|  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- |
| **Bond length** | **A(I)** | **A(II)** | **A(III)** | **A(IV)** | **B(I)** | **B(II)** | **B(III)** | **B(IV)** |
| Be41/Al-O41 | 1.60 | 1.80 | 1.55 | 1.80 | 1.56 | 1.83 | 1.54 | 1.83 |
| Be41/Al -O31 | 1.60 | 1.80 | 1.52 | 1.80 | 1.55 | 1.81 | 1.54 | 1.83 |
| Be32-O31 | 1.54 | 1.55 | 1.60 | 1.54 | 1.56 | 1.55 | 1.54 | 1.54 |
| Be42- O42/AL & S | 1.54 | 1.55 | 1.54 | 2.05 | 1.56 | 1.55 | 1.56 | 2.05 |
| Be52 -O42/Al & S | 1.54 | 1.55 | 1.53 | 2.02 | 1.55 | 1.55 | 1.54 | 2.02 |
| Be52-O51/S | 1.54 | 1.55 | 2.06 | 1.54 | 1.57 | 1.55 | 2.05 | 1.54 |
| Be61-O51/S | 1.54 | 1.55 | 2.02 | 1.56 | 1.55 | 1.55 | 2.02 | 1.56 |
| Be61-O61 | 1.54 | 1.55 | 1.54 | 1.54 | 1.58 | 1.55 | 1.54 | 1.54 |
| Be51-O61 | 1.55 | 1.55 | 1.55 | 1.55 | 1.57 | 1.55 | 1.55 | 1.55 |
| Be51-O41 | 1.54 | 1.55 | 1.55 | 1.55 | 1.55 | 1.55 | 1.55 | 1.55 |
| Be52-O62 | 1.55 | 1.55 | 1.55 | 1.54 | 1.55 | 1.55 | 1.54 | 1.54 |
| **Bond angle** |  |  |  |  |  |  |  |  |
| O41-Be41/Al-O31 | 113 | 105 | 120 | 103 | 118 | 102 | 118 | 101 |
| O31—Be41/Al-O51/S | 116 | 106 | 116 | 111 | 118 | 105 | 115 | 109 |
| Be52-O51/S-Be41/AL | 119 | 123 | 88 | 128 | 115 | 124 | 89 | 127 |
| O31-Be32-O42 | 120 | 119 | 113 | 122 | 117 | 120 | 119 | 123 |
| Be41-O31-Be32 | 119 | 123 | 118 | 128 | 116 | 124 | 117 | 128 |
| Be32-O42-Be52 | 119 | 123 | 121 | 102 | 121 | 123 | 124 | 102 |
| O51/S-Be52-O42 | 121 | 120 | 119 | 124 | 118 | 121 | 119 | 124 |
| O51/S-Be61 -O61 | 118 | 118 | 122 | 118 | 118 | 118 | 122 | 118 |
| Be51-O61-Be61 | 113 | 119 | 121 | 119 | 113 | 118 | 121 | 118 |
| O61-Be51-O41 | 121 | 120 | 123 | 120 | 118 | 121 | 123 | 121 |
| Be41-O41-Be51 | 116 | 117 | 123 | 118 | 115 | 117 | 120 | 118 |
| O62-Be52-O42 | 119 | 120 | 122 | 116 | 121 | 120 | 121 | 116 |
| **Bond length** | **C(I)** | **C(II)** | **C(III)** | **C(IV)** | **D(I)** | **D(II)** | **D(III)** | **D(IV)** |
| Be41/Al-O41 | 1.55 | 1.80 | 1.55 | 1.80 | 1.56 | 1.84 | 1.55 | 1.86 |
| Be41/Al -O31 | 1.55 | 1.80 | 1.54 | 1.80 | 1.54 | 1.82 | 1.54 | 1.84 |
| Be32-O31 | 1.55 | 1.56 | 1.54 | 1.55 | 1.56 | 1.56 | 1.55 | 1.57 |
| Be42- O42/AL & S | 1.55 | 1.56 | 1.55 | 2.06 | 1.56 | 1.56 | 1.55 | 2.10 |
| Be52 -O42/Al & S | 1.55 | 1.55 | 1.55 | 2.02 | 1.56 | 1.54 | 1.55 | 2.04 |
| Be52-O51/S | 1.54 | 1.55 | 2.04 | 1.55 | 1.55 | 1.56 | 2.06 | 1.57 |
| Be61-O51/S | 1.59 | 1.56 | 2.09 | 1.56 | 1.55 | 1.55 | 2.05 | 1.56 |
| Be61-O61 | 1.60 | 1.55 | 1.60 | 1.55 | 1.55 | 1.55 | 1.57 | 1.55 |
| Be51-O61 | 1.53 | 1.55 | 1.54 | 1.55 | 1.55 | 1.55 | 1.56 | 1.55 |
| Be51-O41 | 1.55 | 1.57 | 1.56 | 1.55 | 1.54 | 1.56 | 1.56 | 1.55 |
| Be52-O62 | 1.55 | 1.54 | 1.54 | 1.54 | 1.55 | 1.55 | 1.56 | 1.55 |
| **Bond angle** |  |  |  |  |  |  |  |  |
| O41-Be41/Al-O31 | 117 | 104 | 118 | 103 | 120 | 104 | 118 | 103 |
| O31—Be41/Al-O51/S | 120 | 107 | 116 | 111 | 120 | 104 | 116 | 111 |
| Be52-O51/S-Be41/AL | 113 | 122 | 88 | 127 | 116 | 124 | 92 | 125 |
| O31-Be32-O42 | 118 | 120 | 120 | 123 | 120 | 121 | 121 | 126 |
| Be41-O31-Be32 | 114 | 122 | 117 | 128 | 116 | 126 | 119 | 126 |
| Be32-O42-Be52 | 120 | 123 | 123 | 102 | 118 | 121 | 123 | 97 |
| O51/S-Be52-O42 | 119 | 120 | 119 | 124 | 120 | 123 | 119 | 127 |
| O51/S-Be61 -O61 | 113 | 117 | 117 | 118 | 1118 | 119 | 123 | 120 |
| Be51-O61-Be61 | 117 | 119 | 126 | 119 | 116 | 120 | 116 | 118 |
| O61-Be51-O41 | 121 | 121 | 124 | 121 | 120 | 120 | 124 | 121 |
| Be41-O41-Be51 | 120 | 116 | 125 | 118 | 1115 | 118 | 120 | 118 |
| O62-Be52-O42 | 119 | 121 | 121 | 116 | 120 | 120 | 121 | 114 |

**Table S1** Bond length and bond angle of HCN adsorption on the pristine, Al, S and AlS- doped BeONTs (A to D) models

**Table S2**  Quantum descriptor of pristine, Al-doped (a), S-doped(b),AlS-doped(c) before HCN adsorption .

|  |  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |
| **Name** | **pristine** | **a**  **α spin** | **a**  **β spin** | **b** | | **c**  **α spin** | | **c**  **β spin** | |
| **E HOMO(ev)** | -7.71 | -4.90 | -7.74 | | -7.74 | | -4.95 | -7.77 |
| **E LUMO(ev)** | -0.67 | -0.66 | -1.98 | | -0.7 | | -0.68 | -1.99 |
| **E gap** | 7.04 | 4.24 | 5.76 | | 7.04 | | 4.27 | 5.78 |
| **E Fermi** | -4.19 | -2.78 | -4.86 | | -4.22 | | -2.81 | -4.88 |
| **I** | 7.71 | 4.90 | 7.74 | | 7.74 | | 4.95 | 7.77 |
| **A** | 0.67 | 0.66 | 1.98 | | 0.7 | | 0.68 | 1.99 |
| **ɳ** | 3.52 | 2.12 | 2.88 | | 3.52 | | 2.13 | 2.89 |
| **S** | 0.14 | 0.23 | 0.17 | | 0.14 | | 0.23 | 0.17 |
| **µ** | -4.19 | -2.78 | -4.86 | | -4.22 | | -2.81 | -4.88 |
| **Δϕ** | -3.52 | -2.12 | -2.88 | | -3.52 | | -2.13 | -2.89 |
| **ΔN** | 1.19 | 1.31 | 1.69 | | 1.2 | | 1.32 | 1.69 |
| **W** | 2.49 | 1.82 | 4.52 | | 2.53 | | 1.85 | 4.12 |
| **Χ** | 4.19 | 2.78 | 4.86 | | 4.22 | | 2.81 | 4.88 |

**Fig. S1** Plots of adsorption energy, NBO and dipole moment for adsorption HCN gas on the surface of pristine and Al, S and AlS doped of (4, 4) armchair model of BeONTs for (AI to DIV).

**Table S3** NBO parameters of HCN adsorption on the pristine, Al, S and AlS- doped BeONTs A models

|  |  |  |  |  |  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |
| **Structure** | |  |  | **Model A(I)** | **Donor(i)** |  | **Acceptor(j)** | **E2(Kcal/mol)** | | | **Ej-Ei** | | **F(i,j)** |
|  | | σ Be32-O31 | → | σ\* Be21-O21 | | 3.65 | 1.02 | | 0.054 | |
| σ Be21-O21 | → | σ\* Be41-O51 | | 2.45 | 1.03 | | 0.045 | |
| σ Be52-O51 | → | σ\*Be41-O41 | | 1.87 | 1 | | 0.039 | |
| σ Be52-O51 | → | σ\*Be41-O51 | | 4.33 | 1.01 | | 0.059 | |
| σ Be72-O62 | → | n\* Be62 | | 1.68 | 0.76 | | 0.033 | |
| σ Be22-O22 | → | σ\* Be12-O11 | | 1.29 | 0.98 | | 0.032 | |
|  | |  | | σ C81-N82 | → | σ \*Be41-N82 | | 1.68 | 1.37 | | 0.044 | |
|  | |  | | σ C81-H83 | → | σ \*Be41-N82 | | 3.91 | 0.98 | | 0.056 | |
|  | | | | **Model A(II)** | σ Be52-O51 | → | n\*AL | | 0.24 | 1.2 | | 0.021 | |
| σ Be72-O62 | → | n\* Be62 | | 1.45 | 1.12 | | 0.051 | |
| σ Be22-O22 | → | σ\* Be12-O11 | | 2.32 | 1.38 | | 0.073 | |
| σ AL-N82 | → | σ\*C81-N82 | | 0.68 | 1.35 | | 0.038 | |
| σ C81-N82 | → | σ \*Be41-N82 | | 0.58 | 1.21 | | 0.034 | |
| σ C81-H83 | → | σ \*Be41-N82 | | 2.65 | 1.62 | | 0.057 | |
|  | **Model** **A(III)** | | |  | σ Be32-O31 | → | σ\* Be21-O21 | | 2.93 | 1.07 | | 0.050 | |
|  | σ Be21-O21 | → | σ\* Be41-O51 | | 0.92 | 0.9 | | 0.026 | |
|  | σ Be52-O51 | → | σ\*Be41-O41 | | 3.28 | 0.91 | | 0.053 | |
|  | σ Be52-O51 | → | σ\*Be41-O51 | | 5.82 | 0.71 | | 0.058 | |
|  | σ Be72-O62 | → | n\* Be62 | | 1.35 | 0.73 | | 0.029 | |
|  | σ Be22-O22 | → | σ\* Be12-O11 | | 1.76 | 0.933 | | 0.036 | |
|  | σ C81-N82 | → | σ \*C81-H83 | | 2.05 | 1.47 | | 0.049 | |
|  | σ C81-H83 | → | σ \*C81-N82 | | 2.83 | 1.62 | | 0.57 | |
| **Model A(IV)** | | | | | σ Be52-O51 | → | n\*AL | | 0.11 | 1.11 | | 0.024 | |
| σ Be72-O62 | → | n\* Be62 | | 1.42 | 1.23 | | 0.068 | |
| σ Be22-O22 | → | σ\* Be12-O11 | | 1.95 | 1.2 | | 0.024 | |
| σ AL-N82 | → | σ\*C81-N82 | | 0.62 | 0.98 | | 0.066 | |
| σ AL-N82 | → | σ\* C81-H83 | | 1.98 | 0.097 | | 0.074 | |
| σ C81-N82 | → | σ \*Be41-N82 | | 0.56 | 1.19 | | 0.028 | |
| σ C81-H83 | → | σ \*Be41-N82 | | 2.34 | 1.57 | | 0.055 | |

**Table S4** NBO parameters of HCN adsorption on the pristine, Al, S and AlS- doped BeONTs B models

|  |  |  |  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |
| Structure | |  |  | | **Model B(I)** | Donor(i) |  | Acceptor(j) | E2(Kcal/mol) | Ej-Ei | F(i,j) |
|  | | | σ Be32-O31 | → | σ\* Be21-O21 | 3.49 | 0.97 | 0.052 |
| σ Be21-O21 | → | σ\* Be41-O51 | 0.75 | 1 | 0.025 |
| σ Be52-O51 | → | σ\*Be41-O51 | 3.75 | 1 | 0.055 |
| σ Be72-O62 | → | n\* Be62 | 1.95 | 0.75 | 0.035 |
| σ Be22-O22 | → | σ\* Be12-O11 | 1.45 | 0.92 | 0.033 |
|  | |  | | | σ C81-N82 | → | σ \* C81-H83 | 2.20 | 1.53 | 0.052 |
|  | |  | | | σ C81-H83 | → | σ \* C81-N82 | 2.32 | 1.51 | 0.053 |
|  | | | | **Model B(II)** | | σ Be32-O31 | → | σ\* Be21-O21 | 0.22 | 1.35 | 0.022 |
| σ Be21-O21 | → | n\*AL | 0.1 | 1.18 | 0.014 |
| σ Be52-O51 | → | n\*AL | 2.69 | 1.11 | 0.069 |
| σ Be72-O62 | → | n\* Be62 | 1.54 | 1.14 | 0.053 |
| σ Be22-O22 | → | σ\* Be12-O11 | 2.33 | 1.33 | 0.073 |
| σ C81-N82 | → | σ \*C81-H83 | 0.99 | 1.38 | 0.047 |
| σ C81-H83 | → | σ \*C81-N82 | 0.97 | 1.37 | 0.046 |
|  | **Model B(III)** | | |  | | σ Be32-O31 | → | σ\* Be21-O21 | 3.02 | 1.32 | 0.050 |
|  | | σ Be21-O21 | → | σ\* Be41-O51 | 0.34 | 0.87 | 0.051 |
|  | | σ Be52-O51 | → | σ\*Be41-O51 | 3.54 | 0.94 | 0.053 |
|  | | σ Be72-O62 | → | n\* Be62 | 0.46 | 1.58 | 0.027 |
|  | | σ Be22-O22 | → | σ\* Be12-O11 | 1.02 | 0.93 | 0.036 |
|  | | σ Be32-N82 | → | σ \*C81-H83 | 4.46 | 1.01 | 0.088 |
|  | | σ Be32-H83 | → | σ \*C81-N82 | 3.21 | 1.36 | 0.103 |
| **Model B(IV)** | | | | | | σ Be32-O31  σ Be21-O21 | → | σ\* Be21-O21  n\*AL | 0.2  0.12 | 1.4  1.19 | 0.021  0.015 |
| σ Be52-O51 | → | n\*AL | 2.55 | 1.15 | 0.069 |
| σ Be72-O62 | → | n\* Be62 | 1.65 | 1.14 | 0.055 |
| σ C81-N82 | → | σ \*C81-H83 | 1.07 | 1.49 | 0.051 |
| σ C81-H83 | → | σ \*C81-N82 | 1.15 | 1.56 | 0.054 |

**Table S5** NBO parameters of HCN adsorption on the pristine, Al, S and AlS- doped BeONTs C models

|  |  |  |  |  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |
| **Structure** | |  |  | | **Model CI** | **Donor(i)** |  | **Acceptor(j)** | **E2(Kcal/mol)** | | **Ej-Ei** | **F(i,j)** |
|  | | | σ Be32-O31 | → | σ\* Be21-O21 | | 3.26 | 1.03 | 0.055 |
| σ Be21-O21 | → | σ\* Be41-O51 | | 0.78 | 1.1 | 0.026 |
| σ Be52-O51 | → | σ\*Be41-O41 | | 3.47 | 1.02 | 0.053 |
| σ Be52-O51 | → | σ\*Be41-O51 | | 4.11 | 1.01 | 0.057 |
| σ Be72-O62 | → | n\* Be62 | | 1.78 | 0.75 | 0.034 |
| σ Be22-O22 | → | σ\* Be12-O11 | | 1.61 | 0.96 | 0.035 |
|  | |  | | | σ C81-N82 | → | σ \*C81-H83 | | 2.05 | 1.47 | 0.049 |
|  | |  | | | σ C81-H83 | → | σ \*C81-N82 | | 2.51 | 1.62 | 0.057 |
|  | | | | **Model CII** | | σ Be32-O31 | → | σ\* Be21-O21 | | 0.03 | 1.43 | 0.009 |
| σ Be21-O21 | → | n\*AL | | 0.1 | 1.2 | 0.014 |
| σ Be52-O51 | → | n\*AL | | 0.21 | 1.2 | 0.014 |
| σ Be72-O62 | → | n\* Be62 | | 2.08 | 1.36 | 0.067 |
| σ Be22-O22 | → | σ\* Be12-O11 | | 2.41 | 1.38 | 0.075 |
| σ AL-C81 | → | σ\*Be52-O51 | | 0.59 | 0.8 | 0.008 |
| σ C81-N82 | → | σ \*C81H83 | | 0.32 | 1.37 | 0.027 |
| σ C81-H83 | → | σ \*C81-N82 | | 0.41 | 1.23 | 0.028 |
|  | **Model CIII** | | |  | | σ Be32-O31 | → | σ\* Be21-O21 | | 3.19 | 1.04 | 0.051 |
|  | | σ Be21-O21 | → | σ\* Be41-O51 | | 0.76 | 0.91 | 0.023 |
|  | | σ Be52-O51 | → | σ\*Be41-O51 | | 4.69 | 0.77 | 0.056 |
|  | | σ Be52-O51 | → | σ\*Be41-O41 | | 4.39 | 0.58 | 0.046 |
|  | | σ Be72-O62 | → | n\* Be62 | | 1.06 | 0.73 | 0.026 |
|  | | σ Be22-O22 | → | σ\* Be12-O11 | | 1.71 | 0.96 | 0.036 |
|  | | σ C81-N82 | → | σ \*C81-H83 | | 2.04 | 1.47 | 0.049 |
|  | | σ C81-H83 | → | σ \*C81-N82 | | 2.53 | 1.61 | 0.057 |
|  | | | | | | σ Be21-O21 |  | n\*AL | | 0.34 | 1.24 | 0.026 |
| **Model CIV** | | | | | | σ Be52-O51 | → | n\*AL | | 0.03 | 1.22 | 0.008 |
| σ Be72-O62 | → | n\* Be62 | | 1.64 | 1.37 | 0.060 |
| σ AL-N82 | → | σ\* Be52-O51 | | 0.20 | 0.89 | 0.017 |
| σ C81-N82 | → | σ \*Al63-N82 | | 0.57 | 1.2 | 0.034 |
| σ C81-H83 | → | σ \*Al63-N82 | | 2.7 | 0.76 | 0.058 |

**Table S6** NBO parameters of HCN adsorption on the pristine, Al, S and AlS- doped BeONTs D models

|  |  |  |  |  |  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |
| **Structure** | |  |  | | **Model D(I)** | **Donor(i)** |  | **Acceptor(j)** | **E2(Kcal/mol)** | | | **Ej-Ei** | **F(i,j)** |
|  | | | σ Be32-O31 | → | σ\* Be21-O21 | | 3.70 | 1.03 | | 0.055 |
| σ Be21-O21 | → | σ\* Be41-O51 | | 1.03 | 1.05 | | 0.029 |
| σ Be52-O51 | → | σ\*Be41-O41 | | 3.23 | 1.02 | | 0.051 |
| σ Be52-O51 | → | σ\*Be41-O51 | | 4.86 | 1.06 | | 0.064 |
| σ Be72-O62 | → | n\* Be62 | | 2.19 | 0.75 | | 0.037 |
| σ Be22-O22 | → | σ\* Be12-O11 | | 1.81 | 0.96 | | 0.037 |
|  | |  | | | σ C81-N82 | → | σ \*C81-H83 | | 2.72 | 1.52 | | 0.057 |
|  | |  | | | σ C81-H83 | → | σ \*C81-N82 | | 4.44 | 1.56 | | 0.075 |
|  | | | | **Model D(II)** | | σ Be32-O31 | → | σ\* Be21-O21 | | 0.11 | 1.43 | | 0.009 |
| σ Be21-O21 | → | n\*AL | | 0.42 | 1.2 | | 0.014 |
| σ Be52-O51 | → | n\*AL | | 0.34 | 1.2 | | 0.014 |
| σ Be72-O62 | → | n\* Be62 | | 2.11 | 1.36 | | 0.067 |
| σ Be22-O22 | → | σ\* Be12-O11 | | 2.41 | 1.38 | | 0.075 |
| σ AL-C81 | → | σ\*Be52-O51 | | 0.67 | 0.8 | | 0.008 |
| σ C81-N82 | → | σ \*C81H83 | | 1.32 | 1.37 | | 0.027 |
| σ C81-H83 | → | σ \*C81-N82 | | 2.1 | 1.23 | | 0.028 |
|  | **Model D(III)** | | |  | | σ Be32-O31 | → | σ\* Be21-O21 | | 3.56 | 1.01 | | 0.053 |
|  | | σ Be21-O21 | → | σ\* Be41-O51 | | 0.98 | 1.05 | | 0.026 |
|  | | σ Be52-O51 | → | σ\*Be41-O41 | | 3.15 | 0.9 | | 0.051 |
|  | | σ Be52-O51 | → | σ\*Be41-O51 | | 4.65 | 1.03 | | 0.057 |
|  | | σ Be72-O62 | → | n\* Be62 | | 1.9 | 0.72 | | 0.033 |
|  | | σ Be22-O22 | → | σ\* Be12-O11 | | 1.54 | 0.96 | | 0.037 |
|  | | σ C81-N82 | → | σ \*C81-H83 | | 2.43 | 1.49 | | 0.055 |
|  | | σ C81-H83 | → | σ \*C81-N82 | | 4.32 | 1.52 | | 0.072 |
|  | | | |  | | σ Be21-O21 |  | n\*AL | | 0.11 | 1.18 | | 0.015 |
| **Model**  **D(IV)** | | | | | | σ Be52-O51 | → | n\*AL | | 4.28 | 1.15 | | 0.089 |
| σ Be72-O62 | → | n\* Be62 | | 2.02 | 1.15 | | 0.061 |
| σ C81-N82 | → | σ \*C81-H83 | | 1.30 | 1.51 | | 0.056 |
| σ C81-H83 | → | σ \*C81-N82 | | 2.03 | 1.56 | | 0.071 |