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LARGE SCALE SIMULATION OF NUCLEAR WASTE MATERIALS

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Abstract

Computer-aided simulations are valuable research tools for investigation of the properties of nuclear materials at atomic scale. This is because in principle, comparing with the experimental techniques, any system could be computed, including the experimentally challenging radiotoxic materials, and the only limitation is the availability and performance of the supercomputing resources and the approximate character of computational methods. Here we present an overview of our research activities on atomistic simulations of materials related to nuclear waste management. We discuss various structural, chemical, energetic, thermodynamic and radiation damage resistance properties of phosphate-based ceramic waste forms and nuclear graphite. Emphasis is put on selecting a reliable computational methodology. Our atomistic modeling effort complements the relevant experimental studies. We demonstrate that the combined atomistic modeling and experimental studies result in superior characterization of the investigated nuclear materials.

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1. Introduction

Safe management of nuclear waste is an important issue and strategies to deal with this problem are discussed in nuclear technology utilizing countries [1]. One of the widely accepted approaches is the conditioning of the waste by, for instance, immobilization of radionuclides in ceramic waste forms [2], and final deep geological disposal. Designing a durable nuclear waste disposal form requires in-depth characterization and understanding of the target

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materials, such as monazite-type ceramics [3]. The materials properties of interest include the chemical and thermodynamic stability, the radiation damage resistance, the dissolution behavior and the corrosion resistance. Because many of these materials properties cannot be easily measured, if at all, the reliable atomistic modeling represents an important research tool in this field [4].

Because of the tremendous increase in the availability of the computing power, that rises by a factor of at least 100 per decade [5], it is now possible to perform large scale simulations of properties of chemically and structurally complex materials using *ab initio* (up to a few hundred atoms) and the force-field methods of computational chemistry and materials science [6]. In the atomistic modeling group in the IEK-6 part of the Institute of Energy and Climate Research at Forschungszentrum Jülich (Research Centre Jülich) we perform large scale atomistic simulations of various properties of nuclear waste related materials, including ceramic waste forms such as monazite-, xenotime- or pyrochlore-type ceramics, spent nuclear fuel (UO_2-based) and irradiated graphite. The investigated problems include: a reliable and feasible computational methodology for strongly correlated *f*-electrons systems [7,8,9], the structural [7], the elastic [10] and the thermodynamic materials properties [10,11,12,13], the radiation damage resistance [14,15], the surface chemistry [14,16] and the diffusion and energetics of defect formation [16,17], to name but a few. Here we will discuss in details the results of selected research activities. These include investigation of: the reliable *ab initio* computational approaches, the properties of monazite- and xenotime-type ceramic waste forms and irradiated graphite. In particular, we report new results on the elastic properties and the thermal conductivity of xenotime-type ($LnPO_4$) phosphates. These studies represent a scientific basis for nuclear waste management and the discussed results have been published in various scientific journals as indicated through the text.

2. Method

2.1 Ab initio methods

Density functional theory (DFT) is currently the most commonly used *ab initio* method [6]. This is because of its feasibility and good scaling with the number of atoms comparing to other methods of computational quantum chemistry (such as the post Hartree-Fock methods like hybrid DFT functional, MP2 or CCSD(T)). However, because DFT uses approximations which originate from the homogeneous electron gas, it usually has problems when applied to computation of strongly correlated and localized *d*- and *f*- electrons-containing systems such as lanthanide and actinide-bearing materials that constitute nuclear waste. Among other shortcommings, it overestimates the reaction enthalpies (by as much as 100 kJ/mol) [18,8] and fails even on the qualitative level, predicting metallic state for simple wide band gap actinide-oxides insulators (UO_2, NpO_2 or PuO_2) [19]. We thus utilize the DFT+*U* method, which treats the electronic correlations with the Hubbard model. In this model the strength of the Coulomb on-site repulsion, which causes the electronic correlation on the strongly localized *d* and *f* orbitals, is represented by the so-called Hubbard *U* parameter, which value is usually guessed or sometimes estimated from the sparsely available experimental data. In order to make the method a parameter free approach we derive the Hubbard *U* parameter value using recently developed *ab initio* methods such as the linear response (or constrained local density approximation, cLDA) [20] or the constrained random phase approximation (cRPA) [21]. In our research we use Quantum-ESPRESSO [22], CPMD [23] and FLEUR DFT [24] codes. The core electrons are represented by the ultrasoft pseudopotentials [25] and the plane-waves energy cutoff is set to 50 Ryd. More details on the computational setup are provided in the relevant publications.

2.2 Force fields methods

Because *ab initio* simulations are currently limited to no more than a few hundred atoms, large scale atomistic simulations involving thousands or millions of atoms are performed using force field methods in which the inter-atomic interactions are represented by an applied force field that is usually designed to reproduce some experimentally measured or computed *ab initio* materials properties. In our studies we use simple Buckingham-type force fields which parameters are fitted so the force field reproduces the *ab initio* data computed with the methodology described in the previous section. To perform these simulations we use GULP [26] and LAMMPS [27] simulation packages. The details on the fitting procedure and force fields parameters are provided by Ji et al. [14].

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