Retention and Recycling of Deuterium in Liquid Lithium-Tin Slab Studied by First-Principles Molecular Dynamics

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Abstract

Understanding the retention and recycling of hydrogen isotopes in liquid metal plasma-facing materials such as liquid Li, Sn, and Li-Sn are of fundamental importance in designing magnetically confined fusion reactors. We perform first-principles molecules dynamics simulations of liquid Li-Sn slab with inserted D atoms to provide microscopic insights into the interactions of D with Li-Sn liquid metal. We observe evaporation of D\textsubscript{2} and LiD molecules out of the Li-Sn slabs. With detailed analysis, we unveil a cooperative process of forming D\textsubscript{2} molecules in liquid Li-Sn, where Li atoms act as catalytic centers to trap a D atom before another D comes nearby to form a molecule, and the surplus charges are transferred from D\textsubscript{2} to nearby Sn atoms. Furthermore, we predict a temperature window in which D\textsubscript{2} molecules can escape to vacuum, while LiD molecules cannot. The above findings deepen our understanding of interactions between hydrogen isotopes and Li-Sn liquid metal.

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I. INTRODUCTION

Plasma-facing components (PFCs) in magnetic confinement fusion devices are designed to withstand high heat loads of bombardments from energetic particles. Meanwhile, hydrogen (H) isotopes such as deuterium (D) and tritium (T) are utilized as fuels and their interactions with PFCs need to be thoroughly understood. In particular, the retention and recycling of D/T attract many attentions due to their impacts on the fueling and vacuum pumping systems, and also the plasma conditions. Here, retention and recycling respectively refer to the hydrogen isotopes that remain in PFCs and return back to plasma after the PFCs receive particle bombardments. On one hand, a high retention rate of hydrogen isotopes suggests that the temperature of core plasma is less affected by preventing cold atoms from reentering the plasma, therefore, the plasma performance could be increased with PFCs that can retain a large amount of D/T atoms. On the other hand, a high recycling rate of H isotopes can significantly reduce the fuel cost, and a low retention of D/T atoms in PFCs also reduces safety concerns.

The emergence of liquid-metal PFCs began receiving widespread attention in recent years. Unlike solid metallic PFCs that unavoidably suffer from mechanical failure problems caused by high energetic particles, liquid metals for PFCs own a low melting point, resist erosion and neutron damage to a large extent, and has high heat dissipation capabilities. For instance, lithium (Li) has a low melting point of 453 K and was found to improve the plasma parameters in a variety of experiments. Tin (Sn) has also been tested in several experiments due to its low melting point of 505 K and additionally owns a low vapor pressure that is beneficial for high operating temperatures. Nevertheless, plasma contamination due to accumulation of impurities atoms in plasma is concerned since Sn has a high atomic number. In this regard, Li-Sn eutectic is considered as an alternative candidate for PFCs with combined advantages of both Li and Sn. However, the Li-Sn alloy has been investigated in a few experiments. Therefore, it is of great help to utilize first-principles computational methods to provide fundamental insights into understanding liquid metals and their interactions with hydrogen isotopes.

Experiments have found that hydrogen isotopes interact differently with liquid Li and liquid Sn. For example, it is known that hydrogen isotopes can be largely absorbed by liquid Li due to their strong chemical affinity with Li atoms. In contrast, the low reactivity...
of Sn with hydrogen isotopes leads to a small retention ratio of hydrogen isotopes in liquid Sn.\(^{18}\) It is intuitively expected that D atoms can be trapped by Li atoms in liquid Li-Sn eutectic, resulting in a higher number of retained hydrogen isotopes in liquid Li-Sn than liquid Sn. Surprisingly, a recent experiment observed that Li-Sn eutectic has a smaller\(^{24}\) retention ratio of D than pure Sn against intuition. Although the above experiment may suffer from contaminants such as oxygen and carbon at the surface of the sample, it is still worth understanding the fundamental processes regarding the interactions of hydrogen isotopes with liquid Li-Sn. For example, how do D atoms escape from Li-Sn eutectic, causing the low retention ratio of D in Li-Sn? More importantly, what is the underlying mechanism that explain liquid Li, Sn, and Li-Sn interact differently with D?

In this work, we performed first-principles molecular dynamics (FPMD) simulations to tackle the above issues. We observe the formation and evaporation of D\(_2\) molecules in the Li-Sn slab, which may be a reason for the low retention ratio of D observed in experiment.\(^{24}\) These results are remarkable, because as predicted by recent FPMD studies, D\(_2\) molecules hardly form in liquid Li because D atoms form strong ionic bonds with Li atom,\(^{29}\) whereas forming D\(_2\) molecules is also difficult in liquid Sn, which may be caused by the high diffusivity of D in liquid Sn.\(^{30}\) In addition, how many different types of molecules can escape the surfaces of Li-Sn slab are still inconclusive. Notably, a recent work\(^{31}\) has found the formation of D\(_2\) molecules in bulk liquid Li-Sn systems, while the detailed forming processes of D\(_2\) molecules have not been fully explored. Herein, we unveil a cooperative mechanism of forming D\(_2\) molecules in liquid Li-Sn slabs, where Li atoms play a central role to trap a D atom, and wait for a second D to come nearby, triggering the forming process of D\(_2\) molecule. During the formation of D\(_2\) molecule, the surplus electrons are transferred from D\(_2\) to its adjacent Sn atoms. D\(_2\) molecules then diffuse rapidly by leaving their adjacent Li and Sn atoms. In addition, we find the existence of both D\(_2\) molecules and LiD molecules in the vacuum area of the simulated cell. Our results show that LiD molecules are more difficult to form than D\(_2\) molecules, and we predict a optimal temperature window that allows the evaporation of D\(_2\) molecules from surfaces of PFCs but not for LiD molecules, which may potentially contaminate the plasma. The above findings offer a series of new microscopic insights into our understanding of how hydrogen isotopes interact with liquid Li-Sn eutectic.
II. COMPUTATIONAL DETAILS

All of the FPMD simulations were performed with the ABACUS (Atomic-orbital Based Ab-initio Computation at USTC) package. We adopted norm-conserving pseudopotentials and local density approximation. We chose an energy cutoff of 120 Ry for charge density and only the gamma point to sample the Brillouin zone with periodic boundary conditions. We used numerical atomic orbitals as basis sets to construct the electronic wave functions. Specifically, double-zeta plus polarized (DZP) orbital sets were adopted for D (2s1p), Li (2s1p) and Sn (2s2p1d). The radius cutoffs of numerical atomic orbitals were chosen to be 6.0, 9.0 and 8.0 bohr for D, Li, and Sn elements, respectively. Born-Oppenheimer molecular dynamics simulations were performed in the canonical ensemble NVT with the Nosé-Hoover thermostat at 573, 673, and 873 K. A liquid Li-Sn slab with a vacuum and two concentrations of D atoms in a periodic cell (60 × 14.98 × 14.98 Å³ where the vacuum is placed along x-axis) was studied. A Li$_{0.19}$Sn$_{0.76}$D$_{0.05}$ slab containing 36 Li, 144 Sn, and 10 D atoms is referred as the low concentration system, while a Li$_{0.13}$Sn$_{0.51}$D$_{0.36}$ slab consisting of 36 Li, 144 Sn, and 100 D atoms is referred as the high concentration system for brevity. The time step utilized in molecular dynamics was set to 0.2 fs and the trajectories were run for 20 ps.

The liquid configurations of Li-Sn slabs were generated by performing FPMD simulations as follows. First, we ran FPMD simulations of a Li$_{0.20}$Sn$_{0.80}$ slab for 5.0 ps at 1073 K with a time step of 0.5 fs in order to obtain its liquid structure. Second, we utilized the above liquid Li$_{0.20}$Sn$_{0.80}$ structure and ran FPMD simulations for 20 ps at selected temperatures, i.e., 573, 673 and 873 K. Third, we randomly inserted 10 and 100 deuterium (D) atoms into the Li$_{0.20}$Sn$_{0.80}$ system to generate two systems with different D concentrations, i.e., Li$_{0.19}$Sn$_{0.76}$D$_{0.05}$ and Li$_{0.13}$Sn$_{0.51}$D$_{0.36}$ systems, respectively. Fourth, both Li$_{0.19}$Sn$_{0.76}$D$_{0.05}$ and Li$_{0.13}$Sn$_{0.51}$D$_{0.36}$ systems were equilibrated for 10 ps with a time step of 0.2 fs. Finally, we simulated the Li-Sn systems with the inserted D atoms for another 20 ps at different temperatures. All of our analysis was based on the last 20 ps trajectories.
FIG. 1: (Color online) Ionic density profiles of the Li$_{0.19}$Sn$_{0.76}$D$_{0.05}$ and Li$_{0.13}$Sn$_{0.51}$D$_{0.36}$ slabs along the surface normal direction at 573, 673, and 873 K. The two dashed lines at 20.4 and 36.6 Å represent the surfaces of the Li$_{0.19}$Sn$_{0.76}$D$_{0.05}$ Li-Sn slab, while the other two dashed lines at 16.8 and 39.0 Å indicate the surfaces of the Li$_{0.13}$Sn$_{0.51}$D$_{0.36}$ Li-Sn slab.

FIG. 2: (Color online) Surface coverages of Li, Sn, and D atoms in the Li$_{0.19}$Sn$_{0.76}$D$_{0.05}$ and Li$_{0.13}$Sn$_{0.51}$D$_{0.36}$ slabs at 573, 673, and 873 K.
III. RESULTS AND DISCUSSIONS

A. Li-Sn Surfaces

Figure 1 illustrates the simulated ionic density profiles of liquid Li-Sn systems with both low and high concentrations of D atoms at 573, 673, and 873 K. First of all, we find liquid Li-Sn has some short range orders. The ionic density profiles of Li-Sn systems exhibit a series of sharp peaks, where the heights of peaks decrease at higher temperatures, suggesting a more uniform distribution of atoms in the slabs at higher temperatures. Furthermore, the volume of Li-Sn slab is insensitive to temperatures ranging from 573 to 873 K, but substantially increases with a higher concentration of D. Figure 2 shows the surface compositions of Li-Sn slabs from simulations, where the surface is defined as the space between the first peak of ionic density profile [see Figure 1] and the vacuum. For both Li-Sn systems, we observe a similar trend as compared to experiment\textsuperscript{20–22} that the surface coverage of Li increases at a higher temperature. In addition, the surface coverage of D only slightly decreases with increasing temperature. The reason that Li coverage increases at higher temperature is because Li may reduce the surface tension more effectively than Sn.\textsuperscript{20,23} Notably, in the high-concentration Li-Sn system, the surface coverage of D is twice more than that of Sn, although the total number of Sn atoms is slightly larger than the number of D atoms.

B. Formation of D$_2$ Molecules

Figure 3 illustrates the partial pair distribution functions $g_{\alpha\beta}(r)$ of D-D, Li-Li, Sn-Sn, Li-D, Sn-D, and Li-Sn, which were obtained with FPMD simulations in the liquid Li$_{0.19}$Sn$_{0.76}$D$_{0.05}$ slab at 673 K. The partial pair distribution function $g_{\alpha\beta}(r)$ with two different atomic species $\alpha$ and $\beta$ is defined as

\[
g_{\alpha\beta}(r) = \frac{N_{\alpha} + N_{\beta}}{\rho N_{\alpha} N_{\beta}} \sum_{i=1}^{N_{\alpha}} \sum_{j=1}^{N_{\beta}} \langle \delta(r + \mathbf{R}_i - \mathbf{R}_j) \rangle,
\]

where $\rho$ is the atomic density, $N_{\alpha}$ and $N_{\beta}$ are the numbers of atoms for species $\alpha$ and $\beta$, respectively. $\mathbf{R}_i$ and $\mathbf{R}_j$ are the atomic positions of atoms $i$ and $j$. First of all, we can see that the partial pair distribution functions in Figures 3(b-f) all exhibit liquid-like structural characteristics. In particular, as can be seen from Figure 3(a), $g_{DD}(r)$ with peaks centered...
FIG. 3: (Color online) Partial pair distribution functions of (a) D-D, (b) Li-Li, (c) Sn-Sn, (d) Li-D, (e) Sn-D, (f) Li-Sn in the simulated liquid Li_{0.19}Sn_{0.76}D_{0.05} slab at 673 K.

at around 0.8 Å suggests that D₂ molecules form in the liquid Li_{0.19}Sn_{0.76}D_{0.05} slab, as the typical bond length of a D₂ molecule is 0.74 Å.

From the FPMD trajectories, we observe several D₂ molecules in the vacuum area outside of the liquid Li_{0.19}Sn_{0.76}D_{0.05} slab during the molecular dynamics simulations. Note that each D atom was randomly implanted in the liquid Li-Sn slab at the beginning of the simulations. Hence, D₂ molecules can only form from individual D atoms in the Li-Sn sample during the simulations, and then escape into the vacuum area of the simulation cell. In addition, D₂ molecules are only found to dissociate into D atoms inside the Li-Sn slab but not in the vacuum. Importantly, about 60% of the formed D₂ molecules are released into the vacuum, resulting in a low retention ratio of D in the simulated Li-Sn systems, which provides some clues to the recent experiment. The results also suggest that the formation and releasing D₂ molecule may be an effective way for recycling of D atoms in liquid Li-Sn slabs.

In previous FPMD simulations, the formation of D₂ molecules did not occur when the proportion of D atoms is smaller than that of Li atoms due to the strong bonding between Li and D atoms. Only a small amount of D₂ molecules were recorded when the proportions
FIG. 4: (Color online) A series of steps to form \( \text{D}_2 \) molecules in the \( \text{Li}_{0.19}\text{Sn}_{0.76}\text{D}_{0.05} \) slab at 673 K. (a) Li and Sn trap a D atom at the surface. (b) Two D atoms come closer at the surface. (c) Formation of a \( \text{D}_2 \) molecule. (d) The \( \text{D}_2 \) molecule diffuses away from Li and Sn atoms.

of D and Li are equal. No \( \text{D}_2 \) formation with a long lifetime was observed in pure liquid Sn from FPMD studies.\(^{20}\) In stark contrast, in the Li-Sn slab, a considerable number of \( \text{D}_2 \) molecules form in a system with more Li than D, indicating there may be a different mechanism to form \( \text{D}_2 \) in the Li-Sn eutectic. Based on our analysis, there are typically four steps to form \( \text{D}_2 \) molecules in the Li-Sn slab as shown in Figure 4 (see the movie in the SM)\(^{43}\): In the first step, a D atom is first trapped by a Li atom in the Li-Sn slab to form a Li-D pair, as shown in Figure 4(a). Interestingly, the trapped D keeps moving at a relatively low velocity, setting up a stable environment with adjacent Li and Sn atoms in order to attract other D atoms to form a \( \text{D}_2 \) molecule. Figure 3(a) further demonstrates that D atoms interact strongly with Li atoms, as can be seen by the high first peak of \( g_{\text{LiD}}(r) \). Next, Figure 4(b) shows that a second D atom comes close to the first D, while the position of the Li-D pair does not change much. As will be explained later, we find that the second D is in fact attracted and slows down towards the first D. Third, a \( \text{D}_2 \) molecule starts to form by triggering the charge transfer from D to its adjacent Sn atoms (\textit{vide infra}) and a chemical bond between the two D atoms is established, as shown in Figure 4(c). In the last
FIG. 5: (Color online) Distances from D atoms to its adjacent atoms in the Li$_{0.19}$Sn$_{0.76}$D$_{0.05}$ slab during the formation of D$_2$ molecules. $d_{D-D}$, $d_{D-Li}$, and $d_{D-Sn}$ are defined as the distances between D and its adjacent D, Li, and Sn atoms, respectively.

FIG. 6: (Color online) Velocities of two D atoms (labeled as D-1 and D-2) during the formation of D$_2$ molecules in the Li$_{0.19}$Sn$_{0.76}$D$_{0.05}$ slab.

step, the newly formed D$_2$ molecule diffuses away from its Li and Sn neighbors as illustrated in Figure 4(d). We also provide a movie that shows the formation of a D$_2$ molecule near the surface of Li$_{0.19}$Sn$_{0.76}$D$_{0.05}$ system (673 K) and evaporation of this D$_2$ molecule into the vacuum (see the Supporting Information).
C. Processes of Forming D$_2$

We systematically analyze the processes of forming D$_2$ molecules in order to quantify the above observations in simulations. In particular, we track the D-D pairs in the low-concentration Li-Sn slab and report their interesting dynamics. The criterion to track D-D pairs was selected when any two D atoms come close within 1.1 Å and last for more than 0.2 ps. Moreover, we chose a time window of 100 fs that is relevant to the formation of D$_2$ molecules, emphasizing the formation moment which occurs at around 50 fs corresponding to Figures 4(a-c). By investigating the changes of properties before and after the formation of D$_2$ molecules within the 100 fs, we can obtain insights into the fundamental process of forming D$_2$ molecules. Specifically, the analyzed time-dependent properties include distances of D to its neighbors, instantaneous velocities of D atoms, and number of electrons on D, which are respectively shown in Figure 5, Figure 6, and Figure 7, respectively. These time-dependent properties are averaged over the three temperatures studied.

Figure 5 shows that the distance between two D atoms ($d_{D-D}$) quickly shortens approximately from 2.8 to 0.8 Å in the first 50 fs before the formation of D$_2$, and then stably oscillates around 0.8 Å with a damped amplitude up to 100 fs. The value of 0.8 Å is the location of the first peak of the radial distribution function $g_{DD}(r)$, as illustrated in Figure 3(a). Meanwhile, the distances between D atoms and its adjacent atoms, i.e., $d_{D-Li}$
and $d_{D-Sn}$, slowly elongates from around 2.5 to 2.8 Å during the first 50 fs, which can be understood since Li and Sn atoms should provide spaces for two neighboring D atoms to meet and the process of forming D$_2$ starts. Meanwhile, as displayed in Figures 3(d) and (e), D atoms start to deviate from positions of the first peak in $g_{LiD}(r)$ and $g_{SnD}(r)$, indicating that the interactions between D and Li/Sn atoms are weakened. We then see a dramatic increase of $d_{D-Li}$ and $d_{D-Sn}$ from around 2.8 to 4.0 Å within the second 50 fs, suggesting that the D$_2$ molecule quickly diffuse away after its formation (see Figure 4(d) and the movie in the Supporting Information).

The instantaneous velocities of the selected D-D pairs during the formation of D$_2$ molecules are illustrated in Figure 6. One of the two D atoms (labeled as D-1) has an oscillating velocity ranging from 50–65 Å/ps up to around 40 fs. This is much slower than the velocity of the other D atom (labeled as D-2), which is larger than 100 Å/ps, indicating that the two D atoms are experiencing different environments, i.e., the D-1 atom is trapped by surrounding Li and Sn atoms and the D-2 atom diffuses more freely in the liquid. Next, during the formation of D$_2$ molecule at around 50 fs, the D-1 atom experiences a vast increase of velocity within a time window from 40 to 60 fs, whereas the velocity of D-2 abruptly drops, suggesting a collision between the two D atoms occurs with a shortened distance comparable to the bond length of a D$_2$ molecule as shown in Figure 5. After the formation of D$_2$ molecule, the D-1 and D-2 atoms diffuse together with similar velocities which gradually increase to about 120 Å/ps.

In Figure 7, we also compute the averaged charge change of D atoms and their adjacent atoms during the formation of D$_2$ molecules. We utilized the Bader charge analysis method. From this analysis, we find electrons transfer from D to Sn atoms during the formation of D$_2$ molecules. Quantitatively, before forming D$_2$ molecules at around 50 fs, more than 1.5 electrons are assigned to a D atom while less than 4.0 valence electrons per atom are on its nearby Sn atoms. In the first 50 fs, a small amount of electrons (less than 0.5 electrons per atom) on D transfer to Sn. When D$_2$ forms at around 50 fs, the number of electrons on the D atom abruptly drops to about 1.0 within a very short time, which occurs at the same time when the changes of distances and velocities of D atoms occur. Meanwhile, the electrons on Sn are accumulated to above 4.0. The resulting number of electrons on D is slightly above 1.0, which is close to that of a single D atom. Furthermore, the D-D pair therefore has a bond of around 0.8 Å with two electrons. Therefore, we identify the formation of D$_2$
D. Formation of LiD Molecules

The FPMD simulations show that the D$_2$ molecules diffuse rapidly in the Li-Sn eutectic and some of them evaporate into the vacuum. We did not find evaporation of single D atoms, Li atoms, or Li$_2$ molecules in liquid Li-Sn systems from our simulations. Nevertheless, we found the existence of LiD molecules in vacuum, which is not surprising, as Li and D atoms can form strong ionic bonds. Since Li could potentially contaminate the plasma, it is important to know how the LiD molecules evaporate compared to D$_2$. Figure 8 depicts the distributions of D$_2$ and LiD molecules in the low-concentration Li-Sn slab at 573, 673, and 873 K. D$_2$ molecules distribute in both liquid Li-Sn slab and vacuum at all temperatures considered. The more flat distribution of D$_2$ in vacuum than bulk implies that D$_2$ diffuse more freely in vacuum than in the Li-Sn slab. In contrast, Li-D pairs mostly reside in
the Li-Sn slab at 573 and 673 K, and are occasionally recorded around the surface. At the higher temperature of 873 K, we find a small amount of LiD molecules escape to the vacuum, although the chance to find the LiD molecules is almost an order of magnitude smaller than that of D$_2$ molecules. This finding is significant, because it suggests that the facility should work in an optimal temperature window below 873 K, which allows efficient recycling of D atoms but reduce the risk of LiD molecules from contaminating the plasma.

IV. CONCLUSIONS

To summarize, we carry out FPMD simulations to investigate the retention and recycling of deuterium in liquid Li-Sn with two different concentrations (Li$_{0.19}$Sn$_{0.76}$D$_{0.05}$ and Li$_{0.13}$Sn$_{0.51}$D$_{0.36}$) at three temperatures of 573, 673, and 873 K. First of all, we find the formation of D$_2$ molecules in all of the simulations. Importantly, we unveil a cooperative mechanism of forming D$_2$ molecules in liquid Li-Sn, where Li atoms behave as catalytic centers, allowing efficient generation of D$_2$ molecules in liquid metal Li-Sn. During the formation process, Li traps a D atom until another D atom comes to trigger the charge transfer from D atoms to their nearby Sn atoms. After formation, the D$_2$ molecules diffuse quickly in the Li-Sn liquid and evaporate to the vacuum. The above mechanism reduces the retention of D in liquid Li-Sn slabs, and is beneficial for the recycling of D atoms. This work therefore provides new clues to a recent experimental finding that Li-Sn eutectic has an unexpected low retention rate of hydrogen isotopes. Furthermore, although we do not observe evaporation of single D atoms, Li atoms, or Li$_2$ molecules in our simulations, we observe the formation and evaporation of LiD molecules at 873 K. Note that the chance to find LiD molecules is much smaller than that of D$_2$ molecules. We predict that there is a temperature window below 873 K, allowing efficient recycling of D atoms but reduce the risk of LiD molecules from contaminating the plasma. The new findings deepen our understanding of interactions between hydrogen isotopes and liquid Li-Sn and can be potentially used for improving the design of magnetically confined fusion reactors.
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(See the supplementary material at the website for more simulation details and results.).

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