

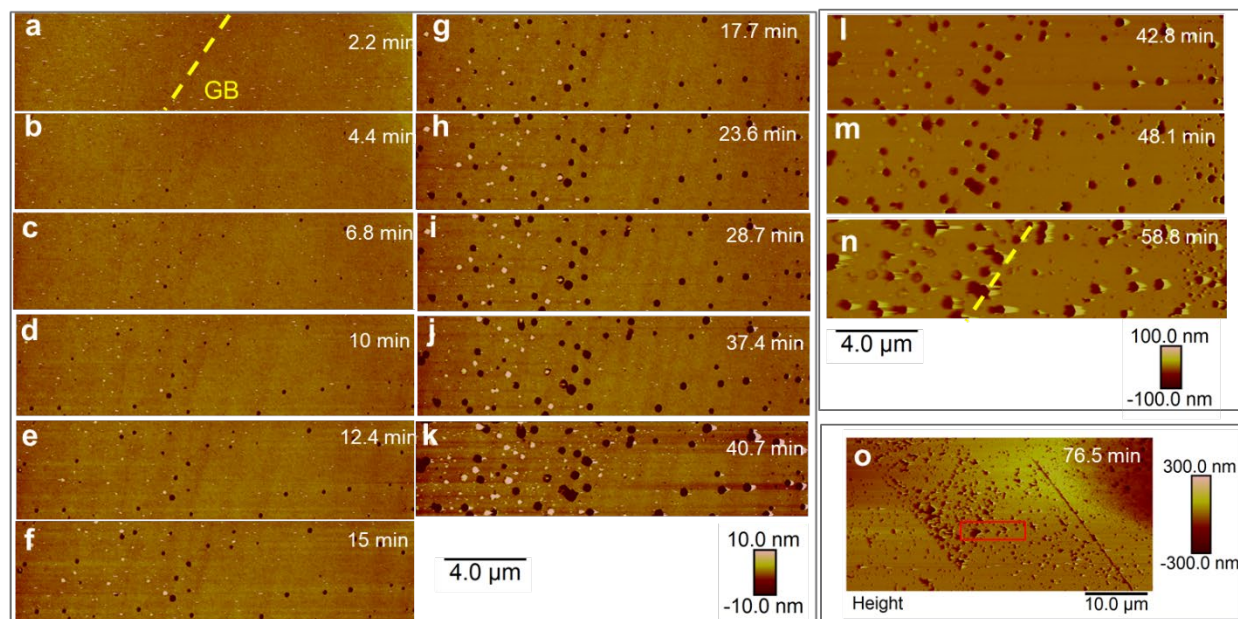
Directly Monitoring the Shift in Corrosion Mechanisms of a Model FeCrNi Alloy Driven by Electrical Potential

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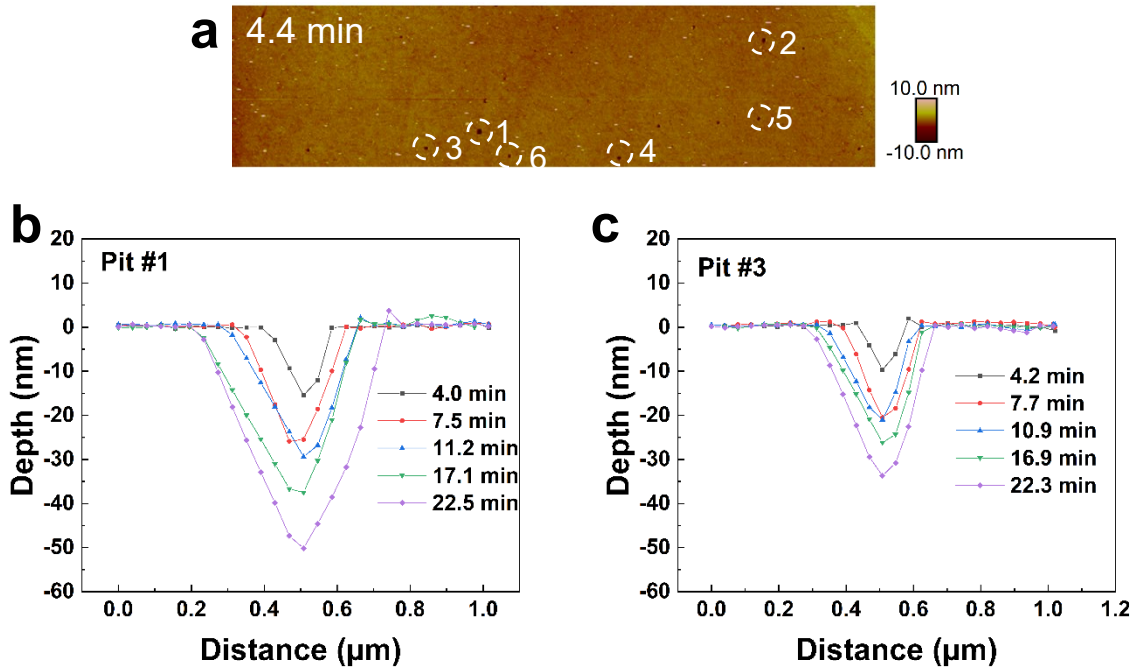
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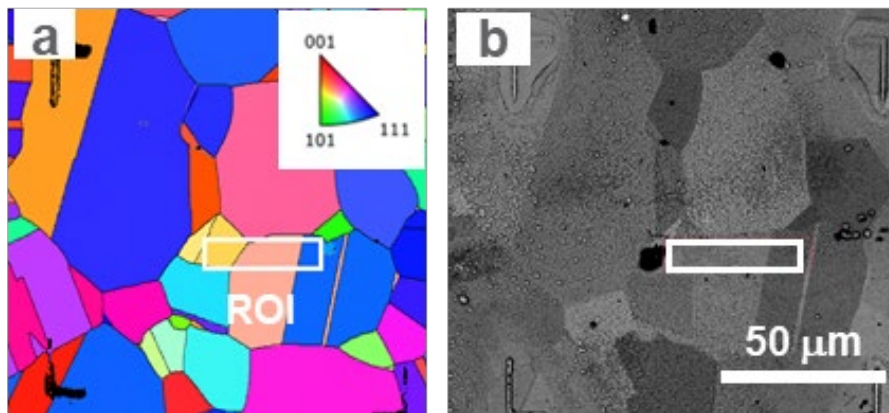
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Supplementary Figure 1. In situ AFM images of the nonbiased sample. (a–o) AFM images from 2.2 min to 76.5 min. The red rectangle in (o) represents the region for in situ monitoring (a–n). The scanning area of in situ measurement is $5 \times 20 \mu\text{m}^2$. The vertical scale of images (a–k) is from -10 nm to 10 nm . The vertical scale of images (i–n) is from -100 nm to 100 nm . The vertical scale of image (o) is from -300 nm to 300 nm .



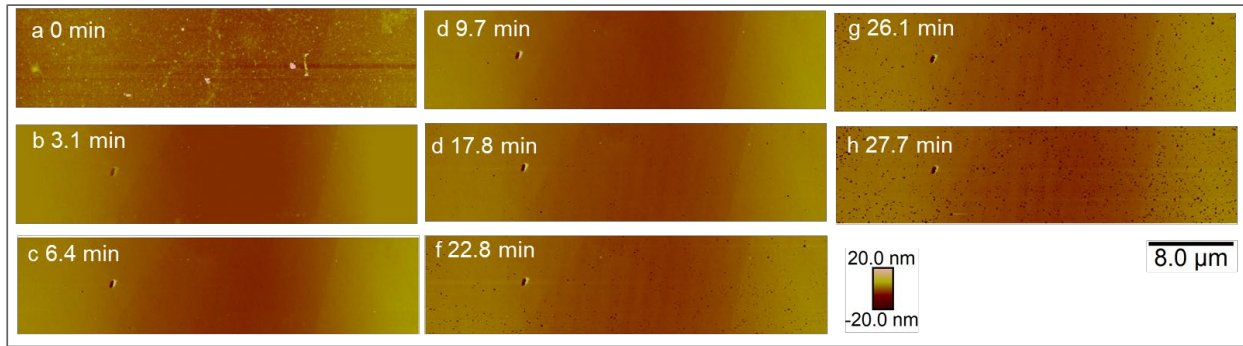
Supplementary Figure 2. Pit profile of selected pits. (a) Selected corrosion pits of the nonbiased sample illustrated on the 4.4 min AFM image for quantitative analysis, and the representative time-dependent pit profiles of (b) Pit #1 and (c) Pit #3 along the horizontal plane.



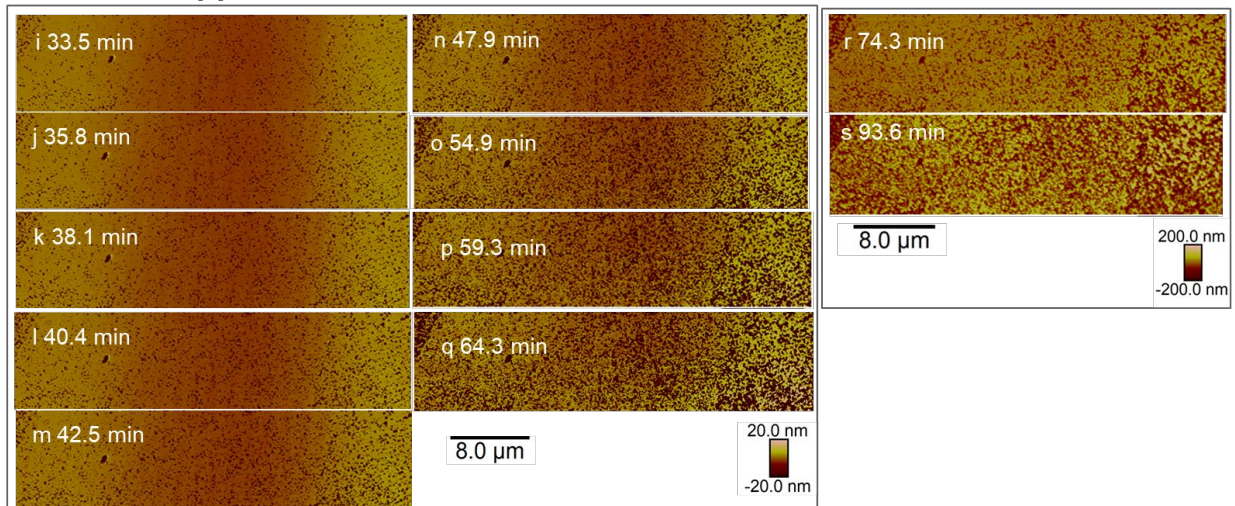
Supplementary Figure 3. EBSD IPF images showing the ROI of the in situ EC-AFM scan area of the biased sample: (a) SEM BSE image of the same location after electrochemical corrosion. The

IPF color code is projected to the surface normal. EBSD results show Grain 1, Grain 2, and Grain 3 are close to the [012], [124], and [323] orientations.

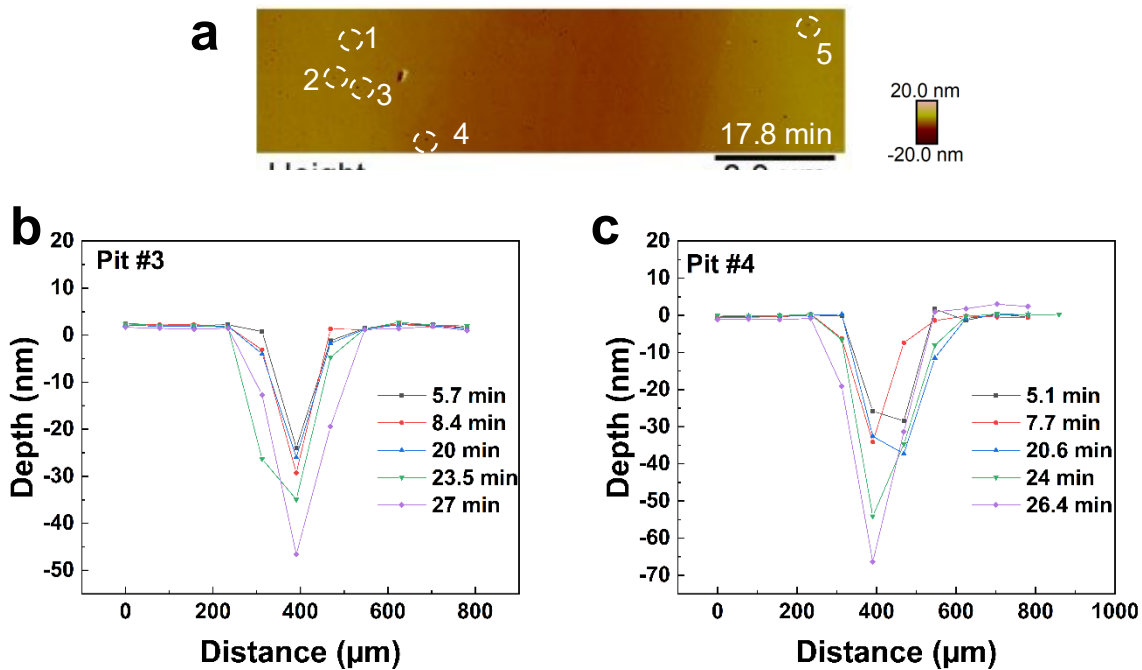
Before bias applied



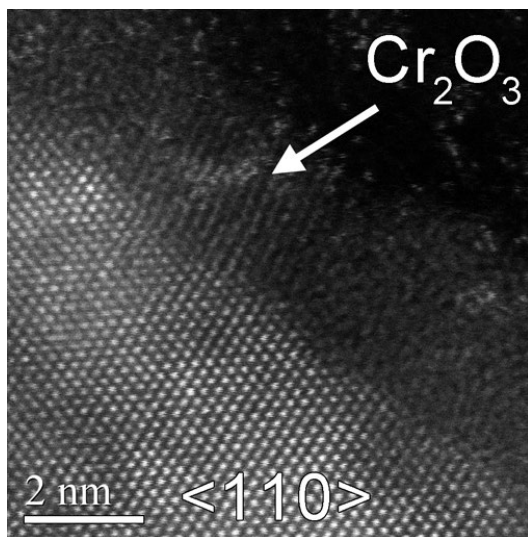
-0.5V bias applied



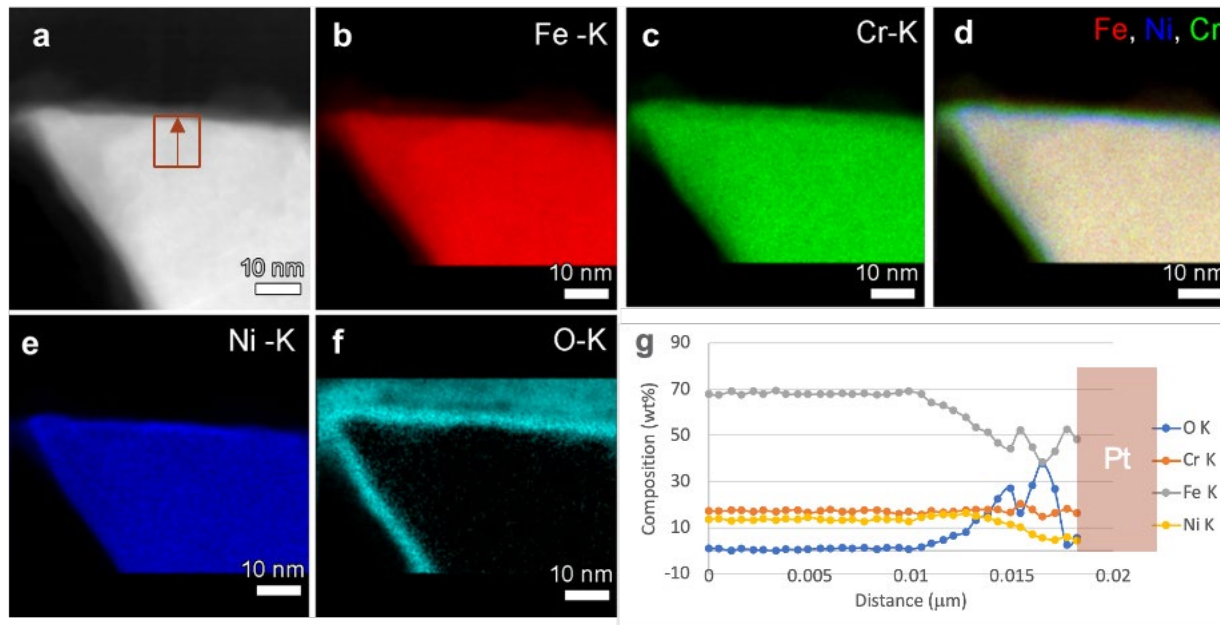
Supplementary Figure 4. In situ EC-AFM images of the biased sample corroded in 0.5 M DCl: (a–h) before bias is applied and (i–s) after a -0.5 V bias is applied. The scan area of the in situ measurement is $10 \times 40 \text{ μm}^2$. The vertical scale of images (a–q) is from -20 nm to 20 nm . The vertical scale of images (r and s) is from -200 nm to 200 nm .



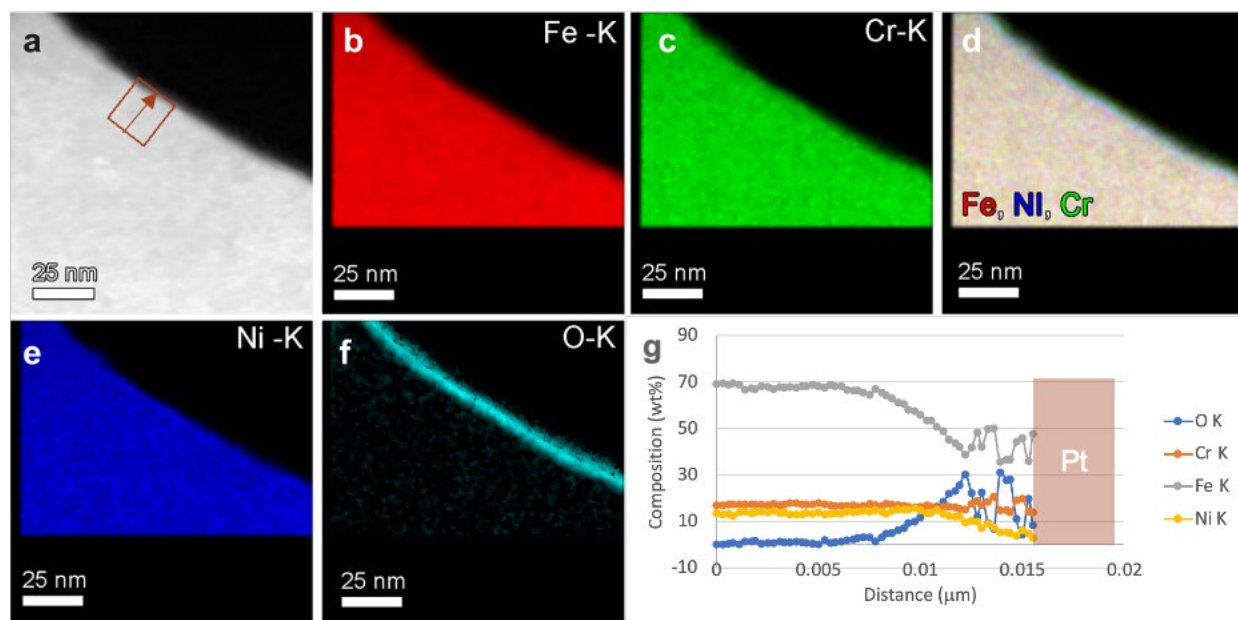
Supplementary Figure 5. Selected pit profiles of the biased sample. (a) Selected corrosion pits of the biased sample illustrated on the 17.8 min AFM image for quantitative analysis, and the representative time-dependent pit profiles of (b) Pit #3 and (c) Pit #4 along the horizontal plane.



Supplementary Figure 6. TEM image of the biased sample showing Cr_2O_3 .



Supplementary Figure 7. TEM EDS mapping of the nonbiased sample: (a) TEM image of the mapping region, (b) Fe mapping, (c) Cr mapping, (d) mixed mapping of Fe, Ni, and Cr, (e) Ni mapping, (f) O mapping, (g) composition (wt%) line scan along the red arrow.



Supplementary Figure 8. TEM EDS mapping of the biased sample: (a) TEM image of the mapping region, (b) Fe mapping, (c) Cr mapping, (d) mixed mapping of Fe, Ni, and Cr, (e) Ni mapping, (f) O mapping, (g) composition (wt%) line scan along the red arrow.