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Original Article

INTERPARTICLE POTENTIAL OF 10 NANOMETER TITANIUM NANOPARTICLES IN LIQUID SODIUM: THEORETICAL APPROACH

SOO JAE KIM [a], GUNYEOP PARK [a], HYUN SUN PARK [b,*], MOO HWAN KIM [c], and JEHYUN BAEK [a]

[a] Department of Mechanical Engineering, POSTECH, 77 Cheongam-Ro, Nam-Gu, Pohang, Gyungbuk 37673, Republic of Korea
[b] Division of Advanced Nuclear Engineering, POSTECH, 77 Cheongam-Ro, Nam-Gu, Pohang, Gyungbuk 37673, Republic of Korea
[c] Korea Institute of Nuclear Safety, 62 Gwahak-Ro, Yuseong-Gu, Daejeon 34142, Republic of Korea

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ABSTRACT

A suspension of titanium nanoparticles (Ti NPs) in liquid sodium (Na) has been proposed as a method to mitigate the violent sodium–water reaction (SWR). The interparticle potential between Ti NPs in liquid Na may play a significant role in the agglomeration of NPs on the reaction surface and in the bulk liquid Na, since the potential contributes to a reduction in the long-term dispersion stability. For the effective control of the SWR with NPs, a physical understanding of the molecular dynamics of NPs in liquid Na is key. Therefore in this study, the nonretarded Van der Waals model and the solvation potential model are employed to analyze the interparticle potential. The *ab initio* calculation reveals that a strong repulsive force driven by the solvation potential exceeds the interparticle attraction and predicts the agglomeration energy required for two 10-nm Ti NPs to be 4×10^{-17} J. The collision theory suggests that Ti NPs can be effective suppressors of the SWR due to the high energy barrier that prevents significant agglomeration of Ti NPs in quiescent liquid Na.

1. Introduction

Sodium (Na) is an excellent coolant as it has extremely high thermal conductivity, low neutron flux, low meting temperature, and high boiling temperature. Therefore, in the development of the next-generation nuclear reactor design, Na-coolant-based reactors have become one of the key design concepts in the GEN-IV program [1]. However, it is well known that when Na comes into direct contact with water or moist air, a sodium–water reaction (SWR) occurs. This violent exothermic reaction rapidly releases a large amount of heat, gases including explosive H_2, and corrosive NaOH. In 1995, a

* *Corresponding author.*
E-mail address: hejsunny@postech.ac.kr (H.S. Park).

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Na leakage accident occurred at the Monju reactor in Japan. In the accident, Na leaked from a pipe and reacted with water causing a fire intense enough to warp steel structures. The lesson learned from the accident was that the elimination of SWR risk should be one of the most significant design criteria for the development of safe sodium fast cooled reactors.

When a leakage occurs in the Na and water heat exchanger, the pressurized water in the secondary loop intrudes into liquid Na, causing an energetic SWR. Therefore, development of technologies to prevent or mitigate SWR becomes a key issue in the fast cooled reactor design. One method involves isolating Na from water by employing a double-walled piping system and guard vessels [1–4]. This technology, however, cannot be an ultimate solution, since the probability of failure of the system still exists. An alternative method is to reduce or eliminate SWR. In this area, recent studies [5–11] suggested that a small quantity of nanoparticles (NPs) suspended in Na reduced the reaction rate between Na and water. The studies showed that the hydrogen production rate and reaction heat generation during the SWR were reduced when titanium (Ti) NPs with 2 at% and 0.214 vol % for 10 nm and <100 nm, respectively, were mixed in the liquid Na. However, it is necessary to further understand the mechanism.

The strong attraction between Na atoms and Ti NP surfaces reduces SWR reactivity [6,7,11], increases liquid Na surface tension by 16%, and reduces its evaporation rate by 18% [6]. Dispersed NPs constrain the movement of many nearby Na atoms, which therefore cannot easily react with H_2O. The SWR can only occur where NPs are not present; this observation indicates that the NPs reduce the effective area of the reaction surface [12], and that areal density of NPs (NPs/m^2) at the Na/H_2O interface is a key parameter that affects the effectiveness of NPs to suppress reactivity.

If NPs are well dispersed and evenly hold Na atoms, as illustrated in Fig. 1A, the reactivity would be reduced effectively. However, when NPs agglomerate into clusters over time, the effective area of the reaction surface increases, thus lowering the effectiveness of NPs to reduce SWR reactivity (Fig. 1B). For this reason, the effectiveness of NPs in mitigating SWR can be determined by the agglomeration behavior of NPs at the Na/H_2O interface over time. Unfortunately, experimental observation of NPs in bulk liquid Na and the reaction surface is difficult, since SWR is very fast and liquid Na is visually opaque. Therefore, in this study, the agglomeration behavior of NPs was analytically studied and theoretically discussed by employing the non-retarded Van der Waals and solvation potential models to evaluate the interparticle potential between 10-nm Ti NPs in liquid Na. In this paper, the computational methodology for the theoretical analysis is described in the second section, the analytical result from the computation is discussed in the third section, and the final conclusion of the study will follow.

2. Materials and methods

2.1. *Computation method*

In order to understand the behavior of colloidal particles in a liquid medium, the force between the particles should be known. Many types of forces act on NPs in liquids; a force can be categorized by its origins as a Van der Waals, an electrostatic, a solvation, an entropic, or a nonequilibrium force [13]. Van der Waals, a relatively weak force, always exists, and it can be attractive or repulsive with respect to the interparticle distance. An electrostatic force, a relatively strong force, arises when a particle is polarized. Coulomb, hydrogen bonding, and electric double-layer forces are typical examples of electrostatic forces. If the charged particles are in a vacuum, Coulomb force will act; however, in nature, this is a rare case. In most cases, neutral colloidal particles are in a specific solvent such as water. Particles in a solvent are polarized due to the ions that are absorbed from the solvent; the electric double-layer force arises due to the oppositely charged ions surrounding the particles. The electric double-layer force is a repulsive force and its strength is proportional to the ion concentration. In the past, most studies have used Van der Waals and electric double-layer forces in order to depict colloidal particle dispersion and stability; this approach is named the Derjaguin and Landau, Verwey and Overbeek (DLVO) theory [14,15]. The DLVO theory predicts the behavior of colloidal particles in typical electrolytes reasonably well when the particles are separated by tenths of a nanometer. However, when the particle distance is lower than a Debye

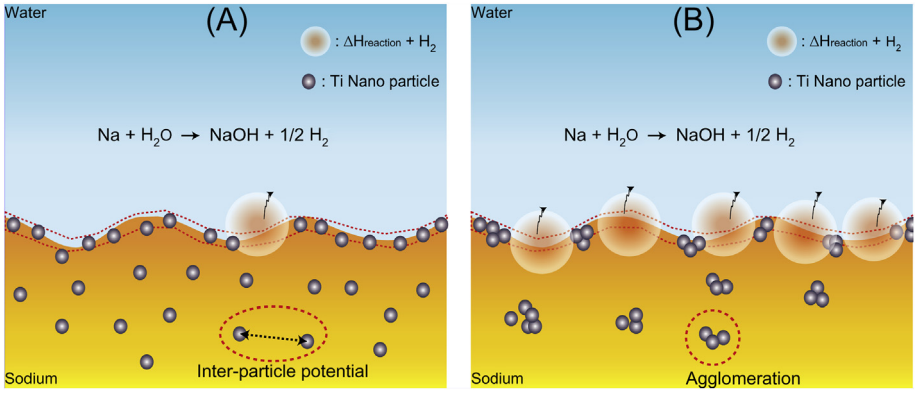

Fig. 1 – Effect of suspended Ti nanoparticles on the sodium–water reaction. (A) Well-dispersed state and (B) agglomerated state.