Applied Surface Science 457 (2018) 745–751

Contents lists available at ScienceDirect

Applied Surface Science

journal homepage: www.elsevier.com/locate/apsusc

Full Length Article

Theoretical study of the interaction of SF_6 molecule on Ag(1 1 1) surfaces: A DFT study

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ARTICLE INFO

Keywords:
SF_6
Ag(1 1 1)
Absorption
DFT study

ABSTRACT

As a kind of gas insulation medium, SF_6 is widely used in the power industry. Due to its high global warming potential value, the degradation and recycle of SF_6 has become a research hotspot in recent years. At present, there have been some achievements in the experimental study on the degradation of SF_6, but there is little research on the catalytic degradation of SF_6 by metal catalysts. Based on the first-principle, this paper investigated the interaction between SF_6 and Ag(1 1 1) and evaluated the possibility of using Ag as a catalyst in SF_6 degradation. It is found that SF_6 can interact with Ag(1 1 1) surface with eight initial configurations. After the interaction, the distance between the F atom in the SF_6 molecule and the Ag surface is significantly shortened and the S—F bond is elongated. There is a strong charge exchange process between the Ag metal interfaces and SF_6 molecule. Ag (1 1 1) transfers electrons of about 1.1e− to the SF_6 molecule and the dissociation of SF_6 by Ag metal shows a catalytic effect. The related research results provide a theoretical reference for the silver-catalyzed degradation of SF_6.

1. Introduction

Sulfur hexafluoride (SF_6) is widely used as a gas insulation medium in various high-voltage electrical equipment due to its excellent insulation properties and excellent arc extinguishing performance [1,2]. In addition, SF_6 is a colorless, odorless, non-toxic, non-flammable and non-corrosive inert gas. It is also widely used in metal smelting, semiconductor manufacturing, medical, chemical, atmospheric tracer and aerospace industries [3]. However, the Global Warming Potential (GWP) of SF_6 is 23,500 times that of CO_2 and its atmospheric life is as long as 3,200 years. In the Kyoto Protocol signed in 1997, SF_6 gas has been listed as one of the six restrictive greenhouse gases [4]. According to statistics, in the past five years, the content of SF_6 in the global atmosphere has increased by 20%, reaching an order of magnitude of $\sim 10^5$ tons [5,6]. Therefore, the harmless treatment of SF_6 is a hot topic in the field of environmental research in recent years.

At present, the treatment of SF_6 mainly includes catalytic thermal degradation, photodegradation, and electrical degradation. Among them, catalytic degradation mainly uses metal oxides such as Al_2O_3, Fe_2O_3 and metal phosphates to degrade SF_6 under high temperature. However, thermal degradation requires high temperature conditions and catalyst consumption [7–10]. Photodegradation mainly use catalysts such as styrene, TiO_2, etc. In the UV or VIS light conditions, the photocatalysts generate electrons to decompose SF_6. Photodegradation products of SF_6 are mostly non-toxic, but the degradation efficiency is low [11,12]. Electrical degradation mainly use plasma generated by microwave discharge, radio frequency discharge, dielectric barrier discharge and other means, to destroy the molecular structure of SF_6. Despite the high efficiency of SF_6 removal, the main products are SO_2F_2, SOF_2, SOF_4, SO_2 and other toxic gases [13–18]. Methods for efficient and harmless degradation of SF_6 need further study.

Silver(Ag), as a transition metal, is widely used in the field of catalysts and sensors. Ming Yan et al. studied the dissociation process of O_2 on transition metal surfaces such as Ag under the participation of H_2O. The transition metal shows a strong adsorption capacity [19]. Yuhua Chi et al. studied the oxidation of CO and the dissociation of O_2 by Ag-Au clusters. Studies have shown that this cluster has a good catalytic effect on CO [20]. Manaschai Kunaseth et al. studied the catalytic process of the adsorption of volatile metal (VOCs) on transition metal-doped graphene with Ag included and proved that the transition metal has a excellent catalytic effect [21]. Many scholars have carried out simulation and experimental studies on the adsorption or growth of materials on Ag (1 1 1) surface, which proved good stability and

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https://doi.org/10.1016/j.apsusc.2018.06.305
Received 27 March 2018; Received in revised form 10 May 2018; Accepted 30 June 2018
Available online 02 July 2018
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research value of Ag(1 1 1) surface. [22,23]. In recent years, many articles have reported experimental studies on SF_6 catalytic degradation, but there is rare report on the simulation of metal catalytic degradation of SF_6.

SF_6 has a regular octahedron molecular structure with high structural stability, and it is difficult to decompose by conventional degradation methods without catalysts. Therefore, the catalytic decomposition of SF_6 needs to be studied. In this paper, based on the density functional theory (DFT), the adsorption and dissociation of SF_6 on the Ag(1 1 1) surface under different initial conditions were studied. The adsorption energy, charge transfer, density of state (DOS) and differential charge density were calculated. The mechanism of the interaction between SF_6 and Ag(1 1 1) surface was discussed. The related research results provide a theoretical reference for the experimental research of Ag-catalyzed degradation of SF_6.

2. Computational methods

The calculations were carried out in the framework of DFT using Dmol 3 module of materials studio 2017 [24]. The unrestricted density functional theory plus dispersion (DFT-D) calculation in this paper were performed. The generalized approximation (GGA) method within the Perdew-Burke-Ernzerhof (PBE) functions is choosed to calculate the exchange-correlation energy [25]. This method is widely used in the calculation of metal materials and their surfaces with DFT calculations [26–29]. The Grimme method is used to correct the Van der Waals' force to obtain more accurate results [30]. The DFT Semi-core Pseudopods (DSSP) and the double numerical atomic orbital augmented by d-polarization (DNP) were applied [31]. The energy convergence tolerance, the maximum force and the maximum displacement of geometry optimizations were 1.0×10^{-5} Ha, 0.002 Ha/Å and 0.005 Å. The global orbital cutoff radius was set at 5.0 Å [32]. The Ag(1 1 1) surface was modeled using a four-layered mode with (3 × 3) super cell. The vacuum layer of 20 Å is added perpendicular to the surface in order to avoid the periodic interactions.The bottom layer of the Ag(1 1 1) was fixed. The Brillouin zone is sampled using a 4 × 4 × 1 Monkhorst-Pack k-point grid with a Methfessel-Paxton smearing of 0.1 eV [33–36].

The adsorption energies (E_{ads}) are calculated as follows:

$$E\text{ads} = Emetal + Egas - Egas/metal \quad (1)$$

where $Egas/metal$is the total energy of SF_6 gas molecule absorbed on the Ag(1 1 1) surface, $Emetal$and $Egas$represented for the total energy of the relaxed metal and gas molecular, respectively. Following this equation a positive value of E_{ads} indicate that adsorption process is exothermic.

In order to further investigate the interaction mechanism of SF_6 gas molecule with Ag(1 1 1) surface, the charge transfer Q_t and the electron density difference of the structure were calculated by Mulliken analysis. If Mulliken charge(e) < 0, the atom is positively charged and if Mulliken charge(e) > 0, the atom is negatively charged. Density of States (DOS) was analyzed to obtain the electronic structure and the properties of the relaxed structure before and after absorption.

3. Results and analysis

3.1. Surface models

Fig. 1 shows the structure of the SF_6 molecule, which is a regular octahedron structure. The bond angle of each S—F bond is 90° and the bond length is 1.608 Å, which is consistent with the calculation result in reference [37].

Fig. 2 shows four typical adsorption sites on the surface of Ag(1 1 1), labeled as Top site, Bridge site, Face-centered-cubic site and Hexagonal-close-packed site. Due to the symmetry of SF_6 molecules, eight initial configurations were constructed to demonstrate the interaction of SF_6 and Ag (1 1 1), as shown in Fig. 3. M1 corresponds to the case where the

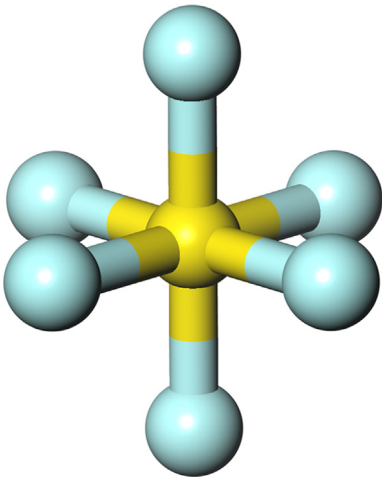

Fig. 1. Molecular structure of SF_6.

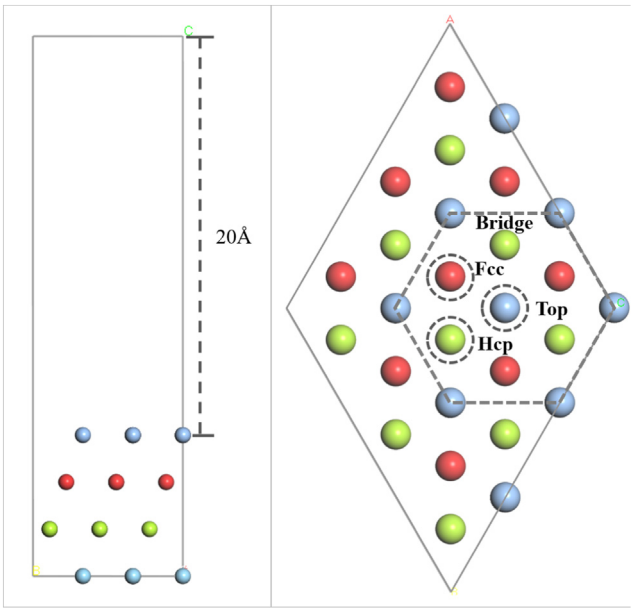

Fig. 2. Structure of Ag (1 1 1) surface. The light blue, green, red, lake blue spheres represent the first, second, third and fourth layer of Ag (1 1 1). Top, Bridge, Fcc and Hcp denote the Top site, Bridge site, Face-centered-cubic site and Hexagonal-close-packed site, respectively. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

straight lines of the two S—F bond of SF_6 are perpendicular to the Ag (1 1 1) metal surface. M2 corresponds to the case where two bottom F atoms are close to the adsorption sites at the same distance. They are defined as M1-Top, M1-Bridge, M1-Fcc, M1-Hcp, and M2-Top, M2-Bridge, M2-Fcc, and M2-Hcp, respectively. The distance of the F atom from the Ag(1 1 1) surface in M1 is 2.000 Å and the two bottom F atoms in M2 are set to 2.000 Å from the adsorption sites on metal surface.

3.2. SF_6 absorbed on Ag(1 1 1) surface

Table 1 shows the adsorption energy and total charge transfer of SF_6 adsorbed on the Ag(1 1 1) surface in different initial configurations. Among the adsorption distances in Table 1, F_x-Ag means the distance from F_x atom to Ag(1 1 1) surface. Fig. 4 shows the optimized structure of the eight initial structures. We calculated the net charge variation based on the Mulliken charge analysis. Q_t is the value of the total charge transfer of SF_6 gas molecule. The negative value of total charge transfer

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