1. Introduction

As a new generation light source, phosphor converted white light-emitting diodes (pc-WLEDs), generally constructing by a blue LED chip coated with the phosphor, are being applied in many emitting diodes (pc-WLEDs), generally constructing by a blue LED chip coated with the phosphor, are being applied in many emitting diodes (pc-WLEDs). In a white LED package, phosphor is always sensitive to high temperature and high moisture, and its stability directly affects the reliability of white LED [3]. Since phosphors are always sensitive to high temperature and high moisture, the thermal quenching and hydrolysis effects become critical concerns on the phosphor’s reliability. Furthermore, phosphor is always mixed within silicone as a light-conversion composite. When the LED operates under harsh application environments, the silicone/phosphor interface is always suffering degradation under conditions of high temperatures [4], high blue light illumination, and high humidity [5–11]. Therefore, the study of interface adhesion in phosphor/silicone composite become one of fundamental researches to improve the reliability of LED package.

In recent years, CaAlSiN₃:Eu²⁺ has attracted much attention for its application in warm white or high color rendering [12–14]. It was firstly developed by K. Uheda et al. under a high-temperature and high-pressure solid state reaction with EuN, Ca₃N₂, AlN and Si₃N₄ [15]. The spectrum measurements conducted by Pan. He et al. showed that the excitation and emission spectra of CaAlSiN₃:Eu²⁺ phosphors could be effectively excited at 467 nm and exhibit a strong red emission at 668 nm, which indicates the CaAlSiN₃:Eu²⁺ phosphor as an excellent candidate for getting red emission for white light-emitting diodes [16]. According to the effective first-principles calculation, the host lattice constants [17], mechanical, electrical and optical properties [18] of CaAlSiN₃:Eu²⁺ and its derivatives were predicted. Although phosphors generally have high thermal quenching temperature and good thermal...
stability, the reliability of CaAlSiN₃:Eu²⁺ red phosphor is still being challenged by the high humidity application condition. Zhu et al. [19] observed the degradation of CaAlSiN₃:Eu²⁺ in high-temperature and -pressure water stream test, which is explained as the oxidation of both the phosphor host and activator under an oxidant-gas penetration. Our research team also experimentally found the hydrolysis phenomenon of CaAlSiN₃:Eu²⁺ red phosphor, that can lower the crystallinity of CaAlSiN₃ and increases its thermal quenching effect [20].

In addition, in a white LED packaging structure, phosphors are always mixed with silicone to form the high-performance composite materials [21]. However, when the phosphors composites are used in the harsh environment (e.g., high temperature and high moisture conditions), the serious lumen degradations and color shifts of a pc-WLED package were observed [22,23]. Luo et al. [7] studied the degradation mechanisms of phosphor/silicone composites used in pc-WLEDs under both high temperature and high humidity conditions. The results show that the hydrolysis of phosphors and the oxidation of silicone under a high moisture environment could accelerate the degradation of phosphor/silicone composites. Moreover, the adhesion behavior between silicone and phosphors is also a serious concern that affects the use of such material in LED packaging. Singh et al. [24] reported the lumen degradation of high-power LEDs aged under high-humidity condition and observed that the voids between silicone and phosphors can promote the adsorption of moisture on silicone, which further could result in subsequent light scattering. However, all current characterizations of phosphors and their silicone composites are studied at macro-level and the understandings on the interactions between phosphor and silicone mainly rely on the characterizations of microstructure and chemical element composition, which cannot provide a deep explanation at atomic level. The explicit physical–chemical effect of humidity condition on the adhesion behavior between silicone and phosphor is still an open question. Those insufficient investigation methods on phosphor/silicone interaction will limit the studies and applications of CaAlSiN₃:Eu²⁺ and its composite.

In this paper, the hydrolyzed CaAlSiN₃:Eu²⁺ red phosphors are obtained via a water immersion experiment, then the pristine and hydrolyzed CaAlSiN₃:Eu²⁺ red phosphors are mixed with silicone to form phosphor/silicone composites respectively. Through tensile tests, their mechanical properties are experimentally obtained and compared. After that, the adhesion properties of silicone molecular on pristine and hydrolyzed surfaces of CaAlSiN₃[0 1 0] are investigated by using Density Functional Theory (DFT) calculations. Through comparing the adsorption energy, bonding nature, electronic structures and sliding energy barriers of silicone molecular on both pristine and hydrolyzed CaAlSiN₃[0 1 0] surfaces, the hydrolysis effect on the interfacial and mechanical properties of CaAlSiN₃:Eu²⁺/silicone composite are discussed.

2. Experiments and methodology

2.1. Sample preparation and mechanical tests

A commercialized CaAlSiN₃:Eu²⁺ red phosphor is used in this study. The test sample preparation procedure for phosphor/silicone composites is shown in Fig. 1 [11,25]: Firstly, red phosphor powders were soaked in deionized water under 55 °C for 1800 s and the hydrolysis reaction was occurred at the surface of phosphor powders as present in Fig. 2. Then, the phosphor powders were filtered and dried from solution. Next, the silicones KJC-1200A and KJC-1200B were mixed with a 1:1 mass ratio. The silicone mixture and the fresh and treated red phosphor powders were thoroughly mixed in a vacuum mixer with mass fractions as 5%, 10%, 15% and 20% respectively. Finally, the phosphor/silicone mixtures were poured into a polyfluorotetraethylene mold and cured in a 100 °C oven for 3 h. The geometric dimensions of prepared test samples with a thickness of 1 mm were designed according to the ASTM D1708 standard. The tensile test was conducted by an Electromechanical Universal Testing Machine from the MTS system (China) Co. Ltd (Model: CMT4204, accuracy: level 0.5).

2.2. Computational method

In this study, all first principles calculations were performed based on the density functional theory (DFT) as implemented in the DMol³ package [26]. The electronic interactions were employed by using the generalized gradient approximation (GGA) with the Perdew-Burke- Ernzerhof (PBE) method [27]. Double numerical basis sets with polarization functions (DNP) were utilized. For geometry optimization calculations, a 5 × 4 × 3 Monkhorst-Pack k-point mesh for the Brillouin zone sampling was used. The convergence criteria of optimized structures are 2 × 10⁻⁵ Ha for energy, 0.002 Ha/Å for force, and 5 × 10⁻³ Å for displacement. Since the chemical elements and composition measurement by using the Energy Dispersive Spectroscopy (EDS) (see the Supplementary Information) indicate that the atomic percentage of Eu element is very small and significantly lower than the other four elements, the host CaAlSiN₃ is used in the DFT calculation to investigate the adhesion behavior between silicone and phosphor particle [25]. A unit cell of CaAlSiN₃ crystal structure was calculated with 24 atoms, placing two Al atoms and two Si atoms in the four tetrahedral sites, as shown in Fig. 3. When all the structures are fully relaxed by employing the conjugate gradient method, the lattice constants of the unit cell are a = 9.8871 Å, b = 5.7134 Å, and c = 5.1146 Å. These computed parameters are in good agreement with the previous studies [17,18].

Normally, CaAlSiN₃ surfaces with three orientations [1 0 0], [0 1 0] and [0 0 1] were reported in previous study [28]. Among these three orientations, the calculations of surface energies show that the CaAlSiN₃ [0 1 0] surface has the lowest surface energy (see the Supplementary Information). Thus, to simplify the calculations of all surfaces of CaAlSiN₃, the most thermodynamically stable one, CaAlSiN₃ [0 1 0] surface, is selected in this study. As shown in Fig. 4(a), the CaAlSiN₃[0 1 0] surface was modeled with ten atomic layers of CaAlSiN₃ and a thick vacuum layer (> 20 Å) was added in the direction perpendicular to the CaAlSiN₃ [0 1 0] surface to ensure the minimum interaction between periodic structures. The bottom two atomic layers of CaAlSiN₃ are fixed at their bulk positions, which avoids the effects of broken bonds at the bottom on the surface properties, as shown in the Supplementary Information. After fully relaxing the CaAlSiN₃[0 1 0] surface, two water molecules were added to the surface structure to obtain the hydrolyzed CaAlSiN₃[0 1 0] surface. Generally, the adhesion behaviors of a common silicone molecular on phosphor surface are very complicated, as in the local area, the adhesion modes are varied from case to case. But such local adhesion behaviors can be understood through the studies of a monomer silicone molecular on phosphor surface at different adsorption sites. Thus, we chose the monomer silicone molecular to study the adhesion property between silicone and CaAlSiN₃ as shown in Fig. 4(b).

To investigate the adsorption of the silicone molecular on CaAlSiN₃[0 1 0] surface, we first calculated the adsorption energy (E_ads) by using the following formulation [29],

\[ E_{ad} = E_{silicone/CaAlSiN₃} - E_{silicone} - E_{CaAlSiN₃} \]

where \( E_{silicone/CaAlSiN₃} \) is the total energy of silicone/pristine or hydrolyzed CaAlSiN₃ structure, \( E_{silicone} \) is the energy of silicone molecular, and \( E_{CaAlSiN₃} \) is the energy of the isolated pristine or hydrolyzed CaAlSiN₃ [0 1 0] surfaces. To link the adsorption energy to its electronic structure, we also calculated the charge density of differences for selected adsorption models via [30],

\[ \rho = \rho_{silicone/CaAlSiN₃} - \rho_{silicone} - \rho_{CaAlSiN₃} \]

where \( \rho_{silicone/CaAlSiN₃} \), \( \rho_{silicone} \) and \( \rho_{CaAlSiN₃} \) are the total charge density of the optimized silicone/CaAlSiN₃ structure, the silicone molecular and the pristine or hydrolyzed CaAlSiN₃[0 1 0] surfaces, respectively. In addition, to investigate adhesion behavior of silicone molecular
sliding along CaAlSiN₃ surface, the transient state (TS) calculation was conducted to determine the minimum energy pathway and energy barrier by using the method of Linear Synchronous Transit-Quadratic Synchronous Transit (LST-QST) calculation with Conjugate Gradient (CG) minimization [31].

3. Results and discussion

3.1. Simulation results

Before calculating the adhesion of silicone molecular on CaAlSiN₃, we first modeled the pristine and hydrolyzed CaAlSiN₃[0 1 0] surfaces. Fig. 5(a) displays the fully relaxed pristine CaAlSiN₃[0 1 0] surface, the positions of the atoms at the top three layers are slightly changed before and after the optimization. Compared to the Si–N–Al angle in the bulk CaAlSiN₃ structure (which is 122.71°), a more open structure with an angle of 152.86° was observed in CaAlSiN₃[0 1 0] surface. In this Si–N–Al, the bond length of Si–N decreases from 1.736 Å to 1.693 Å and the bond length of Al–N decreases from 1.872 Å to 1.810 Å. And the position of Ca atom moves closer to the surface after optimization.

On the basis of fully relaxed pristine CaAlSiN₃[0 1 0] surface structure, the hydrolyzed surface was obtained as shown in Fig. 3(b). The modeling results show that a chemical reaction occurs at the surface of hydrolyzed CaAlSiN₃. The H⁺ and OH⁻ of water molecule are bonded on the surface, forming two N–H bonds with length 1.027 Å, a Si–O bond with distance 1.731 Å and an Al–O bond with length 1.832 Å. Furthermore, the angle of Si–N–Al at the hydrolyzed surface...
is 148.24°, smaller than that in pristine surface structure. These different geometry features between hydrolyzed and pristine CaAlSiN₃[0 1 0] surfaces suggest that both surfaces may have distinctly different adhesion behaviors to the silicone molecular.

3.1.1. Adsorption of silicone molecular on the pristine and hydrolyzed CaAlSiN₃[0 1 0] surfaces

We identify four adsorption sites on the pristine CaAlSiN₃[0 1 0] surface as shown in Fig. 6(a), the tops of two N atoms (site-N1 for the orange ball, site-N2 for the green ball) and the tops of two Ca atoms (site-Ca1 for the black ball, site-Ca2 for the blue ball). On the hydrolyzed CaAlSiN₃[0 1 0] surface, the tops of H atoms are considered as four adsorptions sites as depicted in Fig. 6(b), site-H1 for the orange ball, site-H2 for the green ball, site-H3 for the black ball and site-H4 for the blue ball.

For the case of silicone adsorption, we modeled the adsorption in two modes: the parallel to the surface and the upright to the surface, as shown in Fig. 6(c). The calculated results for both modes on different sites of pristine and hydrolyzed CaAlSiN₃[0 1 0] surfaces are listed in Table 1. It can be seen that the parallel adsorption configurations tend to the higher adsorption energies compared to the upright adsorption.
mode. The values of charge transfer for the parallel configurations are also higher than the upright mode. We ascribe those preferences to the larger contact area of the parallel mode which promotes the interactions between silicone molecular and the surface. From Table 1, we also can find that no matter what adsorption modes, parallel or upright, the average adsorption energy of silicone molecular on the hydrolyzed surface is higher than that on the pristine surface.

Now, we focus on the most thermodynamically stable adsorptions on pristine CaAlSiN$_3$[0 1 0], namely the parallel model of silicone on site Ca1. As plotted in Fig. 7, the silicone molecular is adsorbed on the surface and oxygen atom moves to the position above Ca atom with distance 2.622 Å. Since the interaction with silicone molecular, the height of Ca atom is a little bit higher than the other Ca atoms. Four methyl groups of silicone are downward to the surface and the shortest distance is 2.276 Å as the length between H and N atoms. Before and after the adsorption, the angle of Si–O–Si in silicone molecular changes from 136.42° to 131.0° and the length of Si–O bond changes from 1.677 Å to 1.719 Å. The adhesion energy for this configuration is $-1.994 \text{ eV}$. The plot of charge density difference shows that the charges are depleted at the Ca atom of CaAlSiN$_3$[0 1 0] and methyl groups of silicone, while the charge accumulation appears at the O atom of silicone and N atoms of CaAlSiN$_3$[0 1 0] as shown in Fig. 7(c). Overall, the total charge transfer of this case is very slight ($\Delta Q = -0.003 \text{ e}$) and the pristine CaAlSiN$_3$[0 1 0] surface acts as a charge donor and silicone molecular acts as an acceptor. Above calculated adsorption energy and total charge transfer suggest that the silicone molecule is physically adsorbed on the pristine CaAlSiN$_3$[0 1 0] surface via vdW interactions.

On the hydrolyzed CaAlSiN$_3$[0 1 0] surface, the most stable adsorption site is H2 and the corresponding optimized structure is shown in Fig. 8. The silicone molecular is absorbed on the surface with Si–O–Si parallel to the surface and four methyl groups downward to the surface. The adhesion energy of this configuration is $-2.237 \text{ eV}$ and the shortest distance between silicone and surface is the length between O and H atoms, 2.182 Å. Before and after the adsorption, the angle of Si–O–Si slightly changes from 136.498° to 136.342°, and the length of Si–O bond increases from 1.677 Å to 1.688 Å. Furthermore, according to the definition of hydrogen bond [32], we find that the silicone molecular is absorbed on the hydrolyzed CaAlSiN$_3$[0 1 0] surface via hydrogen bond O–H···O, as shown in Fig. 8(d). The CDD of this configuration is plotted in Fig. 8(c), an obvious charge transfer between hydroxyl and O atom of silicone is observed, where the charges are accumulated at the side of O atom but depleted at the side of hydroxyl of surface. Generally, the total charge transfer from silicone molecular to surface is $-0.028 \text{ e}$ and hydrolyzed CaAlSiN$_3$[0 1 0] surface acts as a charge donor and silicone molecular acts as an acceptor. Compare to the vdW interactions between silicone and pristine CaAlSiN$_3$, the hydrogen bonding contributes a stronger intermolecular interaction. Such different bonding nature of silicone on pristine and hydrolyzed CaAlSiN$_3$[0 1 0] surfaces explains the trend, shown in Table 1, that the silicone adsorbs much stronger on hydrolyzed surface.

### 3.1.2. Sliding of silicone molecular on the pristine and hydrolyzed CaAlSiN$_3$[0 1 0] surfaces

On the basis of thermodynamically most stable adsorptions of silicone on CaAlSiN$_3$[0 1 0] surface, we study the sliding behaviors of silicone on pristine and hydrolyzed surfaces, respectively. On pristine CaAlSiN$_3$[0 1 0] surface, the minimum-energy pathways to slide along [1 0 0] and [0 0 1] directions are plotted in Fig. 9. Along [1 0 0] direction, in one period (from site Ca1 to next site Ca1), the silicone molecular migrates from initial state to intermediate state, passing by the first transit state. Then, the silicone molecular passes the second transit state and finally reaches the final state. The distance of this

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### Table 1

<table>
<thead>
<tr>
<th>Configuration</th>
<th>Site</th>
<th>$E_u$ (eV)</th>
<th>$\Delta Q$ (e)</th>
<th>$d_{min}$ (Å)</th>
<th>Site</th>
<th>$E_u$ (eV)</th>
<th>$\Delta Q$ (e)</th>
<th>$d_{min}$ (Å)</th>
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<td>0.003</td>
<td>2.391</td>
<td>H2</td>
<td>-2.237</td>
<td>-0.028</td>
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<tr>
<td></td>
<td>Ca1</td>
<td>-1.994</td>
<td>-0.003</td>
<td>2.276</td>
<td>H3</td>
<td>-2.195</td>
<td>-0.028</td>
<td>2.248</td>
</tr>
<tr>
<td></td>
<td>Ca2</td>
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<td>0.027</td>
<td>2.227</td>
<td>H4</td>
<td>-2.159</td>
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</tr>
<tr>
<td>Upright</td>
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<td>-0.004</td>
<td>3.221</td>
<td>H1</td>
<td>-1.579</td>
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<tr>
<td></td>
<td>N2</td>
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<td>-0.005</td>
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<td>H2</td>
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<td>-0.015</td>
<td>2.182</td>
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<tr>
<td></td>
<td>Ca1</td>
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<td>2.753</td>
<td>H3</td>
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<td>-1 x 10^-4</td>
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<tr>
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<td>-0.02</td>
<td>2.546</td>
<td>H4</td>
<td>-1.596</td>
<td>-0.004</td>
<td>1.925</td>
</tr>
</tbody>
</table>

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Fig. 7. The thermodynamically most stable adsorption structure of silicone molecular on pristine CaAlSiN$_3$[0 1 0] surface, (a) view along [1 0 0] direction (b) view along [0 0 1] direction. (c) Charge density difference of the silicone/pristine CaAlSiN$_3$[0 1 0] surface, where blue regions represent the charge accumulation and yellow regions mean the charge depletion. The isosurface is taken as $5 \times 10^{-5}$ e/Å$^3$. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

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The sliding process is $L_1 = 9.88 \text{ Å}$. If we set the energy of the initial state as 0 eV, the energies at first and second transit states are 1.16 eV and 1.07 eV, respectively. On pristine CaAlSiN$_3$[0 1 0] surface, silicone molecular at least requires 1.16 eV to overcome the energy barrier to slide along [1 0 0] direction. In [0 0 1] direction, the silicone molecular passes a transit state and reaches the final state with sliding distance $L_2 = 5.71 \text{ Å}$, as shown in Fig. 9(b). The energy barrier along this direction is 1.22 eV.

On the hydrolyzed CaAlSiN$_3$ surface, when silicone slides along [1 0 0] direction, there are two transit states and one intermediate state in one period, as shown in Fig. 10(a). The energies of both transit states are 1.39 eV and 1.40 eV, respectively, so the sliding energy barrier along this direction is 1.40 eV. For the sliding along [0 0 1] direction, the silicone molecular passes a transit state and reaches the final state with energy barrier 1.31 eV, as shown in Fig. 10(b). The embedded pictures in Fig. 10 show that the minimum energy pathways for both directions are passing over the hydroxyls, not through the middle regions between hydroxyls. Compare to the sliding behaviors on pristine CaAlSiN$_3[0 1 0]$ surface, the energy barriers along [1 0 0] and [0 0 1] directions at hydrolyzed CaAlSiN$_3[0 1 0]$ surface are about 20% and 8% higher than that on pristine CaAlSiN$_3[0 1 0]$ surface, respectively. There are two reasons for such differences. Firstly, the hydroxyls bonded on the CaAlSiN$_3$ surface increase the surface roughness, which plays a role as resistance, increasing the friction between the silicone molecular and CaAlSiN$_3$. Secondly, the adhesion energy of silicone on the hydrolyzed CaAlSiN$_3$ surface is higher than that on the pristine surface, so the higher energy is needed to break the adhesion between silicone and hydrolyzed surface. As results, both reasons cause higher energy barriers for the sliding of silicone on hydrolyzed CaAlSiN$_3[0 1 0]$.

### 3.2. Experimental results and explanation

It is well known that the mechanical properties of a composite are dependent on both filler’s and matrix’s properties and the ability to transfer stresses across the filler/matrix interface [33]. In particular, the ability to transfer stress across the interface is often discussed in terms of ‘adhesion’, which, in fact, is related to a complex combination of factors, such as the interfacial shear strength [34]. In the tensile test described in part 2.1, the mechanical properties of silicone/hydrolyzed CaAlSiN$_3$:Eu$^{2+}$ composite were obtained and compared to the silicone/pristine CaAlSiN$_3$:Eu$^{2+}$ composite. As plotted in Fig. 11, after hydrolysis reaction, the tensile strength and Young’s modulus of silicone CaAlSiN$_3$:Eu$^{2+}$ composite were increased. The SEM images in Fig. 2 show that the hydrolysis reaction mainly occurred at the surface of CaAlSiN$_3$:Eu$^{2+}$ and the microstructure of CaAlSiN$_3$:Eu$^{2+}$ has not dramatically crashed after hydrolysis reaction. Thus, in this experiment, the main difference between hydrolyzed and pristine CaAlSiN$_3$:Eu$^{2+}$/...
silicone composites came from their interfaces. Since SEM characterization shows the rougher surfaces of CaAlSiN3 CaAlSiN3:Eu2+ particles after hydrolysis reaction, so we suspect that the increased friction between silicone and CaAlSiN3 is the major contribution to the enhanced mechanical properties of silicone/CaAlSiN3:Eu2+ composite.

Furthermore, the DFT results in part 3.1 provide a new insight to understand the interface of silicone/CaAlSiN3:Eu2+ at atom level. First, the calculated results showed that the hydrolysis reaction of CaAlSiN3[0 1 0] indeed is able to increase surface roughness, which further leads to the higher sliding energy barrier when silicone slides on CaAlSiN3[0 1 0]. It is implied that the friction on hydrolyzed surface is higher than that on pristine CaAlSiN3[0 1 0] surface. The DFT studies also revealed a possible bonding nature between silicone and hydrolyzed CaAlSiN3[0 1 0]: the hydrogen bond O⋯H⋯O. Since the strength of hydrogen bond is obviously higher than the vdW interaction, as a result, the higher adhesion energy is obtained for silicone on hydrolyzed CaAlSiN3[0 1 0]. These simulation results imply that the hydrolysis reaction of CaAlSiN3:Eu2+ can increase the adhesion strength between silicone and CaAlSiN3 and enhance its composite mechanical properties.

4. Conclusions

In this paper, by using both experimental and theoretical methods, the effects of hydrolysis reaction of CaAlSiN3:Eu2+ on the mechanical and interfacial properties of silicone/CaAlSiN3:Eu2+ composite are investigated. In experiments, the hydrolysis reaction of CaAlSiN3:Eu2+ increases its surface roughness, and the tensile tests further show that both tensile strength and Young’s modulus of silicone/hydrolyzed CaAlSiN3:Eu2+ composite are enhanced after hydrolysis reaction. In DFT studies, we find that the adhesion of silicone molecular on the pristine CaAlSiN3[0 1 0] is the weak physisorption via vdW interaction while the adsorption on the hydrolyzed CaAlSiN3[0 1 0] surface is a more complex bonding nature: hydrogen-bonding of O⋯H⋯H. This is well corroborated with the surface electric structures where we clearly observe the corresponding redistribution of charge density on the silicone-CaAlSiN3 interface system. In addition, using the transient state calculations, we reveal that the sliding energy barrier of silicone on hydrolyzed CaAlSiN3[0 1 0] is higher than that on pristine
CaAlSiN$_3$[0 1 0], as the stronger adsorption energy and increased surface roughness. Generally, our experimental and simulation results consistently conclude that the hydrolysis reaction of CaAlSiN$_3$:Eu$^{2+}$ will increase the adhesion of CaAlSiN$_3$: Eu$^{2+}$ phosphor/silicone interface.

Author contribution

Zhen Cui contributes to the modeling and simulations. Jiajie Fan contributes to the experiments and measurements, and provides project administration and funding acquisition. Hendrik Joost van Ginkel contributes the result analysis. Xuejun Fan and Guoqi Zhang support supervision.

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Appendix A. Supplementary material

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