Electronic Supplementary Information

The $Li_2Ti_6O_{13}$ and $Li_2Zr_6O_{13}$ Composite as High-Performance Anode for Alkali-Ion Batteries: A Molecular Dynamics Study

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Examples of LAMMPS input files (at 900K) of mono-, bi-crystalline $Li_2Zr_6O_{13}$ samples, and $Li_2Zr_6O_{13}@Li_2Ti_6O_{13}$ composite are included for reproducibility providing further simulation details for the readers. A zip file is included with the simulation boxes.

1. Input file of monocystalline $Li_2Zr_6O_{13}$

```
############################################################
#------------Variables and cell-------------------------#
############################################################

clear
units       metal       #eV,atomic charge,angstroms,ps,kelvin,bars,g/mol
dimension 3
boundary p p p
atom_style charge
#processors * * * grid numa
read_data MonoLZO.lmp

replicate 2 2 2

group       Li type 1
group       Zr type 2
group       O  type 3

mass 1   6.94000000       # Li
mass 2   91.22400000      # Zr
mass 3   15.99900000      # O
```

```
variable T1 equal 900
variable Timer equal step*dt
log ${T1}.lammps

##############################################################
#---------- Pair styles and electrostatics--------------#
##############################################################

pair_style buck/coul/long 10
pair_coeff * *  0.0 1.0 0.0
pair_coeff 1 2  0.0 1.0 0.0
pair_coeff 1 3  632.1018 0.2906  0.0
pair_coeff 2 3 1453.8 0.35000  0.0
pair_coeff 3 3  22764.3 0.149 27.627

kspace_style pppm 1e-05

##############################################################
# -------------- Run Minimization --------------------#
##############################################################

reset_timestep 0
timestep 0.5
thermo 10
thermo_style custom step enthalpy fmax lx ly lz vol press
min_style cg
minimize 1e-25 1e-25 5000 10000

##############################################################
# -------------- Relax Cell --------------------------#
##############################################################

reset_timestep 0
timestep 0.5
fix 1 all box/relax aniso 1.0 vmax 0.003
thermo 1
thermo_style custom step enthalpy fmax lx ly lz vol press
min_style cg
minimize 1e-25 1e-25 5000 10000
unfix 1

##############################################################
# ------------- Run NPT T1----------------------------#
##############################################################
```

```
reset_timestep 0
timestep 0.002
velocity all create ${T1} 4928 rot yes dist gaussian
fix     2 all npt temp ${T1} ${T1} 0.01 aniso 1.0 1.0 0.1
thermo_style custom step v_Timer cpu temp etotal fmax lx ly lz vol press
thermo 1000
run 5000
unfix 2

reset_timestep 0
timestep 0.002

compute      mymsdLi Li msd com yes
compute       mymsdO O msd com yes
variable     msdxLi equal "c_mymsdLi[1]"
variable     msdyLi equal "c_mymsdLi[2]"
variable     msdzLi equal "c_mymsdLi[3]"
variable     msdtotLi equal "c_mymsdLi[4]"
variable     msdoxO equal "c_mymsdO[1]"
variable     msdoyO equal "c_mymsdO[2]"
variable     msdozO equal "c_mymsdO[3]"
variable     msdototO equal "c_mymsdO[4]"

fix          msdT2 O ave/time 1 1 2500 v_msdoxO v_msdoyO v_msdozO v_msdototO
file msdOxygen${T1}
fix          msdT1 Li ave/time 1 1 2500 v_msdxLi v_msdyLi v_msdzLi v_msdtotLi file
msdLithium${T1}
fix          3 all nvt temp ${T1} ${T1} 0.01
thermo_style custom step v_Timer cpu temp etotal fmax lx ly lz vol press v_msdtotLi
v_msdototO
thermo 5000
dump dynamics all xyz 5000 monoLZO_${T1}.xyz
dump_modify dynamics every 5000 element Li Zr O first yes pbc yes
run          1000000
unfix 3
unfix msdT1
unfix msdT2
```

2. Input file of bi-crystalline Li$_2$Zr$_6$O$_{13}$

```
#############################################################
#------------Variables and cell-------------------------#
#############################################################

clear
units       metal       #eV,atomic charge,angstroms,ps,kelvin,bars,g/mol
dimension 3
```

```
boundary p p p
atom_style charge
#processors * * * grid numa
read_data Bi-crystal_LZO.lmp

group          Li type 1
group          Zr type 2
group          O  type 3

mass 1   6.94000000          # Li
mass 2   91.22400000          # Zr
mass 3   15.99900000          # O

variable T1 equal 900
variable Timer equal step*dt
log ${T1}.lammps


################################################################
#---------- Pair styles and electrostatics--------------#
################################################################

pair_style buck/coul/long 10
pair_coeff * *  0.0 1.0 0.0
pair_coeff 1 2  0.0 1.0 0.0
pair_coeff 1 3  632.1018 0.2906  0.0
pair_coeff 2 3 1453.8 0.35000  0.0
pair_coeff 3 3  22764.3 0.149 27.627

kspace_style pppm 1e-05


################################################################
# -------------- Run Minimization --------------------#
################################################################

reset_timestep 0
timestep 0.5
thermo 10
thermo_style custom step enthalpy fmax lx ly lz vol press
min_style cg
minimize 1e-25 1e-25 5000 10000


################################################################
# -------------- Relax Cell --------------------------#
################################################################
reset_timestep 0
timestep 0.5
```

```
fix 1 all box/relax aniso 1.0 vmax 0.003
thermo 1
thermo_style custom step enthalpy fmax lx ly lz vol press
min_style cg
minimize 1e-25 1e-25 5000 10000
unfix 1


############################################################
# -------------- Run NPT T1-----------------------------#
############################################################
reset_timestep 0
timestep 0.002
velocity all create ${T1} 4928 rot yes dist gaussian
fix     2 all npt temp ${T1} ${T1} 0.01 aniso 1.0 1.0 0.1
thermo_style custom step v_Timer cpu temp etotal fmax lx ly lz vol press
thermo 1000
run 5000
unfix 2

reset_timestep 0
timestep 0.002

compute       mymsdLi Li msd com yes
compute       mymsdO O msd com yes
variable      msdxLi equal "c_mymsdLi[1]"
variable      msdyLi equal "c_mymsdLi[2]"
variable      msdzLi equal "c_mymsdLi[3]"
variable      msdtotLi equal "c_mymsdLi[4]"
variable      msdoxO equal "c_mymsdO[1]"
variable      msdoyO equal "c_mymsdO[2]"
variable      msdozO equal "c_mymsdO[3]"
variable      msdototO equal "c_mymsdO[4]"

fix          msdT2 O ave/time 1 1 2500 v_msdoxO v_msdoyO v_msdozO v_msdototO
file msdOxygen${T1}
fix          msdT1 Li ave/time 1 1 2500 v_msdxLi v_msdyLi v_msdzLi v_msdtotLi file
msdLithium${T1}
fix          3 all nvt temp ${T1} ${T1} 0.01
thermo_style custom step v_Timer cpu temp etotal fmax lx ly lz vol press v_msdtotLi
v_msdototO
thermo 5000
dump dynamics all xyz 5000 2grainsLZO_${T1}.xyz
dump_modify dynamics every 5000 element Li Zr  O first yes
run          1000000
unfix 3
unfix msdT1
```

unfix msdT2

3. Input file of Li₂Zr₆O₁₃@ Li₂Ti₆O₁₃ composite

```
##############################################################
#------------Variables and cell-------------------------#
##############################################################

clear
units        metal      #eV,atomic charge,angstroms,ps,kelvin,bars,g/mol
dimension 3
boundary p p p
atom_style charge
#processors * * * grid numa
read_data LTZO.lmp

group        Li type 1
group        Zr type 2
group        O  type 3
group        Ti type 4


mass 1   6.94000000        # Li
mass 4   47.86700000       # Ti
mass 3   15.99900000       # O
mass 2   91.22400000       # Zr

variable T1 equal 900
variable Timer equal step*dt
log ${T1}.lammps

##############################################################
#---------- Pair styles and electrostatics--------------#
##############################################################

pair_style buck/coul/long 10
pair_coeff * *  0.0 1.0 0.0
pair_coeff 1 4  0.0 1.0 0.0
pair_coeff 2 4  0.0 1.0 0.0
pair_coeff 2 3  1453.8 0.35000  0.0
pair_coeff 4 4  0.0 1.0 0.0
pair_coeff 1 2  0.0 1.0 0.0
pair_coeff 2 4  0.0 1.0 0.0
pair_coeff 1 3  632.1018 0.2906  0.0
pair_coeff 3 4 985.87 0.3760  0.0
pair_coeff 3 3  22764.3 0.149 27.627
```

```
kspace_style pppm 1e-05

##########################################################
# -------------- Run Minimization --------------------#
##########################################################

reset_timestep 0
timestep 0.5
thermo 10
thermo_style custom step enthalpy fmax lx ly lz vol press
min_style cg
minimize 1e-25 1e-25 5000 10000



##########################################################
# -------------- Relax Cell ---------------------------#
##########################################################

reset_timestep 0
timestep 0.5
fix 1 all box/relax aniso 1.0 vmax 0.003
thermo 1
thermo_style custom step enthalpy fmax lx ly lz vol press
min_style cg
minimize 1e-25 1e-25 5000 10000
unfix 1


##########################################################
# ------------- Run NPT T1-----------------------------#
##########################################################

reset_timestep 0
timestep 0.002
velocity all create ${T1} 4928 rot yes dist gaussian
fix     2 all npt temp ${T1} ${T1} 0.01 aniso 1.0 1.0 0.1
thermo_style custom step v_Timer cpu temp etotal fmax lx ly lz vol press
thermo 1000
run 5000
unfix 2

reset_timestep 0
timestep 0.002

compute        mymsdLi Li msd com yes
```

```
compute        mymsdO O msd com yes
variable       msdxLi equal "c_mymsdLi[1]"
variable       msdyLi equal "c_mymsdLi[2]"
variable       msdzLi equal "c_mymsdLi[3]"
variable       msdtotLi equal "c_mymsdLi[4]"
variable       msdoxO equal "c_mymsdO[1]"
variable       msdoyO equal "c_mymsdO[2]"
variable       msdozO equal "c_mymsdO[3]"
variable       msdototO equal "c_mymsdO[4]"

fix            msdT2 O ave/time 1 1 2500 v_msdoxO v_msdoyO v_msdozO v_msdototO
file msdOxygen${T1}
fix            msdT1 Li ave/time 1 1 2500 v_msdxLi v_msdyLi v_msdzLi v_msdtotLi file
msdLithium${T1}
fix            3 all nvt temp ${T1} ${T1} 0.01
thermo_style custom step v_Timer cpu temp etotal fmax lx ly lz vol press v_msdtotLi
v_msdototO
thermo 5000
#dump dynamics all xyz 5000 2grainsLZTO1_${T1}.xyz
#dump_modify dynamics every 5000 element Li Zr  O Ti first yes
run            2500000
unfix 3
unfix msdT1
unfix msdT2
```