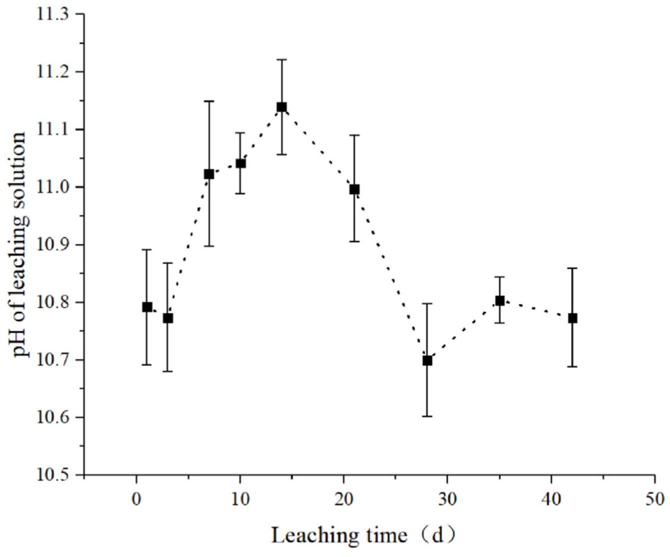


Fig. 6. pH values of leaching solutions of different functional minerals versus leaching time.

3.4. Leaching rate and cumulative fraction leached

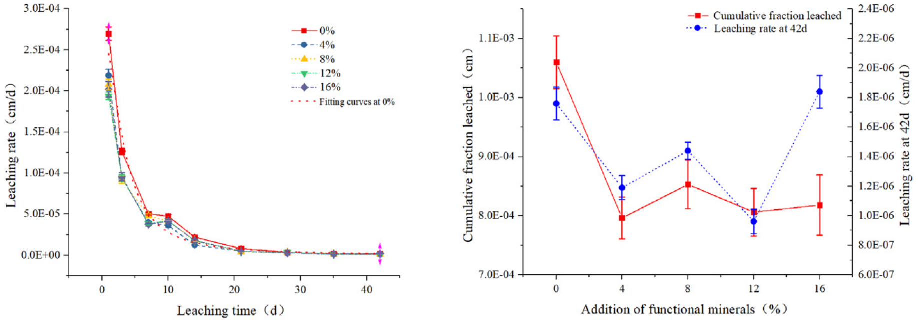
The leaching rate and cumulative fraction leached represent the leaching rate and cumulative amount of uranium leached in a certain leaching cycle, respectively. They consider not only the relationship with the initial value but also the relationship between the geometric surface contact area and the sample volume, so they can characterize the leaching resistance of the filling body better than the uranium concentration of the leaching solution. GB/T 7023–2011 [35] proposed the use of the leaching rate (R_{42}) and the cumulative fraction leached (P_{42}) at 42 d as a quality index of the leaching resistance and reported that R_{42} and P_{42} should be less than 1×10^{-5} cm/d and 0.17 cm, respectively. Therefore, the relationships between R_{42}, P_{42}, and the leaching time of each test group were analyzed (Fig. 7).

When the content of functional minerals was 0%, R_{42} and P_{42} were 1.76×10^{-6} cm/d and 1.06×10^{-3} cm, respectively, which are far below the limits specified above (i.e., by 1–2 orders of magnitude). The leaching rate R_{42} and cumulative fraction leached P_{42} when the zeolite content was 4–16% were less than those when the content of functional minerals was 0%, which indicates that the addition of functional minerals effectively strengthened the leaching resistance of the CUTB.

From the above experimental results, it was found that the CUTB with natural zeolite added as backfill modified material had good mechanical properties and leaching resistance, but the influences of the natural zeolite on these properties were quite different. Theoretically, the mechanical properties are the macroscopic characterization of the characteristics of the CUTB, which are affected by the products of the cementitious materials and the structure of the filling body, and the mechanical properties affect the leaching resistance to a certain extent.

The reason that the addition of natural zeolite reduced the strength of the CUTB and enhanced its leaching resistance can be analyzed from the perspective of its impact on the alkali-activated slag cementitious products. Akbarpour, Özen, and others showed that the increase of natural zeolite content will reduce the early strength of concrete or geopolymer, but will increase the late strength, and will also significantly reduce its rheological properties [39–41]. The addition of natural zeolite as backfill modified material may have led to an insufficient content of low polymer products in the cementitious products, which resulted in a loose filling structure and/or the development of cracks. In addition, due to the high concentration and high viscosity of the filling slurry, holes may have been formed by bubbles in the filling body due to improper vibration during the molding process. These factors may all have reduced the strength of the CUTB.

After adding natural zeolite, the leaching resistance of CUTB is effectively improved. Although there is little research on its impact on the leaching resistance of uranium tailings, Rudzionis showed that the compressive strength of cementitious materials will be reduced after adding natural zeolite, yet the adsorption capacity of heavy metals is high [20]. El-Eswed showed that the fixation of heavy metals in geopolymers may be due to the participation of



(a) Variation in leaching rate with leaching cycle

(b) Variation in cumulative fraction leached and leaching rate with zeolite content

Fig. 7. Relationships between the leaching rate, cumulative fraction leached at 42 d, and leaching time.

heavy metal cations in the balance of aluminum negative charges in unreacted zeolite and geopolymers phase frames [42]. Thomas showed that natural zeolite participates in the hydration reaction, which makes the structure of the solidified body thicker, thereby reducing the leachability of harmful substances [43]. It can be seen that the main reasons are the following aspects: First, the uranium tailings were fully wrapped by the alkali activated-slag cementitious material mixed with natural zeolite to form a relatively homogeneous and dense filling body, which further reduced the contact area between the uranium in the uranium tailings and the leaching agent, resulting in physical fixation. Second, the addition of natural zeolite to the filling material as backfill modified material encouraged the chain structure of the cementitious products to interweave and form a three-dimensional network structure. In addition, the part that does not participate in the reaction has the synergistic effects of separation, wrapping, and adsorption, which strengthen the uranium fixation effect. Moreover, it is also possible for uranium to enter the gelled products instead of some of the elements in the gelled products, and some of the insoluble precipitates formed also have a good uranium fixation effect.

These inferences and the experimental data show that in the process of consolidating uranium tailings via the fixation of uranium by alkali-activated slag mixed with natural zeolite not only produces physical fixation of the uranium, but it also has multiple effects, such as fixation, adsorption, exchange, and precipitation. In future research, it will be necessary to further analyze the mechanism by which the environmental functional mineral backfill modified materials influence the CUTB by studying the microstructure and phase evolution of the filling body.

4. Conclusions

In order to study the influences of the use of environmental functional minerals (i.e., natural zeolite) as backfill modified materials on the characteristics of the CUTB, CUTB samples formed under specific conditions were tested in this study. The UCS of the CUTB and the concentration of uranium in the leaching solution for samples with different natural zeolite contents were determined, and the following conclusions were drawn.

(1) When 4%, 8%, 12%, and 16% zeolite were added, the UCS of the CUTB decreased from 13.45 MPa to 12.37 MPa, 10.05 MPa, 8.95 MPa, and 9.22 MPa, respectively. This shows that the addition of natural zeolite weakened the UCS of the CUTB to a certain extent, and the weakening effect was the most obvious when the amount of zeolite addition was 12%. Nevertheless, the UCS of the CUTB was still higher than the requirement of 7 MPa for a cement solidified body of medium and low-level radioactive waste specified in GB 14569.1–2011. It is also greater than the 4–5 MPa specified for a high-strength filling body proposed in the manual of mining engineers.

(2) When 4%, 8%, 12%, and 16% natural zeolite were added, the maximum uranium concentration of the leaching solution of the CUTB decreased from 11.36 μg/L to 9.44 μg/L, 8.67 μg/L, 8.07 μg/L, and 8.45 μg/L, respectively, indicating that the addition of natural zeolite can effectively improve the leaching resistance of the CUTB, but its strengthening effect on the leaching resistance is not linear. When the zeolite content was 12%, the strengthening effect on the leaching resistance of the CUTB was the best.

(3) For uranium tailings, a type of long-life and large volume very low-level radioactive solid waste with special physical and chemical properties, cemented backfill with alkali-activated slag as the cementitious material is an effective disposal method. When natural zeolite is added as the backfill modified materials, its influences on the UCS and leaching resistance of the CUTB are inconsistent; that is, it has a weakening effect on the UCS and a strengthening effect on the leaching resistance.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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References

[1] World Nuclear Association, Uranium Mining Overview, 2020. https://www.world-nuclear.org/information-library/nuclear-fuel-cycle/mining-of-uranium/uranium-mining-overview.aspx.

[2] M. Ballini, C. Chautard, J. Nos, V. Phrommavanh, C. Beaucaire, C. Besancon, A. Boizard, M. Cathelineau, C. Peiffert, T. Vercouter, E. Vors, M. Descostes, A multi-scalar study of the long-term reactivity of uranium mill tailings from Bellezane site (France), J. Environ. Radioactiv. 218 (C) (2020), 106223.

[3] F.L. Wang, G.L. Chen, L. Ji, Z.P. Yuan, Preparation and mechanical properties of cemented uranium tailing backfill based on alkali-activated slag, Adv. Mater. Sci. Eng. (2020), https://doi.org/10.1155/2020/6345206.

[4] D. Debasis, T. Sreenivas, K.D. Gautam, P. Sandeep, Paste backfill technology: essential characteristics and assessment of its application for mill rejects of uranium ores, T. Indian Metals. 70 (2) (2017) 487–495.

[5] M.T. Zhang, C.H. Yang, M. Zhao, K. Yang, R. Shen, Y.J. Zheng, Immobilization

potential of Cr(VI) in sodium hydroxide activated slag pastes, J. Hazard. Mater. 321 (5) (2017) 281—289.

[6] H.X. Chen, H.H. Yuan, L.Q. Mao, M.Z. Hashmi, F.N. Xu, X.J. Tang, Stabilization/solidification of chromium-bearing electroplating sludge with alkali-activated slag binders, Chemosphere 240 (2020), 124885.

[7] M.M. Alonso, A. Pasko, C. Gascó, J.A. Suarez, O. Kovalchuk, P. Krivenko, F. Puertas, Radioactivity and Pb and Ni immobilization in SCM-bearing alkali-activated matrices, Constr. Build. Mater. 159 (2018) 745—754.

[8] H.A. Abdel-Gawwad, S.A. Mohamed, M.S. Mohammed, Recycling of slag and lead-bearing sludge in the cleaner production of alkali activated cement with high performance and microbial resistivity, J. Clean. Prod. 220 (2019) 568—580.

[9] J. Kiventerä, H. Sreenivasan, C. Cheeseman, P. Kinnunen, M. Illikainen, Immobilization of sulfates and heavy metals in gold mine tailings by sodium silicate and hydrated lime, J. Environ. Chem. Eng. 6 (5) (2018) 6530—6536.

[10] D.Q. Wang, Q. Wang, S.Y. Zhuang, J. Yang, Evaluation of alkali-activated blast furnace ferronickel slag as a cementitious material: reaction mechanism, engineering properties and leaching behaviors, Constr. Build. Mater. 188 (2018) 860—873.

[11] J. Yliniemi, J. Pesonen, M. Tiainen, M. Illikainen, Alkali activation of recovered fuel—biofuel fly ash from fluidised-bed combustion: stabilisation/solidification of heavy metals, Waste. Manage. 43 (5) (2015) 273—282.

[12] P.P. Zhang, F. Muhammad, L. Yu, M. Xia, H.R. Lin, X. Huang, B.Q. Jiao, Y.C. Shiau, D.W. Li, Self-cementation solidification of heavy metals in lead-zinc smelting slag through alkali-activated materials, Constr. Build. Mater. 249 (2020), 118756.

[13] V. Niels, I.R. Ion, C. Robert, S. Pieter, D. Jan, P. Yiannis, S. Sonja, S. Wouter, Alkali-activated materials for radionuclide immobilisation and the effect of precursor composition on Cs/Sr retention, J. Nucl. Mater. 510 (2018) 575—584.

[14] Q.Z. Tian, S. Nakama, K. Sasaki, Immobilization of cesium in fly ash-silica fume based geopolymers with different Si/Al molar ratios, Sci. Total. Environ. 687 (2019) 1127—1137.

[15] M. Komljenovic, G. Tanasijević, N. Dzunuzovic, J.L. Provis, Immobilization of cesium with alkali-activated blast furnace slag, J. Hazard. Mater. 388 (2020), 121765-121765.

[16] K. Gijbels, S. Landsberger, P. Samyn, R.I. Iacobescu, Yiannis Pontikes, Sonja Schreurs, W. Schroeyers, Radiological and non-radiological leaching assessment of alkali-activated materials containing ground granulated blast furnace slag and phosphogypsum, Sci. Total Environ. 660 (2019) 1098—1107.

[17] S.K. Zhou, J.L. Li, L.S. Rong, J. Xiao, Y.J. Liu, Y. Duan, L.P. Chu, Q. Li, L. Yang, Immobilization of uranium soils with alkali-activated coal gangue-based geopolymer, J. Radioanal. Nucl. Chem. 329 (2021) 1155—1166.

[18] F.L. Jiang, J.T. Guo, X.L. Wang, Y. Liu, X.Y. Li, G. Chen, Z. Wang, J. Yang, B. Tan, Experimental study on the leaching performance of U(VI) solidified by uranium tailing cement with different admixtures and ratios, Environ. Technol. Inno 17 (2020), 100506.

[19] F.L. Jiang, B. Tan, Z. Wang, Y. Liu, Y.Y. Hao, C. Zhang, H.N. Wu, C.S. Hong, Preparation and related properties of geopolymer solidified uranium tailings bodies with various fibers and fiber content, Environ. Sci. .Pollut. R 331 (6) (2022) 2761—2777.

[20] Z. Rudzionis, S.K. Adhikary, F.C. Manhanga, D.K. Ashish, R. Ivanauskas, G. Stelmokaitis, A.A. Navickas, Natural zeolite powder in cementitious composites and its application as heavy metal absorbents, J. Build. Eng. 43 (2021), 103085.

[21] C. Napia, T. Sinsiri, J. Chai, P. Chindaprasirt, Leaching of heavy metals from solidified waste using Portland cement and zeolite as a binder, Waste. Manage. 32 (2012) 1459—1467.

[22] M. Vyvail, P. Bayer, Immobilization of heavy metals in natural zeolite-blended cement pastes, Procedia. Eng. 151 (2016) 162—169.

[23] M. Jimenez-Reyes, P.T. Almazan-Sanchez, M. Solache-Ríos, Radioactive waste treatments by using zeolites. A short review, J. Environ. Radioactiv 233 (2021), 106610.

[24] International Organization for Standardization, ISO10390:2005, Soil quality-Determination of pH, Geneva, Switzerland, 2005.

[25] A.X. Wu, H.J. Wang, Theory and Technology of Paste Filling in Metal Mines, Science Press, Beijing, China, 2015.

[26] Standardization Administration of China, GB/T 203-2008: Granulated Blast Furnace Slag Used for Cement Production, 2008. Beijing, China.

[27] H.S. Shi, Eco-cement and Waste Recycling Technology, Chemical Industry Press, Beijing, China, 2005.

[28] Z.S. Lin, Slag Base Serise Eco-Cement, China Building Materials Press, Beijing, China, 2018.

[29] L.J. Vilaplana, J.F. Baeza, O. Galao, G.E. Alcocel, E. Zornoza, Mechanical properties of alkali activated blast furnace slag pastes reinforced with carbon fibers, Constr. Build. Mater. 116 (2016) 63—71.

[30] Q. Liu, J.K. Zhang, Y.W. Su, X.J. Lu, Variation in polymerization degree of C-A-S-H gels and its role in strength development of alkali-activated slag binders, J. Wuhan. Univ. Technol. 36 (6) (2021) 871—879.

[31] Ministry of Industry and Information Technology of the People's Republic of China, JC/T 479-2013, Building quicklime, Beijing, China, 2013.

[32] M. Valaskova, J. Tokarsky, M. Hundakova, J. Zdralkova, B. Smetana, Role of vermiculite and zirconium-vermiculite on the formation of zircon-cordierite nanocomposites, Appl. Clay. Sci. 75/76 (2013) 100—108.

[33] C. Wang, W.J. Li, S.Z. Leng, H.D. Guo, J. Lu, X.J. Zhang, Y. Zhao, Z.P. Ye, Mineralogical characteristics and adsorption property of natural zeolite from Weichang area，Hebei Provence, Acta Petrol. Mineral 38 (6) (2019) 753—760.

[34] K.X. Zhu, J.S. Yuan, Z.H. Yu, Y.P. Fu, L. Zhang, Study on the physical properties and ion exchange performance of the domestic clinoptilolite, Ind. Miner. Process. (7) (2009) 19—23.

[35] Standardization Administration of China, GB/T 7023-2011, Standard Test Method for Leachability of Low and Intermediate Level Solidified Radioactive Waste Forms Beijing, China, 2011.

[36] Ministry of Ecology and Environment of the People's Republic of China, GB 14569.1-2011: Performance Requirements for Low and Intermediate Level Radioactive Waste Form - Cemented Waste Form, 2011. Beijing, China.

[37] L.P. Hu, H. Gao, Nonlinear Regression Analysis and and Their Intelligent Ralization by SAS, Publishing House of Electronics Industry, Beijing, China, 2013.

[38] Ministry of Ecology and Environment of the People's Republic of China, GB 23727—2020: Regulations for Radiation Protection and Radiation Environment Protection in Uranium Mining and Milling, 2020. Beijing, China.

[39] A. Akbarpour, M. Mahdikhani, R.Z. Moayed, Effects of natural zeolite and sulfate ions on the mechanical properties and microstructure of plastic concrete, Front. Struct. Civ. Eng. 16 (1) (2022) 13.

[40] A. Akbarpour, M. Mahdikhani, R.Z. Moayed, Mechanical behavior and permeability of plastic concrete containing natural zeolite under triaxial and uniaxial compression, J. Mater. Civil. Eng. 34 (2) (2022), https://doi.org/10.1061/(ASCE)MT.1943-5533.0004093.

[41] S. Özen, B. Uzal, Effect of characteristics of natural zeolites on their geopolymerization, Case. Stud. Constr. Mat 15 (2021), https://doi.org/10.1016/j.cscm.2021.e00715.

[42] B.I. El-Eswed, R.I. Yousef, M. Alshaaer, I. Hamadneh, S.I. Al-Gharabli, F. Khalili, Stabilization/solidification of heavy metals in kaolin/zeolite based geopolymers, Int. J. Miner. Process. 137 (2015) 34—42.

[43] M. Thomas, M. Osinska, A. Slosarczyk, Long-term behavior of cement mortars based on municipal solid waste slag and natural zeolite—a comprehensive physico-mechanical, structural and chemical assessment, Materials 15 (3) (2022), https://doi.org/10.3390/ma15031001.