

碱矿渣—粘土复合胶凝材料固化Sr、Cs的机理与性能研究

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Research on Mechanism and Properties of Immobilizing Sr and Cs Based on Alkali-Activated Slag-Clay Minerals Composite Cement

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论文单位 中国工程物理研究院,点击次数 10,论文页数 133页File Size9064k

2005-04-20 [论文网](#) http://www.lw23.com/lunwen_402437427/ 碱矿渣-粘土复合凝胶材料;Sr;Cs;固化;机理;性能

Alkali-activated slag-clay minerals composite cement; Strontium; Cesium; Immobilization; Mechanism; Property

放射性核素Sr、Cs的固化是核技术应用的关键环节之一。碱矿渣-粘土复合胶凝材料(AASCM)是具有低钙硅比(C/S)、富铝、富钠组成特征并放成了碱矿渣水泥的高强度、低孔隙率、较好抗侵蚀性与改性凹凸棒石粘土(M-ATT)、沸石(ZEO)良好吸附性能的一种新型放射性核素Sr、Cs的固化基材。研究其固化Sr、Cs的机理和性能,旨在为该材料的组成设计及工程应用提供理论依据和积累实验数据。应用静态吸附法,研究了改性凹凸棒石粘土、沸石等矿物材料吸附Sr、Cs的性能。结果表明:粘土等矿物材料的结构及其中所具有的阳离子的种类决定着它们吸附Sr、Cs的能力。结构中存在低价态取代高价态阳离子(如Al~(3+)取代Si~(4+)或Mg~(2+)取代Al~(3+))的同晶取代现象,形成取代结构吸附活性中心是使其具有较强吸附能力的前提。结构中的电荷数少,离子半径小的阳离子(如Na~+),其更易被交换,对提高其吸附Sr、Cs的能力有利。结构中存在微孔隙发育,具有一些通道(如沸石和凹凸棒石粘土),对吸附小于这些孔道直径的阳离子有益。层状结构的粘土矿物中,取代结构吸附活性中心与阳离子之间的电荷平衡方式为远程中和方式时,有利于阳离子的交换吸附。沸石、改性凹凸棒石粘土具有较强的吸附和选择性吸附Sr、Cs的能力,其中沸石类矿物吸附Sr的能力较强,而改性凹凸棒石粘土吸附Cs的能力较强。运用吸附-逐步浸出试验方法进行了AASCM的配方优化。沸石、改性凹凸棒石粘土及热活化高岭土与碱矿渣水泥复合构成的AASCM水化物的吸附性及在去离子水中抗浸出性较碱矿渣水泥高。在实验条件下,AASCM的组成(HAK/GBS)对其吸附-逐步浸出性能有较大影响。当HAK/GBS为0.176-0.25时,AASCM水化物粉体吸附Sr较好;当HAK/GBS为0.25时吸附Cs最佳。综合组成材料配比对碱矿渣-粘土复合胶凝材料水化物粉体的吸附-浸出及强度性能的影响,较为理想的材料配方为HAK:ZEO:M-ATT:GBS=15:5:10:70(此时HAK/GBS=0.214)。该配方的AASCM水化产物的平均CaO/SiO₂(C/S)为0.80, (CaO+Na₂O)/(SiO₂+Al₂O₃)[(C+N)/(S+A)]为0.75,Al₂O₃/SiO₂(A/S)为0.24。AASCM水化物粉体较之硅酸盐水泥(PC)、铝酸盐水泥(AC)、矿渣硅酸盐水泥(PSC)具有更强的对Sr、Cs的吸附性能。该配方的AASCM净浆28d的抗压强度达到80MPa以上,长期强度稳定。运用XRD、TG-DSC、FTIR、SEM等技术研究了AASCM的水化产物及SrO-CaO-SiO₂-H₂O的水热合成产物。结果表明:热活化高岭土的引入,实现了AASCM水化产物的富铝改性,使AASCM的水化产物具有低C/S、富铝、富钠特性。常温下,AASCM水化产物主要是(Na+Al)-C-S-H,可能存在结晶程度较差且极为分散的沸石类产物;80℃条件下,除了生成(Na+Al)-C-S-H之外,还存在钙十字沸石及菱沸石等水化产物。在80

Immobilization of radioactive nuclide like Sr and Cs is one of the important keys in the application of nuclear technology. Alkali-activated slag-clay minerals composite cement (AASCM) is a new matrix for solidifying Sr and Cs which has characteristics of low C/S, Na and Al enrichment and integrates high strength, low porosity, better resistance to corrosion of alkali-activated slag and good cation exchange adsorption of modified attapulgite(M-ATT) and zeolite(ZEO). The research on mechanism and properties of immobilizing Sr and Cs based on AASCM aims at obtaining theoretical basis and accumulating experimental data for its composition design and engineering application. The adsorption capacity of Sr and Cs in several kind of minerals such as ATT, ZEO etc. were studied by static adsorption method. The results reveal that the adsorption capacity of Sr and Cs are dependent on the structure and sorts of cation of the minerals. The substituted-structural adsorption centers (SSAC) formed from substitution of high-valency cations by low-valency cations (such as Si~(4+) by Al~(3+) or Al~(3+) by Mg~(2+)) in the structure of the are a prerequisite to the high adsorption capacity of the minerals. The cations with low-valency and small radius (such as Na~+) in the structure are more easily exchangeable and are more helpful to the adsorption capacity of Sr and Cs in the minerals. The channels in the structure of the minerals such as ZEO and ATT are favorable for adsorbing cation whose diameter is smaller than that of the channels. When the interaction of SSAC and the cations is long-range neutralization in the charge compensation way, the layer-structural clay minerals have higher cations exchange capacity. The adsorption capacity of Sr in ZEO group minerals is higher than that of Cs while the adsorption capacity of Sr in M-ATT is lower than that of Cs. The adsorption & step-by-step leaching experimental method was applied to the formula optimization of AASCM. The adsorption capacity and resistance to leaching in deionized water of AASCM which is integrated with ZEO, M-ATT, heat-activated kaolinite (HAK) and alkali-activated slag cement(AAS) are higher than AAS. Under the experimental conditions, the formula of AASCM has significant effects on adsorption & step-by-step leaching properties. When HAK/GBS ratio is in range of 0.176-0.25 in AASCM, the adsorption capacity of Sr is higher than at any other ratio while that of Cs at a HAK/GBS ratio of 0.25 is the highest in all investigated HAK/GBS ratios. In consideration of adsorption & step-by-step leaching and strength properties, the suggested formula HAK:ZEO: M-ATT:GBS ratio of AASCM =15:5:10:70 (HAK/GBS ratio is 0.214). The average C/S is 0.80 in the AASCM with suggested formulation while (C+N)/(S+A) is 0.75 and A/S is 0.24. The adsorption capacity of Sr and Cs in hydrated paste power of AASCM is higher than that of PC, AC and PSC. The 28-day compressive strength of the hardened AASCM paste is more than 80MPa and its long-term compressive strength of AASCM does not decrease. The hydration products of AASCM, the hydrothermally synthesized products of SrO-CaO-SiO₂-H₂O systems were studied by means of XRD, TG-DSC, FTIR and SEM. The results indicate that AASCM have characteristics of low C/S, Al and Na enrichment by introducing HAK. The major hydration products of AASCM cured at room temperatures are considered to be Na and Al substituted calcium silicate hydrate gels [(Na+Al)-C-S-H] and poorly-crystallized and well-dispersive products featuring zeolite may exist. Besides (Na+Al)-C-S-H, there are phillipsite-Ca and chabazite in the hardened AASCM cured at 80 ° C. Under the hydrothermal conditions at 80 ° C, when SrO reacts with CaO and SiC₂>2 which are elemental compositions of cements, calcium-strontium silicate hydrate solid solution can be produced and the structure of C-S-H gel will be more disorderly. The static adsorption behaviors of Sr and Cs in hydrated paste powder of AASCM were firstly investigated. The relationship between the adsorption capacity of Sr and Cs in the hydrotherm

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