Contribution ID: **84** Type: 口头报告

氚在高温气冷堆典型材料中输运行为研究进展

摘要

氚作为核能系统中关键的放射性核素，其输运行为对反应堆辐射安全与先进核能系统发展至关重要。针对高温气冷堆（HTGRs）中氚迁移特性与安全挑战，本文系统研究了氚在碳化硅、核级石墨及合金材料中的扩散机制及其抑制策略。研究发现：1）碳化硅表面区域对氚的强滞留效应显著降低其有效扩散系数，复合层扩散模型成功解释了理论与实验的差异；2）通过热解吸实验与密度泛函理论计算，揭示了核级石墨中四个氚解吸峰分别对应基面、非晶表面、扶手椅状边缘和之字形边界的解吸行为，阐明了氚在辐照石墨边缘富集的微观机制；3）基于氢同位素（H/D/T）在 Fe、Cr、W 中的扩散规律分析，验证了高温条件下扩散系数与同位素质量的平方根反比关系，并建立了多元合金（如 2.25Cr1Mo 钢）中氚扩散系数的组分体积分数预测模型；4）结合高温气冷堆示范工程（HTR-PM）实测数据，揭示了氧化膜对氚渗透的抑制效应，提出通过优化材料表面化学状态与掺杂元素降低氚渗透率的工程策略。研究成果为完善 HTGRs 氚源项平衡模型、提升辐射安全水平及核能制氢技术发展提供了理论支撑与材料设计指导。

关键词

高温气冷堆；氚；扩散系数；第一性原理

Abstract

Tritium, as a critical radionuclide in nuclear systems, plays a pivotal role in reactor radiation safety and the advancement of advanced nuclear technologies. Focusing on the migration characteristics and safety challenges of tritium in High-Temperature Gas-cooled Reactors (HTGRs), this study systematically investigates the diffusion mechanisms of tritium in silicon carbide (SiC), nuclear-grade graphite, and alloy materials, along with strategies for permeation suppression. Key findings include: 1) The strong retention effect of tritium at SiC surface regions significantly reduces its effective diffusion coefficient, and a composite-layer diffusion model successfully reconciles theoretical and experimental discrepancies; 2) Thermal desorption experiments combined with Density Functional Theory (DFT) calculations reveal that four characteristic tritium desorption peaks in nuclear-grade graphite correspond to desorption from basal planes, amorphous surfaces, armchair edges, and zigzag boundaries, elucidating the microscopic origin of tritium enrichment at irradiated graphite edges; 3) Analysis of hydrogen isotope (H/D/T) diffusion in Fe, Cr, and W validates the inverse square root relationship between diffusion coefficients and isotopic mass under high temperatures (>800 K), and establishes a component volume fraction-based prediction model for tritium diffusion in multicomponent alloys (e.g., 2.25Cr1Mo steel); 4) By integrating operational data from the HTR-PM demonstration project, the inhibitory effect of oxide films on tritium permeation is demonstrated, with proposed engineering strategies to reduce permeation rates through surface chemistry optimization and elemental doping. These findings provide theoretical foundations and material design guidelines for refining tritium source-term models, enhancing radiation safety, and advancing nuclear hydrogen production technologies in HTGRs.

Keywords

High temperature gas-cooled reactors; tritium; diffusion coefficient; first-principle

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Session Classification: 环、化、材、技、能源战略

Track Classification: 03 口头报告: 环、化、材、技、能源战略