Electronic supplementary information (ESI):

Sign inversion of surface stress-charge response of bulk nanoporous nickel actuator with different surface states

Qingguo Bai,^{†,‡} Conghui Si,[†] Jie Zhang, Zhonghua Zhang*

Key Laboratory for Liquid-Solid Structural Evolution and Processing of Materials (Ministry of

Education), School of Materials Science and Engineering, Shandong University, Jingshi Road

17923, Jinan 250061, P.R. China

[†] The authors equally contribute to this work.

‡ Present address: Institut de Chimie de la Matière Condensée de Bordeaux, 87 Avenue du

Docteur Albert Schweitzer Pessac F-33608 Cedex, France

*Corresponding author. Email: <u>zh_zhang@sdu.edu.cn</u>, Tel/Fax: +86-531-88396978.

Tables

Table S1. Summary of potential range, transferred charge, reversible strain and surface stress-charge coefficient ζ in nanoporous nickel sample with clean surface, oxidation process and oxide-covered surface.

| surface state | potential range vs. SCE (V) | scan rate (mV/s) | electrolyte | Transferred charge (C) | reversible strain (10 ⁻⁴) | ζ(V) |
|----------------------|--------------------------------|------------------------|--------------|------------------------|--|---------------|
| clean surface | -1.1 ~ -0.7 | 1 | 5 M NaOH | 6 | 8 | -1.0±0.004 |
| oxidation process | -1.1 ~ 0.6 | 10 | 5 M NaOH | 65 | / | / |
| | -1.1~ -0.7 | 1 | 5 M NaOH | 0.55 | 1.25 | +1.4±0.02 |
| oxide covered | -0.7~0.6 | 1 | 5 M NaOH | 23 | 65 | +5.0±0.01/ |
| surface | 0.7 - 0.0 | I | 5 101 100011 | 23 | 05 | $+1.8\pm0.01$ |
| | -0.5 ~0.5 | 1 | 0.7 M NaF | 1.5 | 6.5 | $+3.4\pm0.01$ |



Figure S1. Schematic illustration of the measuring apparatus for electrochemical actuation (CE: counter electrode, RE: reference electrode, WE: working electrode).



Figure S2. XRD patterns of (a) the freshly as-dealloyed np-Ni sample and (b) the np-Ni sample after being cycled under positive potentials ($-1.1 \sim 0.6$ V vs. SCE).



Figure S3. Successive CV curves of the freshly prepared np-Ni sample in the 5 M NaOH solution at the scan rate of 10 mV/s. The first cycle is denoted in red color.



Figure S4. Typical reversible strain of the np-Ni sample with a clean surface versus time, together with the potential variation (from -1.1 to -0.7 V vs. SCE) during four successive CV cycles at the scan rate of 1 mV/s.



Figure S5. Typical reversible strain of the np-Ni sample with oxide-covered surface versus time, together with the potential variation (from -0.7 to 0.6 V vs. SCE) during five successive CV cycles at the scan rate of 1 mV/s.



Figure S6. (a) Successive cyclic voltammograms (eight cycles) of the np-Ni with oxide-covered surface with a potential window of $-0.7\sim0.6$ V vs. SCE in the 5 M NaOH solution at a scan rate of 10 mV/s. (b) Strain response versus time corresponding to the CV.



Figure S7. Cyclic voltammograms of the oxide-covered np-Ni sample in the 0.7 M NaF solution at different scan rates (potential range $-1.0 \sim 1.0$ V vs. SCE).



Figure S8. Typical reversible strain of the np-Ni sample with oxide-covered surface versus time, at the four different scan rates: (a) 50 mV/s, (b) 10 mV/s, (c) 5 mV/s, (d) 1 mV/s, together with the potential variation during successive CV cycles.



Figure S9. Strain response versus the transferred charge at different scan rates. The slopes of the thick semitransparent straight lines provide the values of ζ , as indicated by the grey triangles and the potential at which the charge equals to zero is chosen arbitrarily. The data were obtained from Figures S7 and S8.