

Toward Enhanced Combustion Gas Monitoring and Capture: Sulfur Vacancies as Vectors for Selective CO and CO₂ Intercalation in MoS₂

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Supplementary Information

1. Representative Quantum ESPRESSO Input File (RELAX, 6xCO 3x3 MoS₂)

```
&control
calculation='relax',
prefix='QE'
pseudo_dir= ./
outdir = 'files/'
wf_collect=.true.
etot_conv_thr=1.0D-4,
forc_conv_thr=1.0D-3,
! restart_mode = 'restart'
! max_seconds=53900
/
&system
ibrav= 4, a=9.483 c=32
nat= 66, ntyp= 4,
ecutwfc = 50, ecutrho=500,
occupations= 'smearing', smearing='gauss', degauss= 0.01
vdw_corr='DFT-D3'
/
&electrons
electron_maxstep= 150
conv_thr= 1.0d-8
diagonalization='david'
mixing_mode='local-TF'
/
&IONS
ion_dynamics='bfgs',
/
ATOMIC_SPECIES
Mo 95.950 Mo.pbe-spn-rrkjus_psl.1.0.0.UPF
S 32.06 S.pbe-n-rrkjus_psl.1.0.0.UPF
C 12.011 C.pbe-n-rrkjus_psl.1.0.0.UPF
O 15.999 O.pbe-n-rrkjus_psl.1.0.0.UPF
ATOMIC_POSITIONS (angstrom)
```

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C 3.161 4.911015 6.1475
 O 3.161 6.039015 6.1475
 C 0.0 4.911015 6.1475
 O 0.0 6.039015 6.1475
 C 6.322 4.911015 6.1475
 O 6.322 6.039015 6.1475
 C 4.74150 3.301506 6.1475
 O 4.74150 2.173506 6.1475
 C 1.5805 3.301506 6.1475
 O 1.5805 2.173506 6.1475
 C -1.5805 3.301506 6.1475
 O -1.5805 2.173506 6.1475
 Mo 0.000000 1.825004 3.073750
 Mo 4.741500 6.387515 9.221250
 Mo 3.161000 7.300017 3.073750
 Mo 1.580500 0.912502 9.221250
 Mo -3.161000 7.300017 3.073750
 Mo 7.902500 0.912502 9.221250
 Mo -1.580500 4.562510 3.073750
 Mo 6.322000 3.650008 9.221250
 Mo 1.580500 4.562510 3.073750
 Mo 3.161000 3.650008 9.221250
 Mo 0.000000 7.300017 3.073750
 Mo 4.741500 0.912502 9.221250
 Mo 3.161000 1.825004 3.073750
 Mo 1.580500 6.387515 9.221250
 Mo 6.322000 1.825004 3.073750
 Mo -1.580500 6.387515 9.221250
 Mo 4.741500 4.562510 3.073750
 Mo -0.000000 3.650008 9.221250
 S 0.000000 1.825004 7.715113
 S 4.741500 6.387515 4.579888
 S 3.161000 7.300017 7.715113
 S 1.580500 0.912502 4.579888
 S -3.161000 7.300017 7.715113
 S 7.902500 0.912502 4.579888
 S 4.741500 6.387515 1.567612
 S 0.000000 1.825004 10.727388
 S 1.580500 0.912502 1.567612
 S 3.161000 7.300017 10.727388
 S 7.902500 0.912502 1.567612
 S -3.161000 7.300017 10.727388
 S -1.580500 4.562510 7.715113
 S 6.322000 3.650008 4.579888
 S 1.580500 4.562510 7.715113
 S 3.161000 3.650008 4.579888
 S 0.000000 7.300017 7.715113
 S 4.741500 0.912502 4.579888
 S 6.322000 3.650008 1.567612
 S -1.580500 4.562510 10.727388
 S 3.161000 3.650008 1.567612
 S 1.580500 4.562510 10.727388
 S 4.741500 0.912502 1.567612
 S 0.000000 7.300017 10.727388
 S 3.161000 1.825004 7.715113
 S 1.580500 6.387515 4.579888
 S 6.322000 1.825004 7.715113

S -1.580500 6.387515 4.579888
 S 4.741500 4.562510 7.715113
 S -0.000000 3.650008 4.579888
 S 1.580500 6.387515 1.567612
 S 3.161000 1.825004 10.727388
 S -1.580500 6.387515 1.567612
 S 6.322000 1.825004 10.727388
 S -0.000000 3.650008 1.567612
 S 4.741500 4.562510 10.727388
 K_POINTS automatic
 4 4 1 0 0 0

2. Optimized atomic coordinates of lowest energy configurations

2.1. $6xCO\ 3x3\ MoS_2$

C 3.1573272396 5.5935955665 6.5088109601
 O 3.1658423229 6.5786355175 5.9359512540
 C 0.0000000000 5.6071278921 6.4920649444
 O 0.0000000000 6.5958209934 5.9254837760
 C 6.3256727604 5.5935955665 6.5088109601
 O 6.3171576771 6.5786355175 5.9359512540
 C 4.7415000000 2.6126753480 5.8000516808
 O 4.7415000000 1.6226294964 6.3643257443
 C 1.5863549771 2.6144235561 5.7850000234
 O 1.5745830175 1.6327480157 6.3636543620
 C -1.5863549771 2.6144235561 5.7850000234
 O -1.5745830175 1.6327480157 6.3636543620
 Mo 0.0000000000 1.7500606707 1.1064034777
 Mo 4.7415000000 6.4624192928 11.1887873820
 Mo 3.1610247357 7.2237974473 1.1011142313
 Mo 1.5804892657 0.9886738984 11.1938787397
 Mo -3.1610247357 7.2237974473 1.1011142313
 Mo 7.9025107343 0.9886738984 11.1938787397
 Mo -1.5805439037 4.4862717371 1.1009163736
 Mo 6.3220325539 3.7261623107 11.1941769071
 Mo 1.5805439037 4.4862717371 1.1009163736
 Mo 3.1609674461 3.7261623107 11.1941769071
 Mo 0.0000000000 7.2238736220 1.1012462951
 Mo 4.7415000000 0.9885152376 11.1938456812
 Mo 3.1609945009 1.7500972095 1.1065738542
 Mo 1.5805318254 6.4623328589 11.1884712956
 Mo 6.3220054991 1.7500972095 1.1065738542
 Mo -1.5805318254 6.4623328589 11.1884712956
 Mo 4.7415000000 4.4862912209 1.1007186030
 Mo 0.0000000000 3.7261939894 11.1941638715
 S 0.0000000000 1.9005837316 9.6255723761
 S 4.7415000000 6.3118686343 2.6693877773
 S 3.1611328038 7.3787301461 9.6205466195
 S 1.5804175603 0.8337098804 2.6745529906
 S -3.1611328038 7.3787301461 9.6205466195
 S 7.9025824397 0.8337098804 2.6745529906
 S 4.7415000000 6.3112973962 -0.4698018333
 S 0.0000000000 1.9012274521 12.7647108280
 S 1.5805851142 0.8391736297 -0.4634718704
 S 3.1608700043 7.3733637191 12.7585594514
 S 7.9024148858 0.8391736297 -0.4634718704

S -3.1608700043 7.3733637191 12.7585594514
 S -1.5803455779 4.6352055495 9.6229440928
 S 6.3217579156 3.5772858228 2.6719847606
 S 1.5803455779 4.6352055495 9.6229440928
 S 3.1612420844 3.5772858228 2.6719847606
 S 0.0000000000 7.3784983880 9.6202892284
 S 4.7415000000 0.8338717934 2.6747615368
 S 6.3220304823 3.5720243490 -0.4681312342
 S -1.5804713667 4.6403913044 12.7631242121
 S 3.1609695177 3.5720243490 -0.4681312342
 S 1.5804713667 4.6403913044 12.7631242121
 S 4.7415000000 0.8392659402 -0.4632956573
 S 0.0000000000 7.3731130188 12.7583353928
 S 3.1610274480 1.9004535350 9.6256859838
 S 1.5805826290 6.3119763795 2.6693759914
 S 6.3219725520 1.9004535350 9.6256859838
 S -1.5805826290 6.3119763795 2.6693759914
 S 4.7415000000 4.6353494414 9.6232238178
 S 0.0000000000 3.5771578123 2.6719438207
 S 1.5804590329 6.3112962807 -0.4696859237
 S 3.1610221230 1.9010771305 12.7647628857
 S -1.5804590329 6.3112962807 -0.4696859237
 S 6.3219778770 1.9010771305 12.7647628857
 S 0.0000000000 3.5721650041 -0.4681289899
 S 4.7415000000 4.6402719357 12.7633273251

2.2. $6xCO_2$ $3x3 MoS_2$

C 7.3164629981 5.9874075926 6.3696177800
 O 6.8440430227 5.4812246155 5.4212026736
 O 7.7731126185 6.4952039831 7.3208466970
 C 4.1143096507 6.6982420992 6.6036589388
 O 3.4492386144 5.7514652070 6.4079549680
 O 4.7862540476 7.6400010744 6.7976674070
 C 5.3908923322 2.9952651524 6.1733535954
 O 4.9436839201 2.6516977353 5.1459226783
 O 5.8372186603 3.3352102904 7.2033816669
 C 0.8757800352 7.2702946606 5.9659555053
 O 0.5188128518 6.4036345346 5.2632952952
 O 1.2402767006 8.1394393481 6.6669578445
 C 2.4598424144 1.3006412115 5.7230196130
 O 2.0778823482 2.2596940010 6.2782996088
 O 2.8486947137 0.3417504494 5.1711722841
 C 8.7778615958 2.9155023546 6.2984016804
 O 8.4403881672 2.0867694651 5.5384793477
 O 9.0992215571 3.7502205904 7.0557303335
 S 4.7983334953 6.4377945303 -0.9293111647
 S 1.6343191532 0.9655819782 -0.9302083581
 S 7.9546692619 0.9675506579 -0.9344529572
 S 6.3746877609 3.7058856379 -0.9296688681
 S 3.2175570320 3.7016885678 -0.9282328408
 S 4.7941405669 0.9674919727 -0.9321828401
 S 1.6367726341 6.4389031751 -0.9355264486
 S -1.5283185650 6.4413845838 -0.9331681523
 S 0.0536184987 3.7047967674 -0.9329757230
 Mo 0.0505652541 1.8802857985 0.6363805284

Mo 3.2169286956 7.3540654096 0.6344308109
 Mo -3.1068459483 7.3529071281 0.6411454600
 Mo -1.5256918506 4.6174885580 0.6388479072
 Mo 1.6353316231 4.6145553669 0.6377639401
 Mo 0.0548918931 7.3525850410 0.6344008493
 Mo 3.2150730442 1.8779698187 0.6417992013
 Mo 6.3757779877 1.8787939553 0.6385343388
 Mo 4.7959192618 4.6152263234 0.6448095878
 S 4.7938649223 6.4429254962 2.2137541492
 S 1.6265800765 0.9694922975 2.2037928541
 S 7.9550087034 0.9643051197 2.2077727401
 S 6.3806691831 3.7032236111 2.2099337687
 S 3.2123209423 3.7033492494 2.2136066275
 S 4.7984767298 0.9604678419 2.2067206375
 S 1.6393916952 6.4418042624 2.1996470503
 S -1.5229881083 6.4422279954 2.2074058233
 S 0.0539613731 3.7041658529 2.2106718158
 S -0.0948841156 1.8129977950 10.1191354918
 S 3.0662046350 7.2884778998 10.1225117959
 S -3.2570066509 7.2893809782 10.1272944220
 S -1.6769412118 4.5509047318 10.1263621857
 S 1.4863058530 4.5533865048 10.1178727422
 S -0.0939312488 7.2891530796 10.1249831478
 S 3.0684321023 1.8136648901 10.1183104503
 S 6.2282406759 1.8101071925 10.1259577852
 S 4.6470586947 4.5552089240 10.1238127013
 Mo 4.6467381434 6.3776356687 11.6956313681
 Mo 1.4863911514 0.9028237787 11.6909214819
 Mo 7.8087566956 0.9012160391 11.6928863020
 Mo 6.2273220404 3.6376048322 11.6932556996
 Mo 3.0660966517 3.6402655149 11.6896770415
 Mo 4.6472590648 0.9007142080 11.6939841745
 Mo 1.4853502700 6.3756407388 11.6925797802
 Mo -1.6755511012 6.3761450277 11.6938861039
 Mo -0.0935803453 3.6390111490 11.6896721243
 S -0.0931185499 1.8150914633 13.2611364977
 S 3.0649802570 7.2892792180 13.2637674976
 S -3.2554218908 7.2919359585 13.2644627586
 S -1.6738213755 4.5493799994 13.2632356044
 S 1.4863261725 4.5505975245 13.2616347860
 S -0.0950208589 7.2877377787 13.2641848530
 S 3.0662649454 1.8163877024 13.2607830802
 S 6.2283858114 1.8126358348 13.2648097766
 S 4.6465804887 4.5508270666 13.2627143589

2.3. *vac - 8xCO 3x3 MoS₂*

C 0.6005849093 7.3046405825 5.9373478155
 O 1.3334609332 7.2644706656 6.8089524992
 C 6.0565487761 2.3004362348 6.5013660696
 O 7.1362304219 2.5859481577 6.2792468804
 C -2.6232700311 6.2702757826 5.6454491074
 O -2.2566081212 5.6293791278 6.5136392138
 C 0.3593660044 4.1226912894 5.7016025521
 O 0.8552014053 3.5162568827 6.5299632502
 C 3.5519042098 5.3057116638 5.7366271261
 O 4.0480920811 4.7750659991 6.6150851007

C 0.0176849635 0.5687848426 5.6466895914
 O -0.4790522341 -0.0053261633 6.4967498168
 C 3.1107569634 1.5674180756 5.7861319375
 O 2.9336406529 0.9187407776 6.7473322419
 C 3.2976062492 2.6472013169 1.8555547711
 O 3.2783495236 2.2193707871 4.8262648788
 Mo 0.1332844697 0.8224303969 1.0198760633
 Mo 4.7370181800 7.6807401382 11.4204699552
 Mo 3.2992976925 6.3032690138 1.0251929091
 Mo 1.5773496508 2.2041038790 11.4179898306
 Mo -3.0334216886 6.3070095220 1.0213081058
 Mo 7.8987743613 2.2041008707 11.4215909533
 Mo -1.4424770808 3.5497037226 1.0222290751
 Mo 6.3194752577 4.9444634869 11.4204337375
 Mo 1.7290511104 3.5561638988 0.9754445279
 Mo 3.1600117853 4.9402925649 11.4188083411
 Mo 0.1489748159 6.3066423373 1.0209164091
 Mo 4.7371094662 2.2041125867 11.4173126023
 Mo 3.2976537987 0.8384339240 0.9643189525
 Mo 1.5770189150 7.6789743103 11.4185318287
 Mo 6.4635842480 0.8221796842 1.0193624337
 Mo -1.5836427616 7.6799315843 11.4179842320
 Mo 4.8672553375 3.5548851946 0.9754234273
 Mo -0.0030868971 4.9401965265 11.4210333111
 S -0.0053385117 3.1149621162 9.8511087214
 S 4.8644175764 5.3592902346 2.5829272634
 S 3.1561260010 8.5935234303 9.8497301539
 S 1.7341862464 -0.0555065894 2.5739619543
 S -3.1674833092 8.5912421075 9.8497160196
 S 8.0390921980 -0.0879650409 2.5970653494
 S 4.8795846770 5.4109327641 -0.5563323793
 S -0.0021481903 3.1157049846 12.9907177755
 S 1.6954237932 -0.1014974854 -0.5641206030
 S 3.1560237413 8.5916343282 12.9903955198
 S 8.0400906855 -0.0901214715 -0.5518319023
 S -3.1636441612 8.5927542137 12.9886914840
 S -1.5810869638 5.8544597995 9.8509455107
 S 6.4289434129 2.6452666870 2.5821491590
 S 1.5785824190 5.8497989433 9.8526704170
 S -0.0027701696 8.5918947495 9.8484400390
 S 4.8616690182 -0.0547985595 2.5718400641
 S 6.4796651855 2.6399482615 -0.5575410358
 S -1.5827082624 5.8560811224 12.9899373906
 S 3.2972004761 2.6544770654 -0.6739091001
 S 1.5790042461 5.8531086735 12.9884868945
 S 4.9014798330 -0.1018494817 -0.5636859572
 S -0.0035494652 8.5924677225 12.9876710046
 S 3.1572007195 3.1162525533 9.8456941535
 S 1.7349562191 5.3622137600 2.5824041398
 S 6.3187732105 3.1195229636 9.8537400506
 S -1.4414860040 5.3882080648 2.5900639621
 S 4.7400389094 5.8579499917 9.8504284956
 S 0.1691662985 2.6449283183 2.5801007423
 S 1.7192057589 5.4111051969 -0.5568866123
 S 3.1584836171 3.1155627696 12.9880127553
 S -1.4427071456 5.3869604264 -0.5322543919
 S 6.3168100330 3.1173944081 12.9891300428

S 0.1163014666 2.6410850205 -0.5588602573
S 4.7382386843 5.8552866755 12.9900432740

2.4. *vac - 5xCO₂ 3x3 MoS₂*

C 2.4838109977 3.8544781494 7.6950496437
O 3.1088637915 4.8479165877 7.6952446324
O 1.8563418186 2.8651070114 7.6931560694
C 5.3703623918 6.7537216454 7.6180412892
O 4.6699437075 7.6810728861 7.7741695010
O 6.0781044314 5.8315930528 7.4529462194
C 8.2927686128 8.8535348330 7.0682409558
O 8.7745007048 9.3608752193 6.1281874523
O 7.8265541373 8.3225998132 8.0043663865
C 0.8043560665 6.6566928922 7.8369467675
O 0.1757417769 5.6664569082 7.7979296325
O 1.4237234644 7.6528138697 7.8653882678
C 6.6698537474 2.8144220661 7.7778738902
O 5.5306099025 2.8044466498 8.0631505727
O 7.8074558591 2.8192592860 7.4988824846
S 6.2329725497 1.6963403022 14.1954683601
S 4.6521095085 4.4335073330 14.1947925984
S -0.0877407519 1.6977083550 14.1922860300
S 3.0686908405 1.6963934461 14.1933205534
S 3.0708145478 7.1701270151 14.1962923992
S -3.2512712434 7.1736704801 14.1929731728
S 1.4877022742 4.4325265289 14.1927866745
S -1.6710983488 4.4317189941 14.1908415560
S -0.0921137062 7.1712885931 14.1945564676
Mo 7.8149923589 0.7818510123 12.6262060751
Mo 4.6502387229 0.7849297648 12.6254156738
Mo 6.2312889537 3.5207634183 12.6228038694
Mo 3.0711644149 3.5217914899 12.6249842618
Mo 4.6500327344 6.2590106051 12.6256051102
Mo 1.4897494629 0.7854847830 12.6198464509
Mo -0.0922799987 3.5200611314 12.6193829442
Mo 1.4904723933 6.2596444959 12.6255548484
Mo -1.6729026529 6.2572401388 12.6231387413
S 6.2355124047 1.6902324610 11.0616104672
S 1.4906033842 4.4349164761 11.0555580687
S -0.0928703301 7.1724492570 11.0603111069
S 4.6501524670 4.4360096066 11.0555039014
S -0.0922405761 1.6951305453 11.0486575709
S 3.0721061405 1.6998696284 11.0536913688
S 3.0710129639 7.1708421621 11.0546438594
S -1.6738373094 4.4339467341 11.0489521140
S -3.2531720432 7.1699265996 11.0547383302
S -0.4290995477 3.7070268806 4.3364264168
S 1.1198693129 6.5061604979 4.2956264905
S -1.9777971641 6.5089266486 4.2953546501
S 7.3995745206 0.9614745588 4.2990806566
S 1.2155572424 0.9649335028 4.2959419507
S 5.8511473430 3.6461164098 4.2975934891
S 2.7671879608 3.6485859918 4.2889338102
S 4.3085277890 6.4429081427 4.2783106259
Mo -2.0099759274 4.6173652232 2.7614564413
Mo 1.1477685455 4.6184255072 2.7568461625

Mo -0.4333559502 1.8825898632 2.7612081366
Mo 2.7073189063 7.3699685036 2.7248335339
Mo -3.5699148213 7.3691697875 2.7261475631
Mo 4.3113844561 4.5915533844 2.7249088457
Mo -0.4306715143 7.4027903922 2.6920446543
Mo 2.7680197804 1.8558553932 2.6917924366
Mo 5.8501710767 1.8553734388 2.6955441901
S -0.4317777678 3.7059699623 1.2041590947
S 4.3107969498 6.4427915696 1.1730836533
S -1.9893309840 6.4335482949 1.1603820382
S 1.1268881162 6.4355087967 1.1588755895
S 1.1497963234 0.9922330487 1.1609154402
S 7.4669230353 0.9940197791 1.1596728519
S 5.9122128633 3.6879801099 1.1619580128
S 2.7109410293 3.6886756396 1.1583140127
S 4.3104302238 0.9681231581 1.1186096871