Supplementary information for

**Interfacial adsorption mechanism of diethyldithiocarbamate in high-sulfur residue flotation**

Hong Liu[1], Jing He[1], Tao Luo[1], Jie Dai[1], Shuqiong Cao[1], Shenghai Yang[1][2], Chaobo Tang[1], Changhong Wang\*[1], Yongming Chen\*[1][2]

[1] School of Metallurgy and Environment, Central South University, Changsha, Hunan, 410083, China

[2] Hunan Provincial Key Laboratory of Nonferrous Value-added Metallurgy, Changsha, 410083, Hunan, China

\*Correspondence: Changhong.Wang@csu.edu.cn; csuchenyongming@163.com

**The SI contains 2 figures, 1 table and 1 text section.**



Figure S1 Electrostatic potential distribution of DDTC.

Gaussian parameters: m062x/6-311++g(2d,p), em=gd3, scrf=(solvent=water,iefpcm)



Figure S2 a) HOMO orbitals of DDTC; b) LOMO orbitals of DDTC.

Gaussian parameters: m062x/6-311++g(2d,p), em=gd3, scrf=(solvent=water,iefpcm)

Table S1 INCAR file for VASP optimization process

|  |  |  |
| --- | --- | --- |
| INCAR | Keywords | Parameters |
| Basic parameters | ISTART | 0 |
| ICHARG | 2 |
| ISPIN | 1 |
| ENCUT | 400 |
| PREC | Normal |
| Electronic relaxation | ISMEAR | 0 |
| SIGMA | 0.05 |
| NELM | 200 |
| NELMIN | 3 |
| EDIFF | 10-5 |
| Ionic relaxation | NSW | 100 |
| IBRION | 2 |
| POTIM | 0.5 |
| ISIF | 2 |
| EDIFFG | -0.05 |

**Supplemental DFT calculation Information**

**S1. Electrostatic potential and frontline orbit of DDTC (Related to Figure S1 and Figure S2)**

Figure S1 showed the electrostatic potential on the surface of DDTC. The polar group sulfur atom was the region of negative potential concentration of DDTC. An electrostatic potential minimum of the DDTC existed in the middle of the two sulfur atoms (-155.14 kcal/mol). Two extreme points of electric potential existed around the two sulfur atoms (-108.31kcal/mol), which were symmetrically distributed. This meant that the sulfur atoms in DDTC were much likely to adsorb on the surface of the mineral phase.

Figure S2 showed the HOMO and LOMO orbitals of DDTC. The HOMO was mainly composed of sulfur atoms in polar groups and the LOMO was mainly composed of two ethyl groups. In the flotation process, the collector usually acted as the electron donor and its HOMO orbitals usually transferred electrons to the mineral surface. Therefore, the sulfur atom in DDTC was the active site for chemisorption.