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水力压裂处置中镎、钚迁移行为的模拟研究

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摘要 研究了在我国拟实施水力压裂处置某地的元素Np、Pu在钻孔地下水中的存在形式及地下水与围岩(页岩)、水泥固化体之间的相互作用。用地球化学模式程序EQ3/6模拟计算结果表明:1)在处置区地下水中,Np的存在形式主要为NpO2(CO3)34-、NpO2(CO3)23-、NpO2CO3-和NpO2(CO3)35-,Pu为PuO2(CO3)2-2和Pu(OH)5-;2)水岩作用模拟结果表明,处置地点的天然地下水中各组分间的相互作用已达平衡;3)地下水 水泥固化体相互作用过程大体上可划分为两个阶段,第一阶段为起始后的前10d,反应进程快,第二阶段为10d后,反应进程减缓,并在71d达到两相间的平衡,反应10d后,因碱性物质不断溶解及积累导致体系pH快速增长;4)在水泥固化体中,Am、Np和Pu基本上处于固定状态,只有当水泥固化体被溶解和破坏后,被破坏部分的水泥固化体中的核素方被释放进入水中。当地下水与水泥固化体之间的相互作用达到平衡时,地下水中Am、Np和Pu的放射性活度分别为12 66、1 405、132 48Bq。

关键词 水力压裂处置 镎 钚 存在形式 水岩作用

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Migration of Neptunium and Plutonium in Hydrofracturing Disposal

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Abstract Neptunium and plutonium are two important elements in middle radioactive wastes for the present study. The speciations of the elements in groundwater and the interactions between water shale, water cement in the potential area of hydrofracturing disposal of China are simulated by using geochemical modeling code EQ3/6. The simulated results are as follows:1) The main species are NpO 2(CO 3) 3 4- , NpO 2(CO 3) 2 3- , NpO 2CO 3 - and NpO 2(CO 3) 3 5- for Np, and PuO 2(CO 3) 2- 2 and Pu(OH) 5 - for Pu, even some of them rarely occurs in the case of nature and experiment;2) The equilibrium of interaction among species of groundwater in this potential areas of China have been reached;3) The water cement interaction process can divided into two stages, the fast stage with a high reaction rate during the initiative 10 d , and consequential slow stage 10 d later with a increased pH values due to the dissolved and deposited of the alkaline substance, the equilibrium of interaction between groundwater and cement have reached after 71 d;4) Both Np and Pu are almost immobile in the cement but they would release into the groundwater when the cement dissolved and damaged. The calculated activities of Am, Np, Pu in the equilibrium aqueous of water cement are 12.66, 1.405, 132.48 Bq,respectively.

Key words hydrofracturing disposal Np Pu speciation water-rock interaction

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