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Full Text

Preamble

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Abstract

We use microscopic Monte Carlo simulation techniques to investigate the impact of H2 diffusion energy on the recombination efficiency of H2 on interstellar dust grain surfaces under diffuse and translucent cloud conditions. We constructed five models representing different possible conditions encountered by adsorbed H and H2 on interstellar dust grains. We implemented adsorption sites with multiple binding energies for surface species; the Encounter-Desorption mechanism was also included. The study focused on silicate surfaces in diffuse clouds and water ice surfaces in translucent clouds. The results show that the recombination efficiency of H2 on dust surfaces decreases as H2 diffusion energy increases. An interesting finding of this work is that considering different binding sites for H and H2 gives rise to multiple steady phases, during which the recombination efficiency remains constant with a change in H2 diffusion energy.

Key words: astrochemistry – atomic processes – ISM: molecules

1. Introduction

Molecular hydrogen (H2) is the most abundant molecule in the universe. Almost all the hydrogen is in its molecular form in dense and translucent clouds. Because of its abundance, H2 plays a critical role in much of the chemistry in star-forming regions \cite{Cazaux&Tielens2002, Cazaux&Tielens2004, Wakelam_{{{et}}*{al}}*{2017}, Bialy_&*{{{Sternberg}}}{{2019}}}, Tacconi_{{{et}}*{al}}*{2020}, Chevance_{{{et}}*{al}}*{2022}, Nakazato_{{{et}}*{al}}*{2022}}. H2 is also overwhelmingly present in various other regions of the interstellar medium (ISM) such as photon-dominated regions, outflows of planetary nebulae, jets, shocks, and supernova remnants \cite{Cazaux&Tielens2004, Iqbal_{{{et}}*{al}}*{2012}}. Ion-molecule reactions involving H2 and dissociative recombination processes are crucial steps in the chemical pathways leading to numerous simple and abundant interstellar species \cite{Cazaux&Tielens2002, Cazaux&Tielens2004, Wakelam_{{{et}}*{al}}*{2017}}. Additionally, H2 is the primary collider in dense clouds and serves as an important coolant, particularly in regions of warm gas \cite{Herbst_&*{{{Klemperer}}}{{1973}}}, Cazaux&Tielens2004, Sternberg_{2006}, Bialy_&*{{{Sternberg}}}{{2015}}}}. Since H2 plays a key role in determining the physical and chemical evolution of the ISM, the study of how H2 is formed remains of fundamental importance.

Studies in the past few decades have established that the gas-phase formation

rate of H2 in the ISM is inefficient for explaining the observed abundance of molecular hydrogen, and one needs to focus on different formation mechanisms on interstellar dust grains to produce the observed abundance of H2 \cite{Gould_&*{{{Salpeter}}}{{1963}}}, Herbst_&*{{{Klemperer}}}{{1973}}}, Cazaux&Tielens2002, Cazaux&Tielens2004, Iqbal_{{{et}}*{{al}}}{2014}, Wakelam_{{{et}}*{{al}}}{2017}}. However, the exact mechanism of H2 formation on dust grain surfaces remains a subject of intense focus.

In the literature, we find that researchers have mostly concentrated on the Langmuir–Hinshelwood (LH) mechanism at low temperatures to study H2 formation on dust surfaces \cite{Watson_&*{{{Salpeter}}}{{1972}}}, Tielens_&*{{{Allamandola}}}{{1987}}}, Cazaux&Tielens2004, Iqbal_{{{et}}*{{al}}}{2012}, Wakelam_{{{et}}*{{al}}}{2017}}. In this mechanism, H atoms accrete on dust grains and then adsorb onto the dust surface through a process known as physisorption, facilitated by weaker binding energies. Once adsorbed, the H atoms overcome the diffusion energy and hop across the dust surface. When an H atom encounters another H atom while hopping, a chemical reaction occurs, forming a H2 molecule. Additionally, H atoms can also desorb from the dust surface if they gain energy higher than the desorption energy.

The desorption and diffusion rates of an H atom increase rapidly with the surface temperature (T) as they are governed by exponential equations of the form $\exp(-E/T)$, where E is the diffusion or desorption energy in Kelvin. To account for efficient H2 formation, an H atom must remain on the surface long enough to meet another H atom and form H2. To facilitate this, (1) the surface temperature must be such that the desorption probability is significantly lower than the encounter probability of two H atoms on the surface, and (2) the temperature should be high enough to enable efficient hopping of H atoms. These requirements ask for conflicting conditions that are hard to meet in various astronomical sources in the ISM. Recent research shows that a possible solution to this conflict could be in the treatment of the desorption energy of H atoms, which is not as uniform as previously assumed in chemical models but follows a Gaussian distribution \cite{Grassi_{{{et}}*{{al}}}{2020}}.

Further, since H atoms do not react with H2 on grain surfaces, early models did not consider the presence of H2 on the dust surface. However, Hincelin et al. \cite{Hincelin_{{{et}}*{{al}}}{2015}} introduced the Encounter-Desorption (ED) mechanism, demonstrating that adsorbed H2 on dust surfaces can affect the formation of H2. Chang et al. \cite{Chang_{{{et}}*{{al}}}{2021}} extended the ED mechanism to include both H and H2. Zhao et al. \cite{Zhao_{{{et}}*{{al}}}{2022}} used the ED mechanism in their simulations and confirmed that the presence of adsorbed H2 on dust surface affects the recombination efficiency of H2. Their findings indicated that the diffusion energy of H2 on the dust surface could impact the recombination efficiency of H2. However, Zhao et al. \cite{Zhao_{{{et}}*{{al}}}{2022}} did not investigate how uncertainties or variations in the diffusion energy of H2 on grain surface impact the recombina-

tion efficiency of H2, and also, in the literature we do not find any work done previously to investigate it. Hence, this study further improves on the work presented in Zhao et al. \cite{Zhao_{{{et}}{{al}}}{2022}} by investigating various models to understand the impact of diffusion energy of H2 on the recombination efficiency of surface H2.

This paper is structured as follows: Section 2 introduces the model developed. Section 3 explains the numerical methods used. The results of the numerical simulations are presented in Section 4. Finally, Section 5 provides a summary and discussion.

2. Model Description

Following the work of Zhao et al. \cite{Zhao_{{{et}}{{al}}}{2022}, we consider three types of binding sites on the grain surface, namely the shallow binding sites, the medium binding sites, and the deep binding sites. The reason for choosing only three types of binding sites is already explained in detail in Zhao et al. \cite{Zhao_{{{et}}{{al}}}{2022}, so we do not discuss it here in further detail. The binding energy at the medium binding sites is 40% higher than that at the shallow binding sites and 40% lower than that at the deep binding sites (again, see Zhao et al. \cite{Zhao_{{{et}}{{al}}}{2022} for details).

In Figures 1 and 2, we illustrate the possible scenarios on the dust grain surface implemented in our model in this work. Initially, during the accretion process, the sites on the dust grain surfaces are unoccupied by other species, and each site can accommodate up to four layers of species on top of each other (generally not more than two layers are formed in our simulations, so to be on the safe side we implemented a 4-layer model). Only the species on the first layer are connected with the grain surface. Species on the higher layers do not interact with the surface. When an atom or a molecule from the gas phase accretes to a site without any horizontal neighbors, it forms a vertical bond with the dust grain surface (species 1 in Figure 1), indicating a shallow binding site. If the accretion site of the species already has one horizontal neighbor, then the species forms both a vertical bond with the surface and a lateral bond with the horizontal neighbor (species 4 in Figure 1), indicating a medium binding site. When the accretion site has two horizontal neighbors, then the species forms two lateral bonds in addition to the vertical bond (species 2 and 3 in Figure 1), indicating a deep binding site.

However, when an atom or a molecule accretes to a site on the dust grain surface that is already occupied by another atom or molecule, the aforementioned situation changes. This is explained using H and H2 as an example in Figure 2. As shown in Figure 2, when an H atom accretes to site A, which is already occupied by an H2, it cannot form lateral bonds with the two horizontal neighbors available at site A. Consequently, deep binding site A, with a significantly higher effective binding and diffusion energy (similar to site C), behaves as a shallow binding site (similar to site B with no lateral binding sites) due to the

presence of an adsorbed H2 molecule. Similarly, a medium or deep binding site behaves as a shallow binding site when occupied by an H atom. This is called the ED mechanism, as mentioned earlier.

Other parameters to consider here are the binding and diffusion energies on different substrates. For example, on an H2 substrate, the desorption energies for H2 and H are significantly lower than those on silicate and water ice. Calculations indicate that at a temperature of 10 K, the desorption energy of H on an H2 substrate is 45 K, while the desorption energy of H2 on an H2 substrate is approximately 23 K \cite{Pierre_{{{et}}{{al}}}{1985}, Vidali_{{{et}}{{al}}}{1991}}. Recent quantum chemical calculations by Das et al. \cite{Das_{{{et}}{{al}}}{2021}} found that the desorption energy of H2 on an H2 substrate varies between 23 and 25 K, and the desorption energy of H on an H2 substrate varies between 67 and 79 K.

Further, H atoms also have the ability to enhance the desorption rate of H2. According to estimates presented in Cuppen & Herbst \cite{Cuppen_&{{{Herbst}}{2007}}}, the desorption energy of H2 on a site previously occupied by an H atom is 30 K, which is significantly lower than the desorption energy of H2 on bare silicate surface. To account for this, in our model the desorption energy of H2 is reduced to 30 K whenever a H2 molecule occupies a site previously held by an H atom. This emphasizes the role of H in regulating the desorption rate of H2. Therefore, in our model, the diffusion and desorption energies for H2 on different surfaces such as top of an H atom, H2 molecule, and bare dust grain surface are considered to be different. Similarly, diffusion and desorption energies for H on top of H2 substrate and bare dust grains are different.

In this work, our aim is to investigate the H2 formation efficiency on the interstellar dust surfaces in the diffuse and translucent clouds, taking into account the possible variations in H2 binding and diffusion energies on different types of surfaces. According to Biham et al. \cite{Biham_{{{et}}{{al}}}{2001}}, the gas phase temperature in diffuse clouds is around 100 K, with dust grains consisting of silicate surfaces. Meanwhile, according to Wakelam et al. \cite{Wakelam_{{{et}}{{al}}}{2017}}, the gas phase temperature in translucent clouds ranges from 15 to 40 K, with dust grains covered by water ice surfaces. To account for these differences in different types of sources, we construct four models (M01, M02, M03, and M04) to investigate the effect of the diffusion energy of H2 on the formation efficiency of H2 on the silicate surface. Additionally, a fifth model, M05, was developed to study the effect of the diffusion energy of H2 on water ice on the formation efficiency of H2. In our models, we also use a parameter μ to represent what percentage of newly formed H2 is instantly released in the gas phase through a process called chemical desorption \cite{Garrod_{{{et}}{{al}}}{2007}}.

In model M01, the desorption energies of H2 and H on the H2 substrate are 23 K and 45 K, respectively and the chemical desorption coefficient μ is set to "1" which means all newly formed H2 is released instantly in the gas phase. To investigate the effect of μ on H2 formation efficiency, model M02 was run with

the value of μ set to "0" while all other parameters are kept the same as in model M01.

To the best of our knowledge, the specific values of the diffusion energy of H2 on different substrates are not well described in the literature. We could only find that Pierre et al. \cite{Pierre_{{{et}}{{al}}}{1985}}, Vidali et al. \cite{Vidali_{{{et}}{{al}}}{1991}}, and Das et al. \cite{Das_{{{et}}{{al}}}{2021}} provide different values as discussed earlier. Hence, to test the impact of different possible values for the diffusion energy of H2 on H2 formation efficiency, we run model M03. This model is similar to M01, but with the diffusion energy of H2 on H2 substrate and the diffusion energy of H on H2 substrate set to 73 K and 24 K, respectively. In models M01, M02, and M03, we assume that the deep binding sites for H atoms are also deep binding sites for H2 molecules, which is a general case. To test a more specific case where deep binding sites are different for different species, we run model M04, in which the deep binding sites for H atoms and H2 molecules are not the same. In models M01-M04, H2 diffusion energy on dust grain surfaces was varied from 308 to 488 K in increments of 12 K to study the effect of the diffusion energy on the recombination efficiency of H2 on dust grain surfaces. Finally, the parameter R in the model is the ratio of diffusion energy to desorption energy on an empty binding site. The ratio between the diffusion energy and desorption energy of a surface species on sites occupied by H2 is R_{H2} and the ratio of H2 diffusion energy to its desorption energy on sites occupied by H atoms is R_H. Table 1 lists parameters described above in different models.

Model M05 was designed to investigate the effect of the diffusion energy of H2 on the synthesis of H2 on dust grain surfaces in translucent clouds, where the dust grain surface is assumed to be covered with water ice. In this model, diffusion energies of H and H2 at sites occupied by H2 are 45 K and 23 K, respectively. The diffusion energy of H2 on water ice was varied, increasing from 194 to 356 K in steps of 9 K.

In Table 2, we summarize the diffusion and desorption energies at shallow, medium, and deep binding sites on dust surfaces that are not occupied by H2. Following the work of Wakelam et al. \cite{Wakelam_{{{et}}{{al}}}{2017}}, the desorption energies (for medium binding sites) used for H atoms on silicate surface and water ice are 510 K, and 580 K, respectively.

3. Numerical Method

This study employed the Microscopic Monte Carlo (MMC) method for simulations. For MMC simulations, binding sites on interstellar dust were arranged in a square lattice with total number of sites $(N_s) = L^2$, where L represents the square root of the total number of sites on the dust surface. Tiné et al. \cite{Tine_{{{et}}{{al}}}{1997}} suggested that setting L to 100 was sufficiently large to eliminate finite-size effects. Therefore, L was fixed at 100 in this study, which saved significant computational time. Each site on the square

lattice has four nearest neighboring sites.

Species from the gas phase can adsorb onto these sites, becoming surface material on the lattice. These species can randomly hop from one site to any of their nearest neighboring sites. We keep tracking the position of each surface species during the process. When two reactants are located at the same site, a chemical reaction occurs. In our models, the only possible reaction that can occur is between H atoms to form H2. Whether H2 remains on the dust surface depends on the value of μ as described earlier. An adsorbed species can also desorb from the surface if it has energy larger than desorption energy.

In our model, H2 is formed immediately on recombination of two H atoms. So, there is no separate time calculated for H recombination event. Thus, there are only three types of events in our simulation: accretion, hopping, and desorption. We calculate the absolute time for each event (accretion, hopping, and desorption) to occur. The event corresponding to the smallest time is found and executed. The system time is reset to this time. We check again which is the next event in the list of events and execute it and increase the system time to the execution time of that event. Thus, the system time evolves with each execution of events until a fixed time is reached and simulation is terminated.

The accretion rate of a species depends on its accretion flux, which is a function of gas temperature and the density of the species and can be written for H as follows:

$$f_H = \frac{n_H v_H}{4 n_s}$$

where v_H and n_H are, respectively, average velocity and number density of H in the gas phase, and n_s is number density of sites on the grain surface. The expression for v_H is:

$$v_H = \sqrt{\frac{8 k_B T_{\rm gas}}{\pi m_H}}$$

where m_H is mass of H atom, $T_{\rm gas}$ is the gas temperature, and k_B is Boltzmann's constant. The accretion rate $R_{{\rm acc},H}$ of H atoms in the units of atoms per sec is given by:

$$R_{{\rm acc},H} = N_s f_H$$

where σ is the cross section of the dust grain which is assumed to be spherical. Now, total number of binding sites on grain surface is given as:

$$N_s = 4\pi a^2 n_s$$

Using Equations (3) and (4), we obtain:

$$R_{\mathrm{acc},H} = \pi a^2 n_H v_H$$

Similarly for H2, we can write:

$$R_{\mathrm{acc},H_2} = \pi a^2 n_{H_2} v_{H_2}$$

In our model, we assume an H density (n_H) of 10 cm^{-3} for the diffuse cloud and a gas temperature of 100 K \cite{Biham_{{{et}}{{al}}}{2001}}. Thus, accretion flux of atomic hydrogen in the diffuse cloud (using Equation (1)) is $f_H = 1.8 \times 10^{-9}$ ML s^{-1}, and we considered that 50% of hydrogen nuclei in the diffuse cloud have already converted to H2. Thus, effective f_H becomes 9×10^{-10} ML s^{-1}. We recall that $N_s = L^2 = 10^4$. Thus we get $R_{\mathrm{acc},H} = N_s f_H = 9 \times 10^{-6}$. Similarly, the accretion rate of H2 is calculated to be 3.18×10^{-6} ML s^{-1}.

In translucent clouds, we assume a gas temperature of 30 K and a hydrogen nucleus density of 1000 cm^{-3} \cite{Wakelam_{{{et}}{{al}}}{2017}}. Under translucent cloud conditions, we assume that 75% of the hydrogen nuclei have already converted to H2, resulting in an H flux of 1.3×10^{-8} ML s^{-1}. Using this in Equation (5), we get the accretion rate of H to be 3.474×10^{-5} ML s^{-1}. Similarly, the accretion rate of H2 is 3.275×10^{-5} ML s^{-1}.

After accretion, species on the dust grain surface can either hop to one of the neighbor sites or desorb to the gas phase. The hopping and desorption rates are calculated, respectively, using the following equations:

$$k_{\mathrm{hop}} = \nu \exp\left(-\frac{E_D}{T}\right)$$

$$k_{\mathrm{des}} = \nu \exp\left(-\frac{E_b}{T}\right)$$

where ν is called the attempt rate, and is set at 10^{12} s^{-1}. E_b and E_D are the diffusion and desorption energies of the species on the dust grain surface, and T is the temperature of the dust grain surface. We calculate k_{hop} and k_{des} for both H and H2 using respective values of parameters.

Both hopping and desorption are random processes and there is a race between hopping and desorption. To know if the species will hop or desorb, we use a random number X between [0, 1] and compare the value of X with the following expression:

$$\frac{k_{\mathrm{hop}}}{k_{\mathrm{hop}} + k_{\mathrm{des}}}$$

If the above expression is true then the species hops, otherwise, it is desorbed to the gas phase.

In our simulations, the waiting time for the accretion of H is obtained using the following equation:

$$\tau_{\rm acc} = -\frac{\ln(Y)}{R_{{\rm acc},H}}$$

where Y is another random number, also between [0, 1], and $R_{{\rm acc},H}$ is the accretion rate of H obtained using Equation (5). Similarly, we also calculate the accretion time ($\tau_{{\rm acc},H_2}$) for H2.

Again, the times for hopping or desorption for both H and H2 are calculated using the following equation:

$$\tau_{\rm hop/des} = -\frac{\ln(Y)}{k_{\rm hop} + k_{\rm des}}$$

where $k_{\rm hop}$ and $k_{\rm des}$ are calculated using Equations (7) and (8).

At the beginning of the simulation, the surface population of H and H2 fluctuates but keeps increasing with simulation time. Over time, the population of H and H2 reaches a steady state and starts to fluctuate around a constant value. Once the steady state is achieved, a sufficiently large time interval Δt is selected. Simulation is allowed to run in the steady state for this time interval Δt and then terminated. The time Δt is chosen to be sufficiently large to ensure that there is a minimum statistical error in the calculation of H2 formation efficiency η due to the fluctuation in H and H2 surface population during the steady state. At the end of time Δt, we calculate the total number of H2 formed (N_{H_2}) and the total number of H adsorbed (N_H) during the time Δt. Finally, the recombination efficiency η is determined using the following equation:

$$\eta = \frac{2N_{H_2}}{N_H}$$

4. Research Results

In this section, we present our results for different models (M01-M05), divided into two sections: diffuse clouds and translucent clouds.

4.1. Diffuse Clouds

In this subsection, we present results for diffuse cloud conditions using representative models M01-M04. Panel (A) in Figure 3 [Figure 3: see original paper] shows the effect of H2 diffusion energy on H2 recombination efficiency η on dust grain surface with the surface temperatures of 10, 12, and 15 K and using model

M01. Here we note that, for any given surface temperature, recombination efficiency η decreases with increasing H2 diffusion energy. To better understand the simulation outcome, the total numbers of H and H2 occupying shallow, medium, and deep binding sites in steady state are plotted, respectively, in panels (A) and (B) in Figure 4 [Figure 4: see original paper], all results are for simulation with 10 K surface temperature. We note that, as the diffusion energy of H2 increases, more and more H2 are likely to occupy deep binding sites, and when the diffusion energy of H2 reaches approximately 375 K, almost all deep binding sites are occupied by H2. Since in model M01 presented here, the deep binding sites for H2 are also the deep binding sites for H, the presence of H2 in all the deep sites causes fast desorption of surface H atoms due to the ED mechanism explained earlier. Consequently, H atoms do not stay long enough on the grain surface to make H2, resulting in a sharp decrease in the recombination efficiency with increasing H2 diffusion energy.

Panels (B) and (C) in Figure 3 show the recombination efficiency η as a function of H2 diffusion energy on the dust surface (at different surface temperatures of 10 K, 12 K, and 15 K) in models M02 and M03, respectively. These results are similar to results obtained in model M01 and shown in Panel (A) in Figure 3. This indicates that the instant desorption of newly formed H2 and the diffusion energies of H and H2 on the H2 substrate have negligible impact on the recombination efficiency of H2 on the dust surface. Therefore, these cases are not discussed in more detail in this work.

We present our simulated H2 recombination efficiency as a function of H2 diffusion energy for model M04 in Panel (D) in Figure 3, where black, red, and blue lines represent the cases for different surface temperatures of 10 K, 12 K, and 15 K, respectively. The only difference in model M04 compared to model M01 is that the deep binding sites for H atoms and H2 are different in model M04. Similar to model M01 results, the results obtained here also show that, at a fixed surface temperature, the H2 recombination efficiency decreases with increasing H2 diffusion energy. However, unlike model M01, we find in model M04 that there are multiple steady phases between certain ranges of H2 diffusion energy. Within this range of H2 diffusion energy, H2 recombination efficiency remains constant. For example, at a surface temperature of 12 K, H2 recombination efficiency remains constant when H2 diffusion energy changes from 308 to 368 K and again from 428 to 488 K.

To explain this dependence of H2 recombination efficiency on H2 diffusion energy, we plotted the statistical distribution of H atoms (Figure 4, panel (C)) and H2 (Figure 4, panel (D)) at shallow, medium, and deep binding sites at 10 K surface temperature. Here in panel (D) in Figure 4, we can see that with increasing H2 diffusion energy, more and more H2 diffuse to deep binding sites, causing an increase in H2 population on grain surface, and hence H population decreases due to the stronger impact of the ED mechanism. This trend continues until H2 diffusion energy has reached approximately 375 K, and at this point, all deep binding sites are filled with H2 causing the impact of the ED mechanism

(arising due to H2 sitting in deep binding sites) to reach its maximum. Hence, we see no further change in H population on the grain surface, and H2 recombination efficiency becomes constant and remains so until H2 diffusion energy is approximately 425 K. Once H2 diffusion energy is further increased, we notice a transition as more and more H2 now start to occupy medium binding sites (see the rising red curve in panel (D) in Figure 4). Now, the H2 population on the grain surface starts to increase with increasing H2 diffusion energy, causing a stronger impact of the ED mechanism on the H atom population on the grain surface. As a result, the H atom population on the grain surface starts to decrease again, causing a gradual decrease in H2 recombination efficiency.

4.2. Translucent Clouds

In this subsection, we discuss results for translucent cloud conditions \cite{Zhao_{{{et}}{{al}}}{2022}} using representative model M05. In Figure 5 [Figure 5: see original paper], we show the effect of H2 diffusion energy on the recombination efficiency on dust surfaces at temperatures of 12 K (black lines) and 15 K (red lines) using model M05. According to Zhao et al. \cite{Zhao_{{{et}}{{al}}}{2022}}, 12 and 15 K are the critical cloud conditions, where changes in the recombination efficiency on water ice occur. Therefore, these two temperatures were chosen for the study. In Figure 5, it can be seen that (at the same surface temperature) the recombination efficiency of H2 on the dust surfaces decreases as the diffusion energy of H2 on dust surfaces continuously increases, highlighting the impact of H2 diffusion energy on water substrate in translucent cloud-like conditions.

5. Discussion

In this work, we use five different models (M01-M05) to do a detailed study to understand how the desorption energy of H2 on the bare dust grain surface (silicate) with three different types of binding sites and the dust grain surface with a water substrate affects the recombination efficiency of H2. The major finding of this research is that (on both silicate surface and water ice) the recombination efficiency of H2 on dust grain surfaces decreases as the desorption energy of H2 increases.

Results obtained from models M01-M03 show that instant desorption of newly formed H2 and the desorption energy of H and H2 on H2 substrate have negligible impact on the recombination efficiency of H2 on the dust grain surfaces.

We also find (comparing model results from M01 and M04) that H2 recombination efficiency is higher when the deep binding sites for H are not the same as those for H2. Under such conditions, a higher population of H2 does not limit deep potential wells available to H. Even if H2 occupies all the deep binding sites available to H2, there are still separate deep binding sites available for H atoms to adsorb. As a result, the surface can retain H atoms in deep binding sites specific to H atoms. These H atoms help to produce H2 efficiently.

Although our model results show that the H2 diffusion energy is important for the recombination efficiency, this energy value is not well known yet. Recently, Meng et al. \cite{Meng_{{{et}}{{al}}}{2023}} found that the ratio of H2 diffusion energy to its desorption energy is approximately 0.57 on diamond-like carbon (DLC) surfaces based on the experimentally measured coverage dependent desorption energy of H2 on DLC surfaces \cite{Tsuge_{{{et}}{{al}}}{2019}}. Assuming this ratio on the DLC surfaces is the same as that on the silicate surfaces, the diffusion energy of H2 is about 350 K in the medium binding sites on silicate surfaces. If the diffusion energy of H2 is 350 K, Models M01 and M02 predict that the recombination efficiency is approximately 0.3, 0.85, and 0.2 at 10 K, 12 K, and 15 K respectively. On the other hand, Gry et al. \cite{Gry_{{{et}}{{al}}}{2002}} found that the minimum H2 recombination efficiency is 0.5 based on the observed H2 formation rate in the diffuse ISM. Therefore, models M01 and M02 still underestimate the recombination efficiency at 10 and 15 K. Similarly, model M03 also underestimates the recombination efficiency at 10 and 15 K while model M04 can reasonably well estimate the recombination efficiency at 10 and 12 K if the diffusion energy of H2 is 350 K.

An interesting finding of this work is that considering different binding sites for H and H2 gives rise to multiple steady phases, during which the recombination efficiency remains constant with a change in H2 diffusion energy. The first steady phase starts when all deep binding sites are filled with H2 and the second steady state starts when all medium binding sites are filled with H2.

The proposed model only considered physisorption, although H can also chemisorb onto silicate or carbon surfaces. It remains unclear if considering both chemical adsorption and encounter desorption mechanisms in future work will help to better explain the formation efficiency of H2 in diffuse clouds. Further research is necessary to uncover the mysteries of H2 formation in diffuse clouds.

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Appendix A

Extra Figures Showing the Relationship between the Difference in Diffusion Barriers between Mid and Deep Binding Sites, and the Ratio of H2 Population at Mid and Deep Binding Sites

The plots in Figure A1 show the relationship between the H2 population (in the mid and deep binding sites) and the energy difference between the mid and deep binding sites. The left and right panels in Figure A1 are for models M01 and

M04, respectively. From the plots, it can be observed that they do not simply follow a Boltzmann distribution. This complexity arises because our simulations incorporate encounter desorption mechanisms, which complicate the situation when species on the dust surface react. This affects the distribution observed. When the dust grain temperatures are 12 K or 15 K, the H2 content on the dust surface is relatively low, hence closer to a Boltzmann distribution. However, at 10 K, where the H2 content on the dust grain surface is higher, the impact of the encounter desorption mechanism is significant, leading to a distribution that differs significantly from that observed at 12 K or 15 K.

Figure A1. Plot showing the relationship between the H2 population (in the mid and deep binding sites) and the energy difference between the H2 population in the mid and deep binding site as a function of energy difference between the mid and deep binding sites. The left and right panels are for models M01 and M04, respectively.

Appendix B

Extra Figures for Fractional H and H2 Population on Grain Surface as a Function of H2 Diffusion Energy

The plots in Figure B1 are same as Figure 4, but show fraction of H and H2 population instead of actual surface population.

Figure B1. Panel (A): The fraction of hydrogen atoms occupying different binding sites. Here, H(0), H(1), and H(2) are, respectively, the fraction of hydrogen atoms in the shallow, medium, and deep binding sites. Panel (B): The fraction of H2 occupying different binding sites. Here, H2(0), H2(1), and H2(2) are, respectively, the fraction of H2 in the shallow, medium, and deep binding sites. Both (A) and (B) panels are for model M01 at 10 K surface temperature. Panels (C) and (D) are same as panels (A) and (B), respectively, but for model M04.

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Note: Figure translations are in progress. See original paper for figures.