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Simulation on the effect of supercritical carbon on Cu ions leaching behavior in cement solidification body

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Abstract

Residue of city garbage disposal such as fly ash contains lots of harmful substances such as heavy metals, which need to be treated before landfill or recycling utilization. The modification effect of supercritical accelerated carbonation process on cement solidification body (CSB), make up for the deficiency of large porosity. To study the effect of supercritical carbonation on leaching behavior of heavy metal ions in the CSB, the numerical simulations were performed on the leaching rate and leaching distribution pattern of Cu ion. The results showed that supercritical carbonation reduced the leaching rate of Cu ion in the CSB, which was more obvious when leaching time was shorter than 14 days. The start time was brought forward on stable cumulative leaching concentration of Cu ions, due to the Carbonization process. Furthermore, it was found that the distribution of leaching rate on the CSB section was alternative positive and negative. The supercritical carbonation increased the scope of the outermost positive region layer, and changed the plane distribution of the Cu leaching rate in the CSB, which made contribution to the inhibition of heavy metal ion leaching.

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Keywords: Supercritical carbonization; solidification; heavy metal; leaching simulation;

1. Introduction

The garbage incineration technology gets more and more application due to its harmless, reduction and recycling. However, lots of fly ash was produced through waste incineration, which may be harmful to the environment and

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society. The fly ash is hazardous waste for containing higher heavy metals and dioxins. Usually, harmless process is ahead before landfill disposal, for example by cement solidification processing into the CSB. However, the fixed effect of ordinary the CSB is not ideal due to the larger porosity.

To improve the solidification performance of fly ash in CSB, the accelerated carbonation technology adopted to modify of the solidified body. The solidified effect could be largely improved due to the change of chemical state of hydration after carbonization [1]. It is good for curing performance of the CSB using carbon dioxide, with the strength improved at 28 days age, the porosity decrease and leaching rate of harmful substances reduced [2]. Until now, many researches had done on the accelerated carbonization affected certain metal leaching rate in the CSB [3]. It is different for different metal ions on the solidification in the CSB, according to the leaching property of Pb $\$ Cd $\$ As and Cr [4]. The supercritical carbonization technology was also applied on solidification of radioactive waste in the CSB, the results showed that pore structure was changed, free liquid content was decreased, and radionuclide leaching resistance was improved [5,6]. Related studies showed that accelerated carbonation treatment could effectively improve the solidification effect of heavy metal in the CSB.

On the numerical analysis for accelerate carbonization and leaching behavior on the CSB contains heavy metals, there were many beneficial exploration had been performed. A simulation of leaching behavior of Pb, Cd, As and Cr in cement waste had established based on the chemical dynamics and thermodynamic equilibrium function of cement –based material [7]. The simulation had finished the study on the diffusion of Ca, Mg, AL, Pb ions in the CSB considering without, part or complete carbonization [8].

The simulation on cement hydration process was studied using geochemical software GEMS - PSI, and analyzed the temperature influence on the hydration product and porosity [9]. It could provide a reference for establishing hydration model, to simulate the early hydration reaction considering the carbonization. The long time diffusion performance of heavy metal ions were also simulated, combined phase equilibrium, surface adsorption and multicomponent diffusion module, considering the migration and exchange of ion in concrete [10].

Related researches show that after supercritical carbonization, the porosity structure of CSB changes, free liquid content decreases, stabilization of heavy metals improves. As one treatment technology, the accelerate carbonization technology had caused the attention by many scholars, but still not enough [11-13]. It needs to further-study the accelerated carbonation influence on leaching action and mechanism of heavy metal ions. There is also little study on the distribution of leaching rate. This article will focus on the leaching rate of Cu ion in the CSB with and without carbonization.

2. Simulation Model

2.1. Numerical model information

In order to study solidification technology and modification performance of the CSB, Our group established carbonization numerical model using software COMSOL, based on cement-base material under carbonization. The model contains chemical reaction rate equation, mass conservation equation, momentum conservation equation and energy conservation equation, mass transfer of gas liquid two phase and heat transfer [14].

The leaching function of model will use a diffusion analysis model, finished by Wang [15], considering the carbonization and leaching behavior. This model considers function of ion diffusion, adsorption, bond coefficient and thermal diffusion. Langmuir equation was adopted as isothermal adsorption nonlinear simulation of ion. The ion coefficient was used to simulate solidification by ion chemical reaction, which caused the reduction of ion concentration in the original body. As internal ion migration mainly through diffusion behavior, this model uses the nonequilibrium heat and mass transfer mode. The effective diffusion coefficient was adopted in this model to reflect the diffusion behavior of ion migration.

2.2. Model verification

According to the test conditions on materials and reaction, adjusted related parameters, other model parameters was the same as the original model [14]. The leaching rate of Cu ion in the CSB with and without carbonization, then compared with the test using leachate at 25 °C, as shown in figure 1.

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