Supporting Information

Mechanism of lithium dendrites growth on iron surfaces toward high-performance and safe anode-free lithium metal batteries

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Fig. S1. Transformation of randomly placed Li atoms into Li (110) crystal surface.

Fig. S2. (a) Atomic snapshot of homogeneous deposited Li atoms on Fe(110) surface; red color ball represents Fe atom and blue color ball represents Li atom. (b) (c) Top view of the first layer and second layer of Li atoms on the Fe surface after homogeneous deposition. (d) (e) 2D potential energy distribution of Li atoms on the first and the second layers.

Fig. S3. (a) Atomic snapshot of homogeneous deposited Li atoms on Fe(111) surface; red color ball represents Fe atom and blue color ball represents Li atom. (b) (c) Top view of the first layer and second layer of Li atoms on the Fe surface after homogeneous deposition. (d) (e) 2D potential energy distribution of Li atoms on the first and the second layers.

Fig. S4. Atomic snapshot of Li dendrite on (a) Fe(110) and (b) Fe(111) surface of inhomogeneous deposited Li atoms.

Fig. S5. Self-healing process of block Li with surface defects at 300K. Blue, green, red and gray represent BCC, FCC, HCP and other states of Li atoms, respectively.

Fig. S6. (a–c) Atomic diagram of the Li dendrite growth process on a cracked Fe(111) surface. (d) Difference of diffusion energy barriers of Li atoms on cracked and smooth surfaces of Fe(100) and Fe(111).

Fig. S7. Atomic diagram of the Li dendrite growth process on Fe(100) surface with point defect. Yellow color ball represents the first layer atoms of Fe substrate.

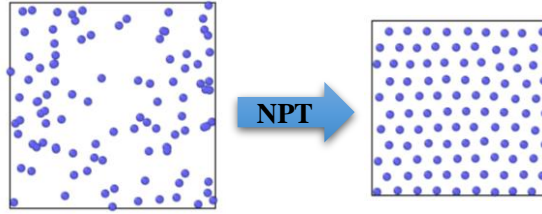**Fig. S8**. Atomic snapshots of Li atoms inhomogeneously deposited on Fe(100) surfaces with crack.

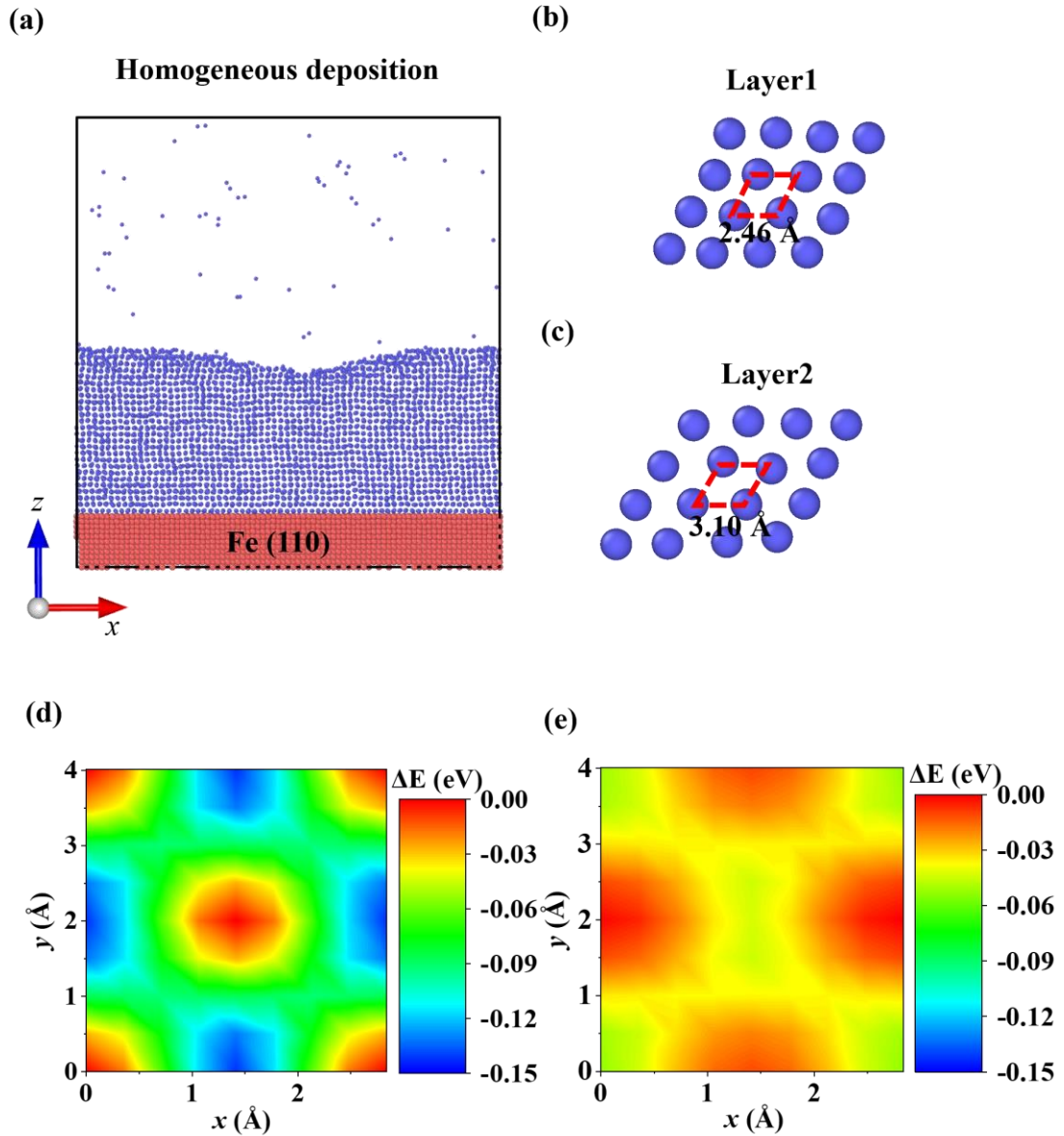**Fig. S1**. Transformation of randomly placed Li atoms into Li (110) crystal surface.

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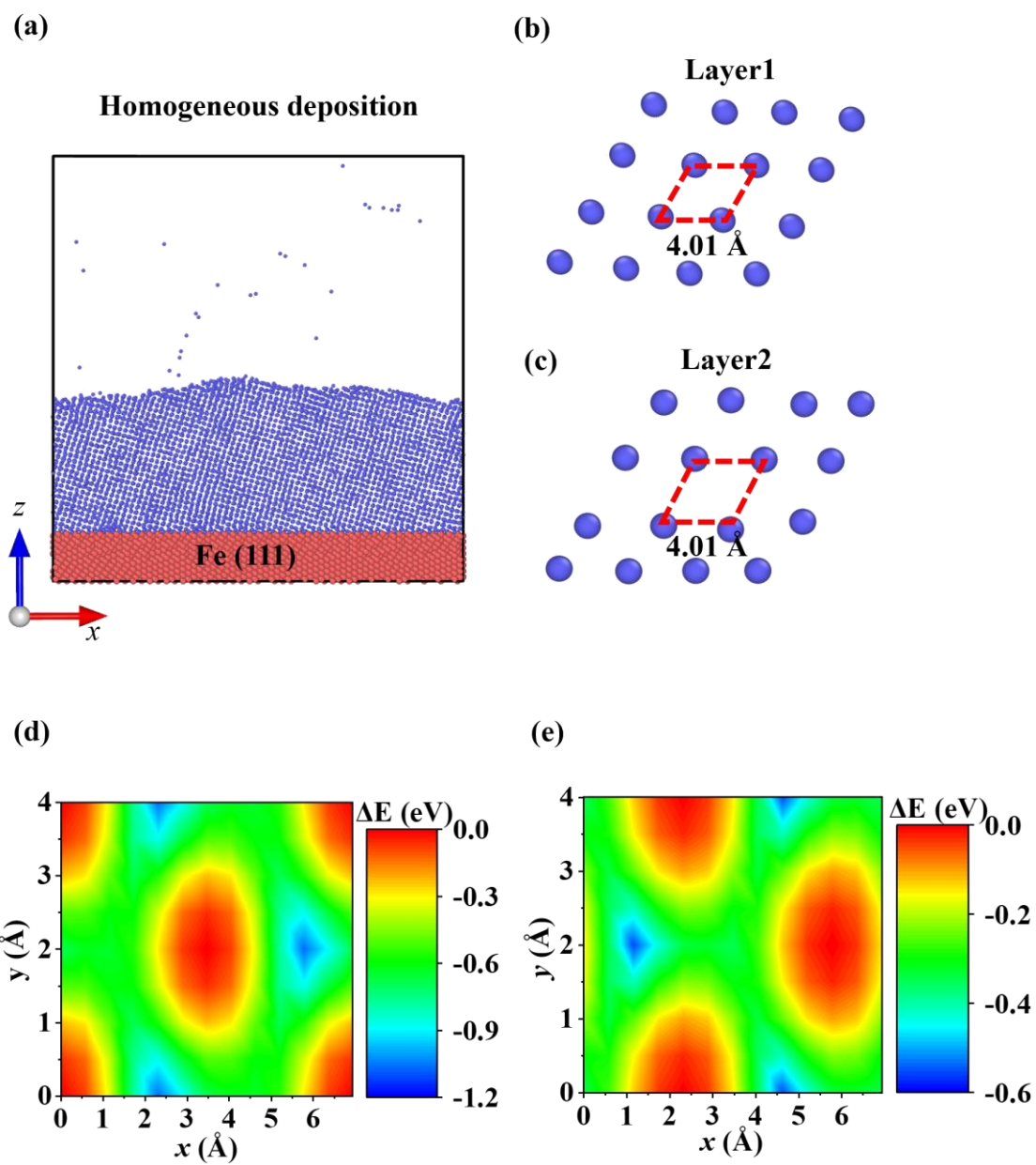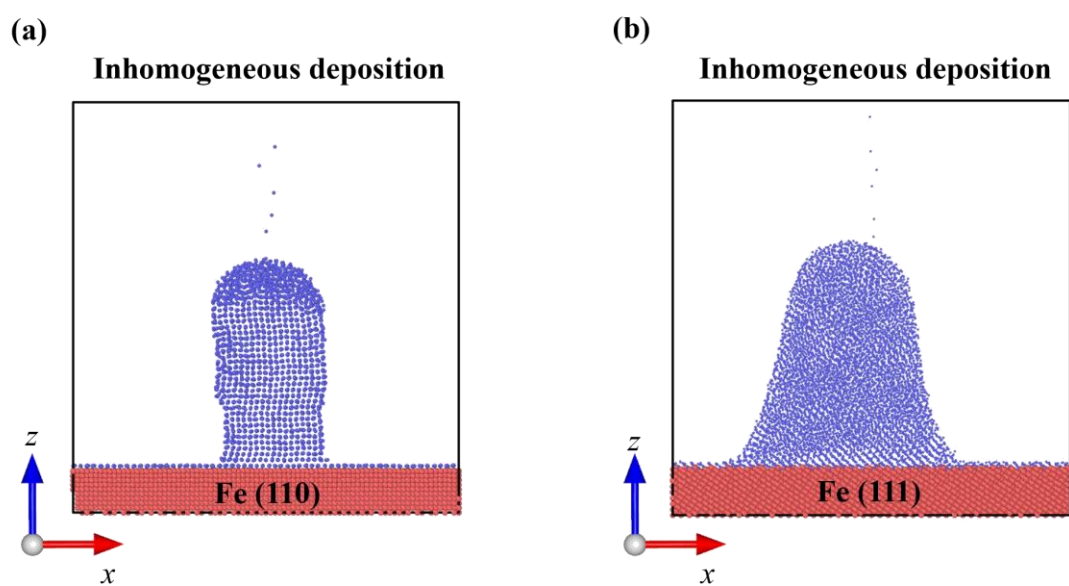
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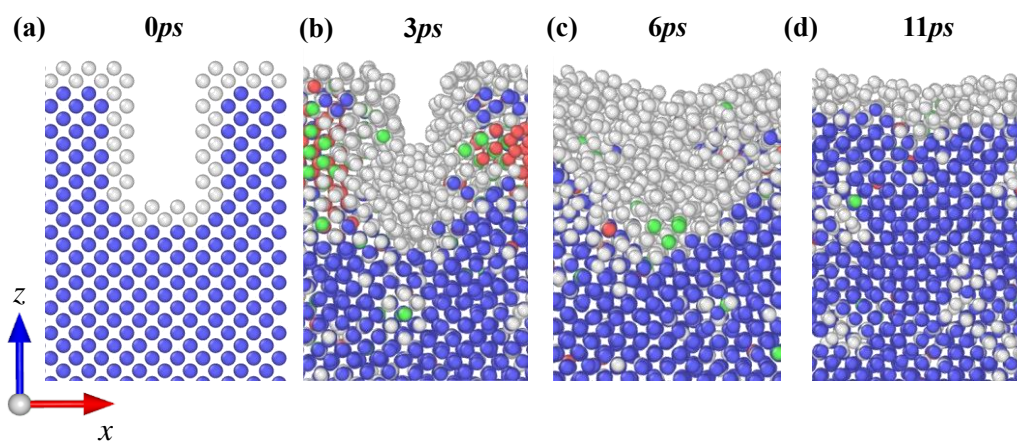

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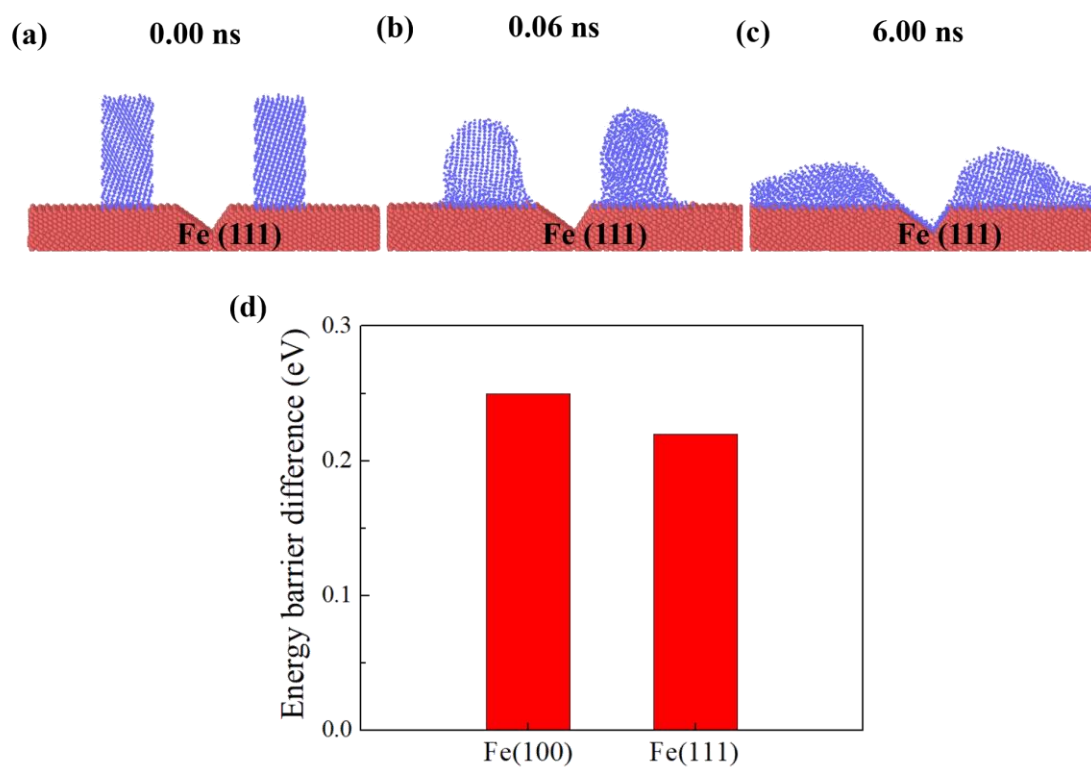
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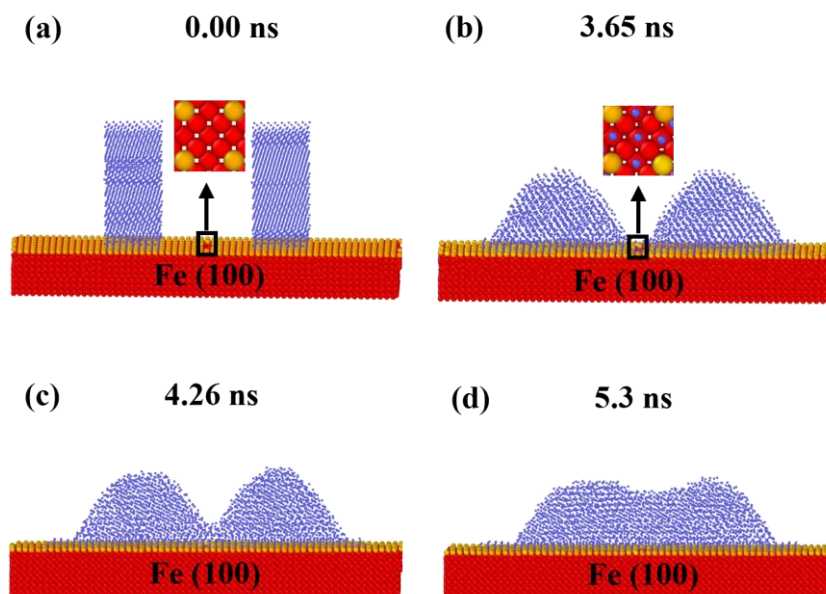

Fig. S7. Atomic diagram of the Li dendrite growth process on Fe(100) surface with point defect.

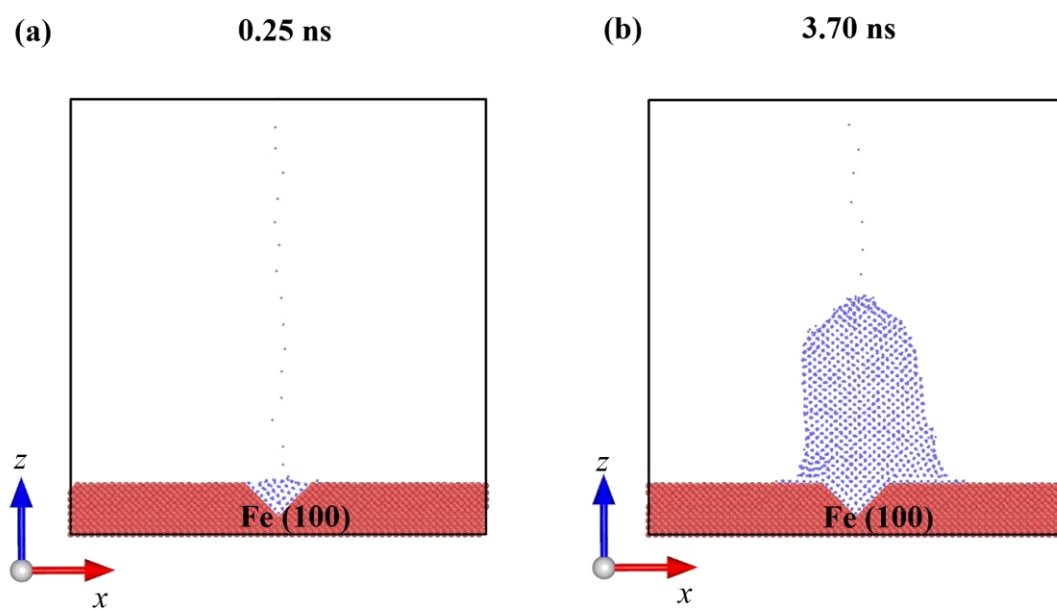Yellow color ball represents the first layer atoms of Fe substrate.

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