



Effect of irradiation on silicate aggregates' density and stiffness

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ABSTRACT

When exposed to high-energy radiations, the silicate aggregates used in concrete can exhibit some swelling. This, in turn, can result in some internal stress, which can lead to micro-cracks or de-bonding along the cement–aggregate interface. However, there is presently some uncertainty regarding the long-term effect of such high-energy irradiation on aggregates and the associated risk of cracking and failure. Here, based on atomistic simulations, we investigate the effect of neutron irradiation on eight silicate minerals belonging to three different mineralogical families. We demonstrate that the irradiation-induced alterations do not depend on the silicate family but rather on the inherent structure and composition of each mineral. Interestingly, we show that vitrification can be used as a surrogate to assess the upper limit of irradiation-induced swelling. Further, we observe that this swelling potentially leads to high internal stresses, which can result in the initiation and propagation of cracks. These findings can guide the selection of optimal aggregate minerals for which the risk of concrete failure upon irradiation is minimized.

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1. Introduction

Silicate minerals form approximately 90% of the Earth's crust [1]. Due to their abundance, these minerals are widely used in applications involving exposure to nuclear radiation, such as the aggregates for nuclear power plants concrete [2–4] and for the immobilization of low-level nuclear waste [5,6]. In structural concrete used for biological shields [7], aggregate minerals constitute approximately 70% (by volume) and form a major component of the load-bearing structure. As such, developing a fundamental understanding of the effect of irradiation on the mechanical properties of silicate minerals is essential to predict and improve the service life of concrete structures.

When exposed to radiations, silicate minerals can undergo some atomic scale disordering, which can ultimately lead to the

amorphization of their structure—a mechanism that is also known as metamictization [8,9]. Metamictization, in many cases, is associated with a volumetric expansion of the mineral, which is found to be as high as 18% in quartz [10]. In addition, metamictization causes changes in the mechanical properties as well, such as the elastic modulus and strength of these mineral [11]. It has been observed that various silicate minerals can exhibit distinct behavior upon irradiation depending on their specific structure and composition [2–4]. The differential volumetric expansions of these minerals under confined conditions (e.g., in concrete), can impose some severe stress on the surrounding matrix, which can ultimately lead to the formation of micro-cracks or macroscopic fracture [4,7,12]. While extensive studies have been conducted on some archetypical silicate minerals such as quartz [10,11,13–15] and zircon [8,16–19], the effect of irradiation on the mechanical properties of more complex silicate minerals remains largely unknown [2–4,20]. As such, this gap in the understanding of the effects of irradiation on aggregate minerals raises serious concerns regarding the integrity of concrete in nuclear environments.

Based on the connectivity of the silicate groups within the

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atomic network, silicate minerals can be classified into different groups, such as nesosilicates, inosilicates, phyllosilicates, and tectosilicates, among others [1]. Most experimental studies on irradiation have focused on the critical amorphization dosage and the resulting dimensional change of aggregate-forming silicate minerals [2,21,22]. However, these studies do not provide a comprehensive mechanistic understanding of irradiation-induced degradation and its effect on the engineering properties of silicate aggregates. Further, the analysis of these experimental results is complicated by the following issues. (i) Mineral samples can contain impurities, which can affect the experimental observations. (ii) Assessing the upper limit of irradiation-induced damage—i.e., after complete metamictization—requires long durations of controlled exposure to radiation. In many cases, it is challenging to ensure that the material has indeed reached its saturated state. (iii) Upon irradiation, mineral samples can exhibit micro-cracks or fracture [3,21], thereby leading to an overestimation of their swelling. (iv) Experiments are highly sensitive to ambient conditions such as temperature [14] and boundary conditions, which renders difficult a meaningful comparison between different experimental results. All these factors make it extremely challenging to do a systematic study on the effect of radiation on silicate minerals belonging to different families.

Molecular dynamics (MD) simulations provide an alternate route to study radiation damage in minerals [18,23–26]. When properly conducted and validated, MD simulations are extremely useful as they provide a direct access to the details of the dynamics of the atoms in irradiated minerals, which are usually invisible to conventional experiments. This can yield an improved understanding of the fundamental mechanisms governing irradiation-induced disordering in minerals. Herein, we systematically analyze the irradiation-induced structural disordering and swelling in eight silicate minerals—almandine, diopside, enstatite, jadeite, albite, anorthite, nepheline, and quartz—belonging to different groups. In each case, structural data are used to elucidate the mechanism of irradiation-induced swelling, if any, which is then compared to that induced upon vitrification. Finally, we use the outcomes of the MD simulations to assess the magnitude of the stress developed in minerals upon irradiation to estimate the risk of cracking.

2. Methodology

2.1. Crystal simulation

Eight different silicate minerals, belonging to three different mineralogical families—nesosilicate, inosilicate, and tectosilicate [1]—are chosen for the present study. These minerals include almandine (nesosilicate, garnet group), diopside, enstatite, jadeite (single-chain inosilicates, pyroxene group), albite (tectosilicate, feldspar group, plagioclase and alkali-feldspar end-member), anorthite (tectosilicate, feldspar group, plagioclase end-member), nepheline (tectosilicate, feldspathoid group), and quartz (tectosilicate). The structures of these minerals are explained below. Each of these minerals is initially relaxed in the isothermal–isobaric ensemble (*NPT*) at 300 K and zero pressure for 100 ps. This ensures that the crystalline structure is in equilibrium and free of any initial stress. The simulation is continued for another 20 ps to obtain the average structural and thermodynamic properties of the equilibrium structure.

2.1.1. Nesosilicates: almandine ($Fe_3Al_2Si_3O_{12}$)

Nesosilicate minerals consist of isolated silicate tetrahedra $[SiO_4]^{4-}$ that are connected by interstitial cations to form a three-dimensional structure [27]. Garnet and zircon groups form some

of the major sub-categories of the nesosilicates—extensive studies on the effect of radiation on zircon can be found in the literature [8,16–19,28]. Here, we focus on the mineral, almandine ($Fe_3Al_2Si_3O_{12}$). Almandine has a cubic crystal structure with Si and Al atoms in tetrahedral and octahedral coordination states, respectively [27]. The mineral represents the ferrous end-member of the garnet series with the Fe atoms exhibiting a nominal charge of 2+. Almandine is a dense mineral with a mass density close to 4.2 g/cm³.

2.1.2. Inosilicates: diopside ($MgCaSi_2O_6$), enstatite ($MgSiO_3$), and jadeite ($NaAlSi_2O_6$)

Inosilicate minerals consist of silicate tetrahedra that are connected to form a chain-like structure [29–31]. Both single- and double-chained structures are found in inosilicates. Pyroxenes (single-chain) and amphiboles (double-chain) represent the major sub-categories of inosilicates. Here, we consider three minerals belonging to the pyroxene group—jadeite ($NaAlSi_2O_6$), enstatite ($MgSiO_3$), and diopside ($MgCaSi_2O_6$).

Diopside is a pyroxene with a monoclinic crystal structure [30]. O atoms exist as both bridging oxygen (BO), i.e., forming Si–O–Si connections between the SiO_4 tetrahedra, and non-bridging oxygen (NBO) atoms that are charge-balanced by Mg and Ca cations. Enstatite is the magnesium end-member of the enstatite–ferrosilite series [29]. Enstatite exhibits an orthorhombic crystal structure comprising some BO and NBOs atoms, charge-balanced by Mg cations in the latter case. Jadeite is an aluminosilicate mineral exhibiting a monoclinic crystal structure, with Na atoms acting as charge-balancing cations [31]. The Al atoms present in jadeite show an octahedral coordination state, forming six bonds with the surrounding O atoms, whereas Si atoms show a tetrahedral environment. Note that all the minerals considered here for this family exhibit high densities, namely, 3.02 g/cm³ for enstatite, 3.10 g/cm³ for jadeite, and 3.20 g/cm³ for diopside.

2.1.3. Tectosilicates: albite ($NaAlSi_3O_8$), anorthite ($CaAl_2Si_2O_8$), nepheline ($NaAlSiO_4$), and quartz (SiO_2)

Tectosilicate minerals form nearly three-fourths of the minerals in the Earth's crust [1]. These minerals are aluminosilicates—with the exception of the quartz group—and are characterized by a three-dimensional silicate network made of Si tetrahedra. Here, we analyze the irradiation response of four tectosilicate minerals—albite ($NaAlSi_3O_8$), anorthite ($CaAl_2Si_2O_8$), nepheline ($NaAlSiO_4$), and quartz (SiO_2).

Albite is the sodium end-member of the plagioclase feldspar [32,33]. It exhibits a triclinic crystal structure made of tetrahedral four-fold coordinated Si and Al atoms. Na atoms act as the charge-compensating cation to balance the charge of the AlO_4 units and all the O atoms exist as BOs. Anorthite is the calcium end-member of the plagioclase feldspar [34]. This mineral, similarly to albite, exhibits a triclinic structure made of four-fold Si and Al tetrahedra. Ca atoms act as charge-compensating cations for the AlO_4 units and all the O atoms exist as BOs. Nepheline belongs to the feldspathoid family and shows a hexagonal crystal structure [35]. Similarly, to the other tectosilicates, Si and Al atoms are found in a tetrahedral state and all O atoms form BOs. Finally, quartz is an archetypical silicate mineral with a three-dimensional network formed of interconnected Si tetrahedra [36]. Quartz exhibits different polymorphs depending on the temperature and pressure conditions. Here, we consider α -quartz, which shows a trigonal crystal structure.

2.2. Neutron-irradiation simulation

MD simulations of irradiation-induced damage in minerals are here carried out using the open-source LAMMPS [37] package,

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