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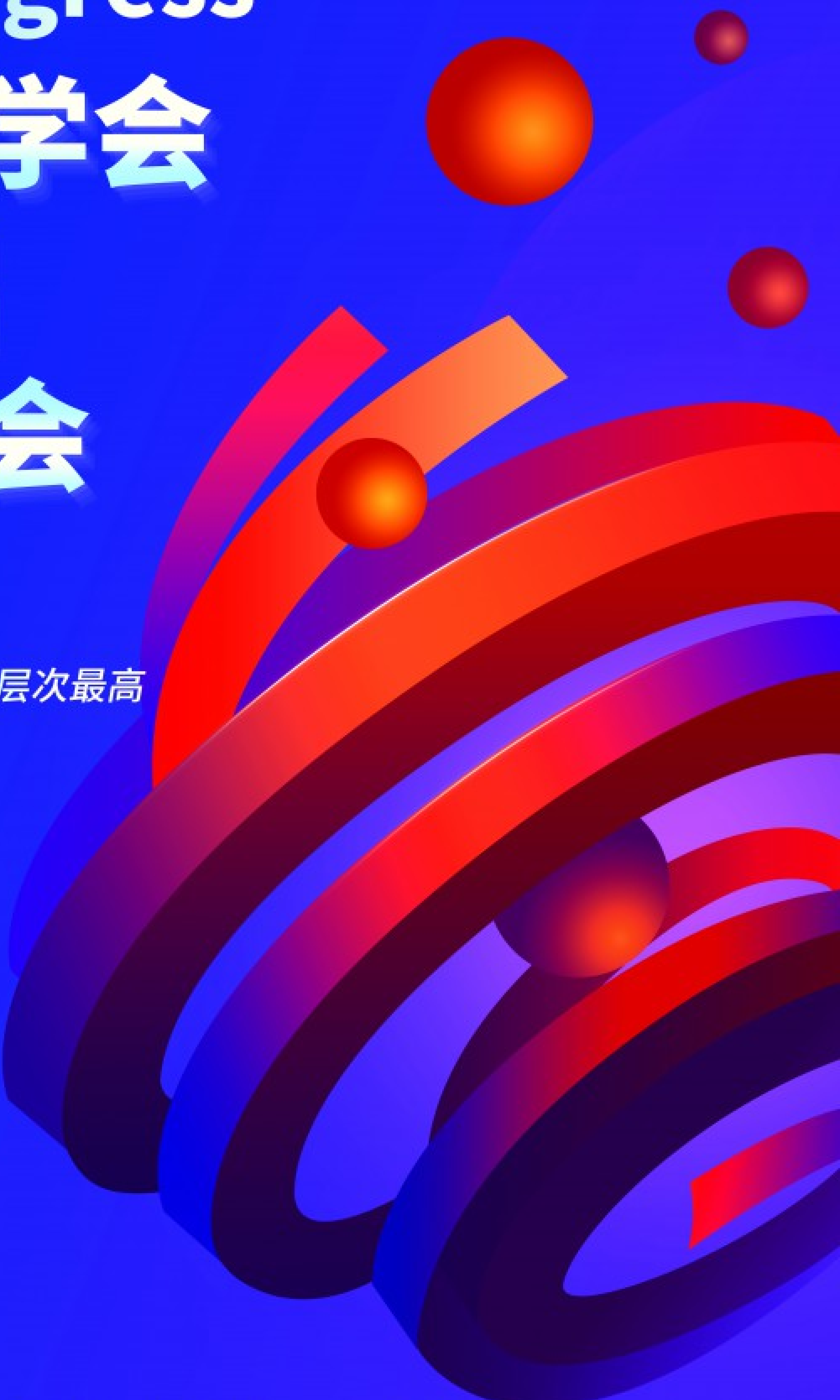
中国化学会
第33届
学术年会



「国内化学及相关领域
规模最大、门类最全、层次最高
的学术盛会」

2023年
6 月17-20日
山东 · 青岛
June 17-20, 2023
Qingdao Shandong

学术分会

01 物理化学前沿
02 表面物理化学
03 超快光谱
04 纳米传感
05 生物分析化学
06 分析仪器与技术
07 生物标记与分析
08 无机合成化学
09 生物无机化学
10 配位超分子化学
11 无机化学前沿
12 元素/金属有机
13 天然产物化学
14 有机合成化学
15 高分子
16 应用化学
17 基础化学教育
18 高等化学教育
19 职业化学教育
20 核化学与放射化学
21 二氧化碳资源化利用
22 理论与计算催化
23 介观催化
24 功能晶态材料
25 流变学
26 电子结构理论及其应用
27 化学中的量子与经典动力学
28 化学热力学与热分析
29 有机/钙钛矿光伏材料与器件
30 有机固体
31 色谱分离分析
32 超分子组装与软物质材料
33 化学信息学与化学计量学
34 环境化学
35 光化学
36 小分子探针与疾病诊疗
37 生物大分子与生命过程
38 基础电化学
39 先进化学电源
40 胶体与界面化学
41 多孔材料
42 绿色化学
43 公共安全化学
44 纳米器件、纳米表征与检测技术
45 纳米生物分析与纳米药物化学
46 纳米材料的合成与组装
47 纳米催化
48 纳米碳材料
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50 纳米体系理论与模拟
51 质谱分析
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53 手性化学
54 物理有机化学
55 燃烧化学
56 木质素
57 农业化学
58 糖化学
59 分子聚集发光
60 仿生材料化学
61 能源化学
62 分子诊断与治疗
63 高压化学
64 有机反应和工艺化学

中国化学会第33届学术年会

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CCS Chemistry

中国化学会第33届学术年会

优秀墙报奖

张颖曦

所在分会：第四十九分会：能源纳米材料物理化学

题目：钼基化合物的 d-p 带隙对于多硫化物转化机制调控的研究

中国化学会
2023年6月

CHINESE CHEMICAL SOCIETY 中国化学会

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Deciphering the d-p band gap on the Polysulfides Conversion with Mo-based Compounds

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Abstract

Transition-metal compounds have been widely proven to be effective catalysts in Li-S chemistry. However, variations in their electrochemical performance make the successor unable to obey the design principle of catalyst engineering. Herein, we have systematically studied thermodynamics and kinetics behaviors of Mo-based compounds and their heterostructure in Li-S chemistry, unraveling the interfacial orbital modulation mechanism with the d-p band gap variation on lithium polysulfide (LiPSs) conversion patterns. Among the compounds, $Mo_2C–MoO_2$/NC as its narrow d-p band gap reflects a new LiPSs conversion pattern which could reduce 1/3 of the activation energy. Therefore, it can promote the capacity retained at 2C almost 5 times the pristine sulfur electrode. The theoretical model uncovered that the d-p gap of catalyst could regulate the Mo-S interfacial bonding and S-S orbit of absorbed LiPSs, varying its electrochemical and thermaldynamic performance. This model provides guidance for band engineering design on Li-S chemistry.

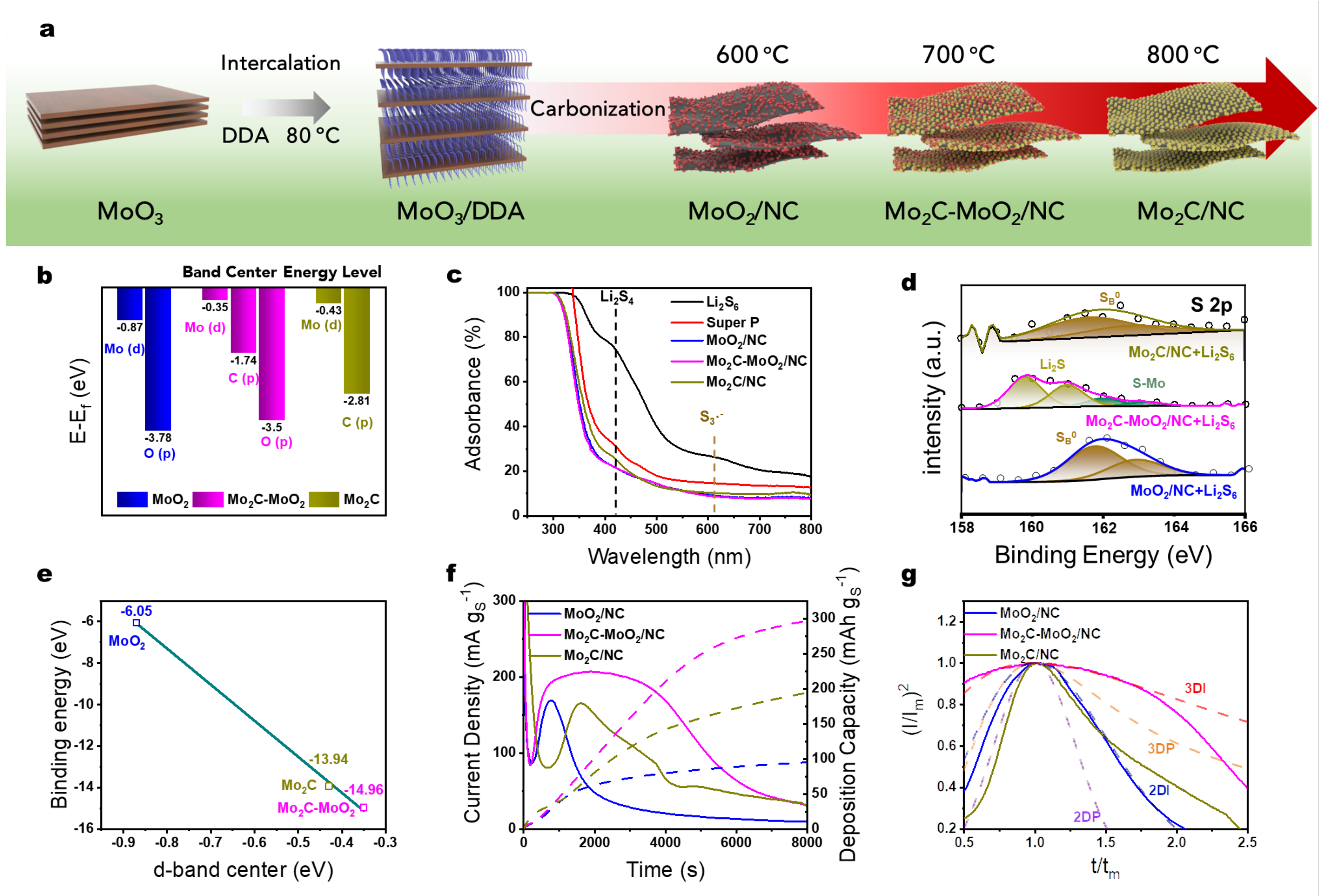

Figure 1. The preparation and thermodynamic performance of Mo-based compounds as LiPSs conversion catalysts.

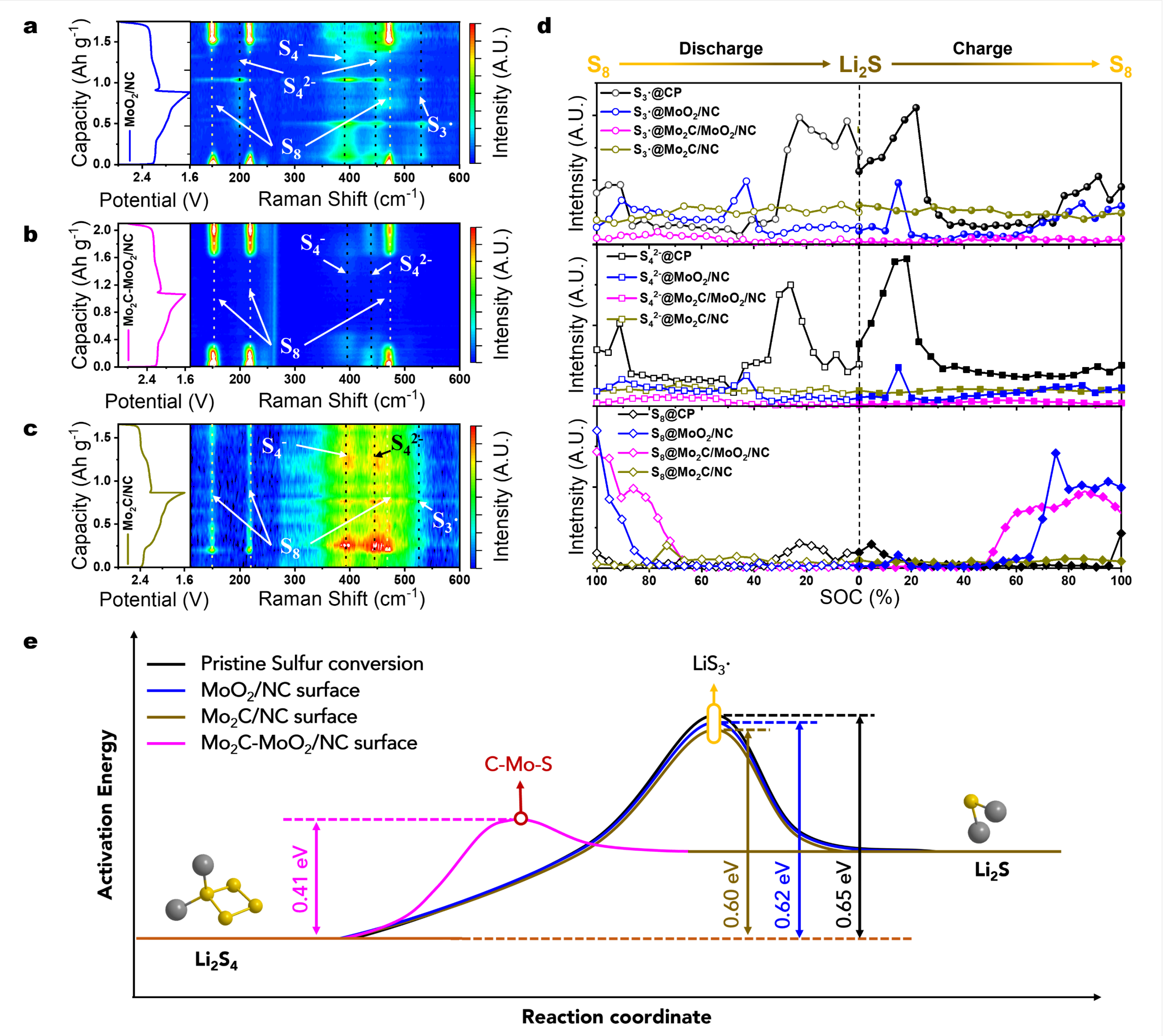

Figure 3. The LiPSs conversion patterns variation by Mo-based compounds.

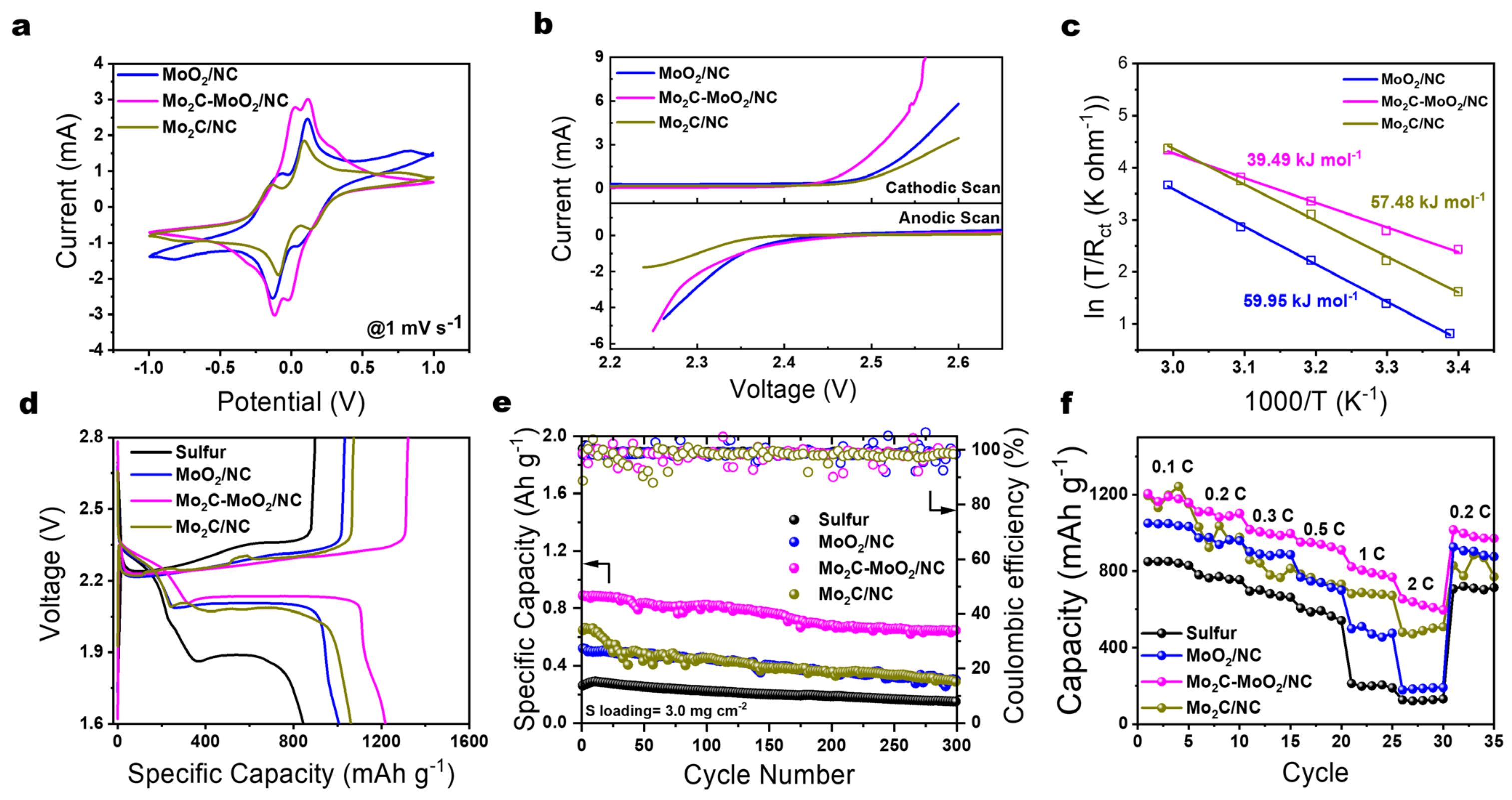

Figure 2. The LiPSs conversion kinetics improvement and electrochemical performance of Mo-base compounds as Li-S cell additive.

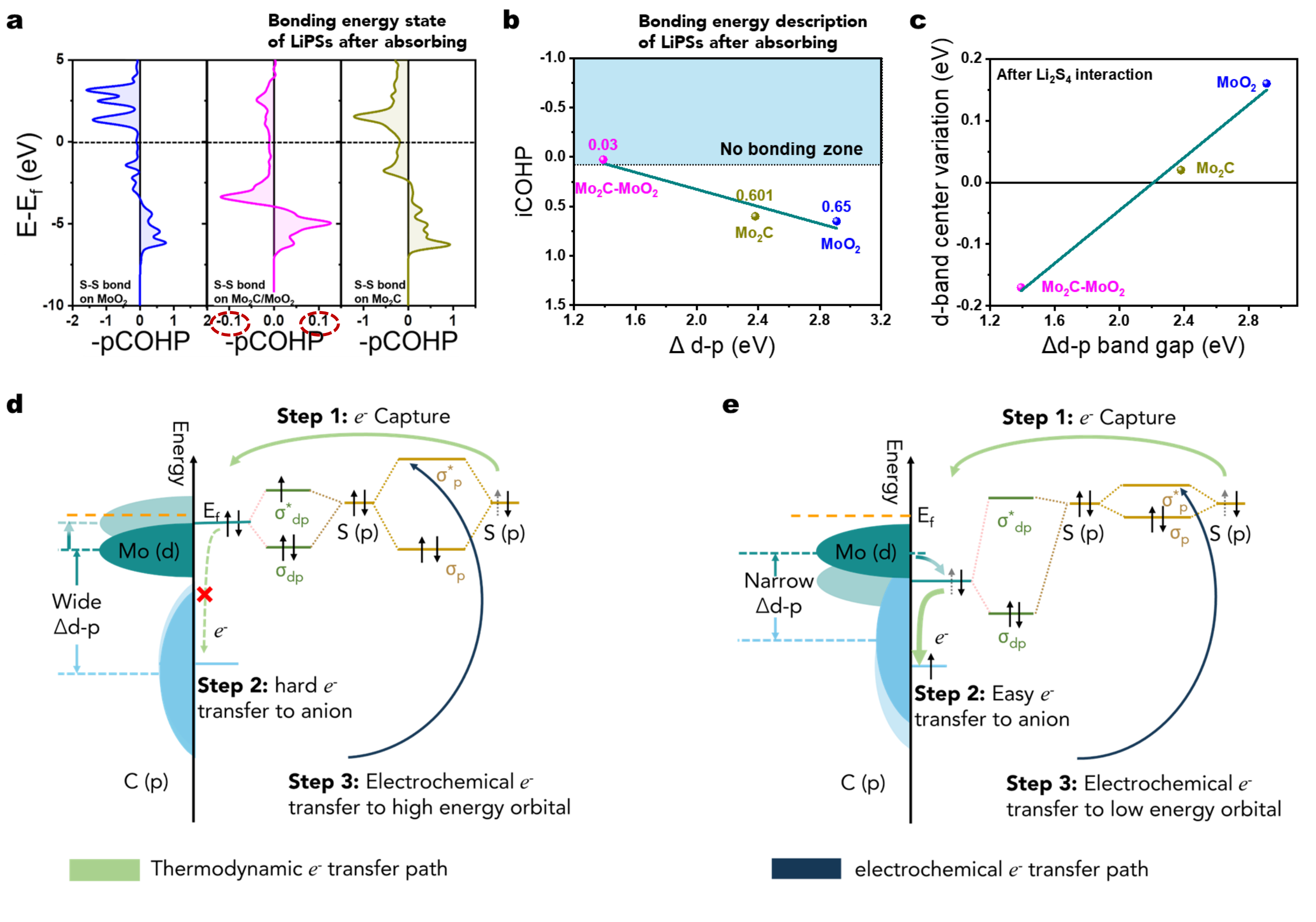

Figure 4. DFT calculation unravel the Effects of d-p band gap on LiPSs conversion

Conclusion

I. The anion of the narrow d-p band gap catalyst can act as the electron container for the extra-captured electron from the Mo cations as it interacts with LiPSs, softening the d-band variation of Mo and the active sites impedance.

II. While the gap is lower than a certain value as Mo_2C-MoO_2/NC does, the d-band state of Mo would not be positive shifting after interacting with LiPSs. Therefore, it can decrease the antibonding orbital energy level and directing the formation of C-Mo-S bond, avoiding the active site block of the cation.

Yingxi Zhang, et al. Deciphering the d-p band gap on the Polysulfides Conversion with Mo-based Compounds, Joule (Second round revising)