

Lattice templating and galvanic coupling effects on the electrochemical performance of core/shell battery materials

Elahe Moazzen*¹, Elena V. Timofeeva¹, Carlo U. Segre¹
¹Illinois Institute of Technology, Chicago, IL

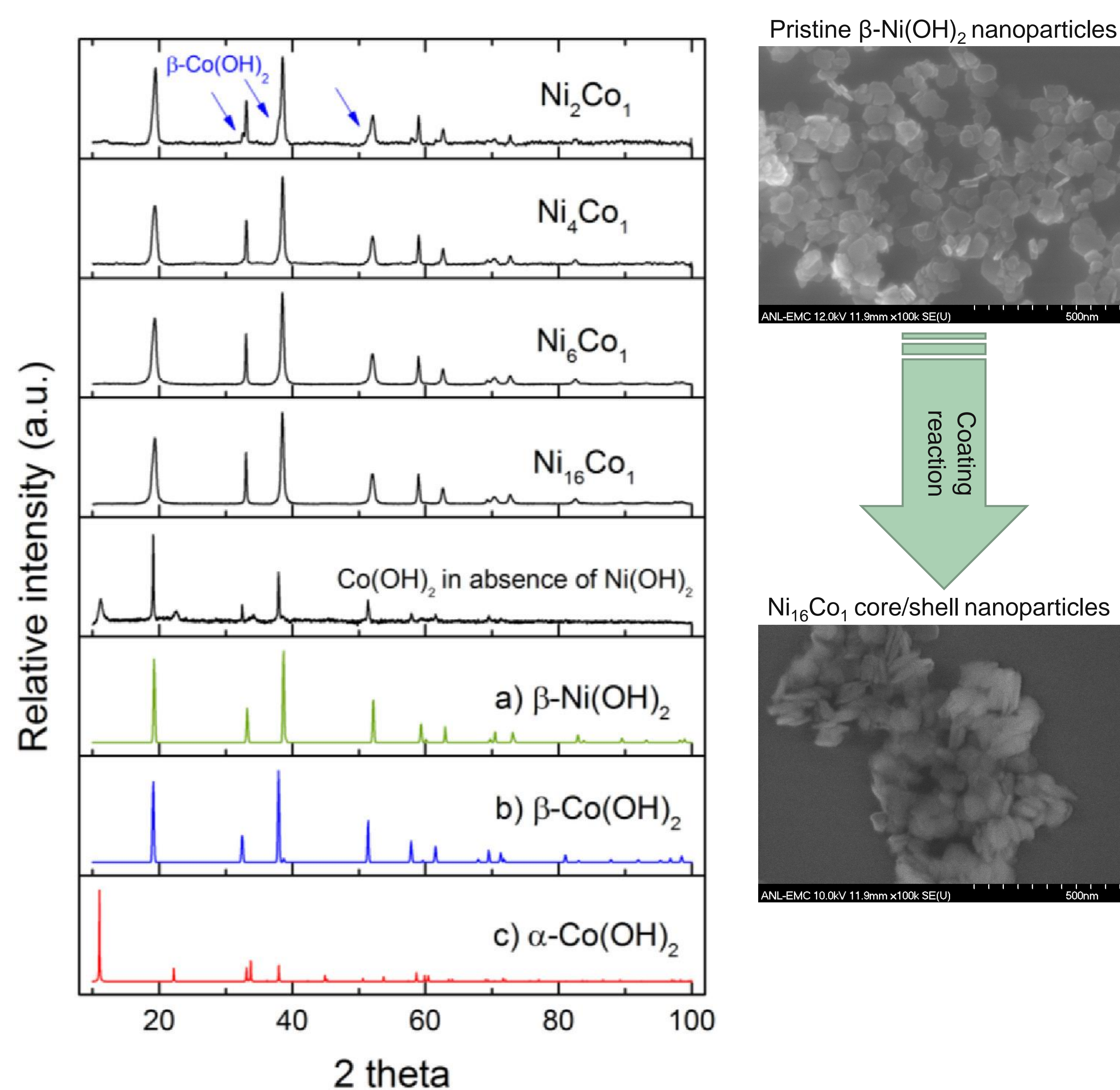
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Introduction

β -Ni(OH)₂ cathode material possesses high theoretical capacity, whereas, it does not perform efficiently at high rates of charging due to its low electrical conductivity resulting in incomplete discharge of the active material and slow charge/discharge rate. This study is focused on improving the discharge of the material and consequently capacity and performance in fast charging by a material activation mechanism. Ni(OH)₂ nanoparticles were synthesized and coated with epitaxial Co(OH)₂ shells and a series of Ni(OH)₂/Co(OH)₂ core/shell nanoplatelets with varying shell thickness were systematically investigated with a combination of scanning electron microscopy (SEM) with energy dispersive x-ray analysis (EDX), x-ray diffraction (XRD), in situ and ex situ x-ray absorption fine structure spectroscopy (XAFS), and electrochemical tests.

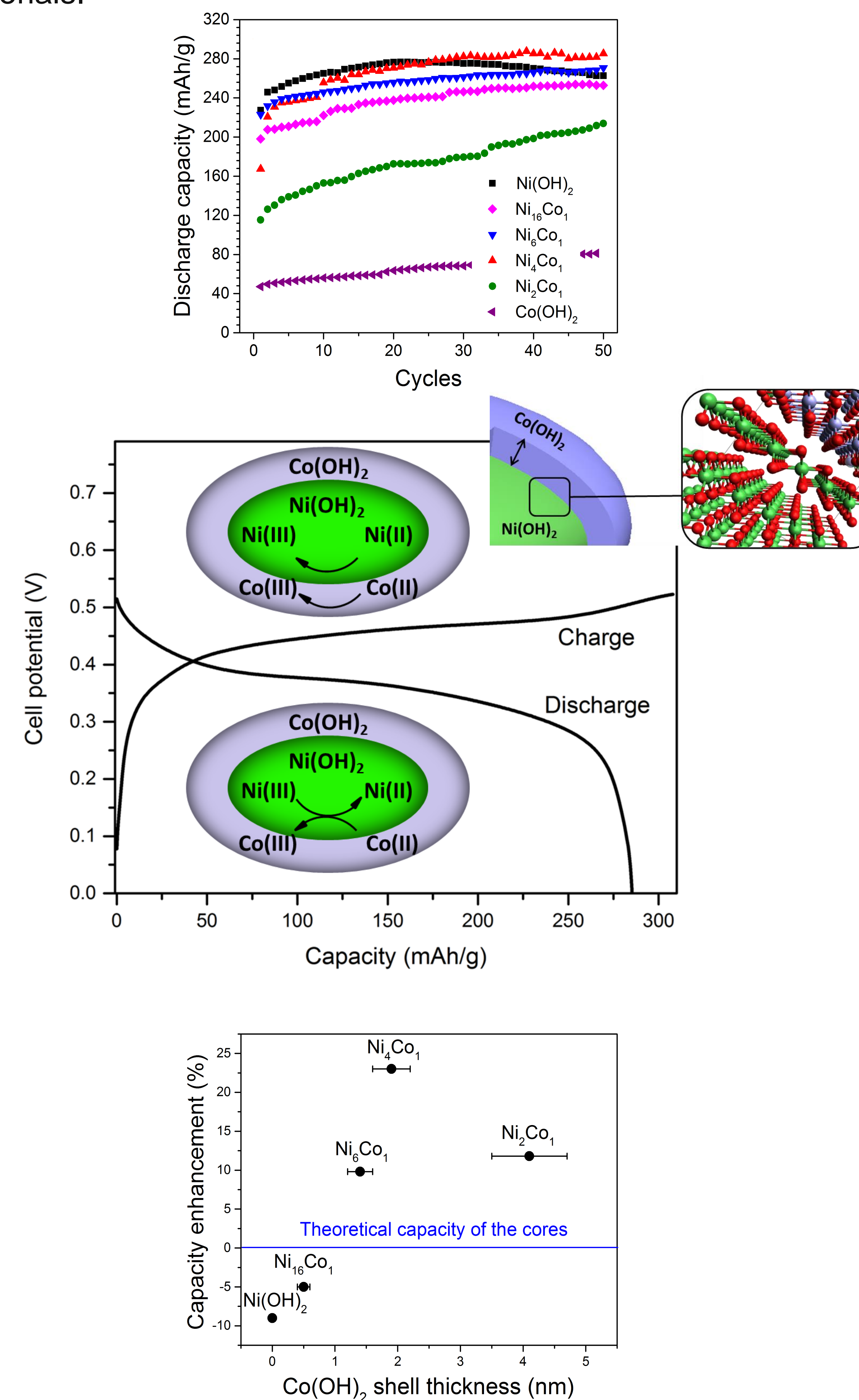
XRD and SEM characterization

Although the pristine Co(OH)₂ (precipitated without Ni(OH)₂ cores) is composed of 44 wt% α -Co(OH)₂ per Rietveld refinement, XRD patterns do not show α -Co(OH)₂ in the crystalline composition of the core/shell nanoparticles, suggesting a templating effect on the shells growth.



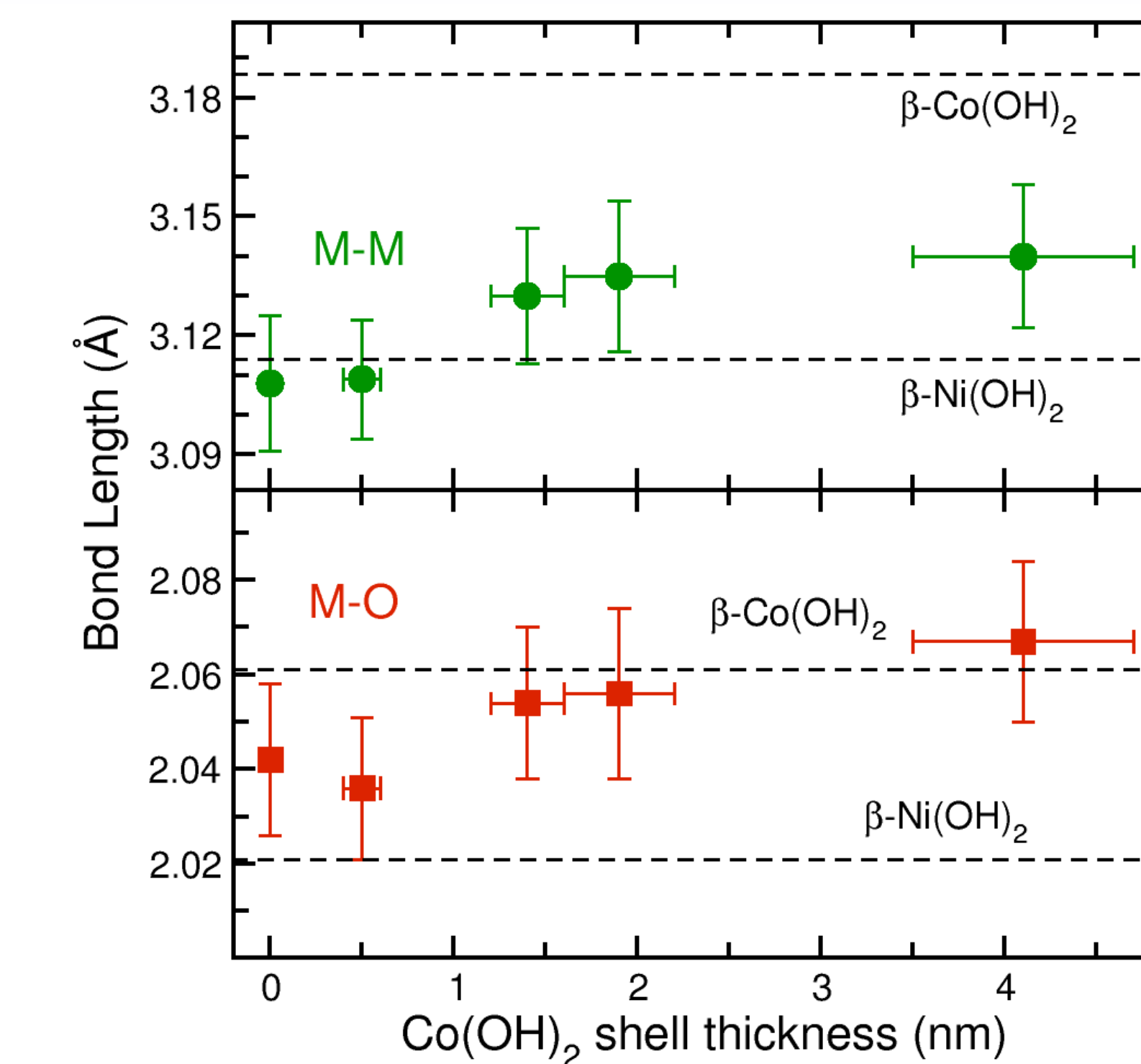
Solid state electrochemical Tests

Structure-property correlations revealed that electrochemical behavior and reversibility of Co(OH)₂ redox conversion is improved non-linearly with the shell thickness due to the lattice templating effect, and the galvanic coupling of core and shell materials.



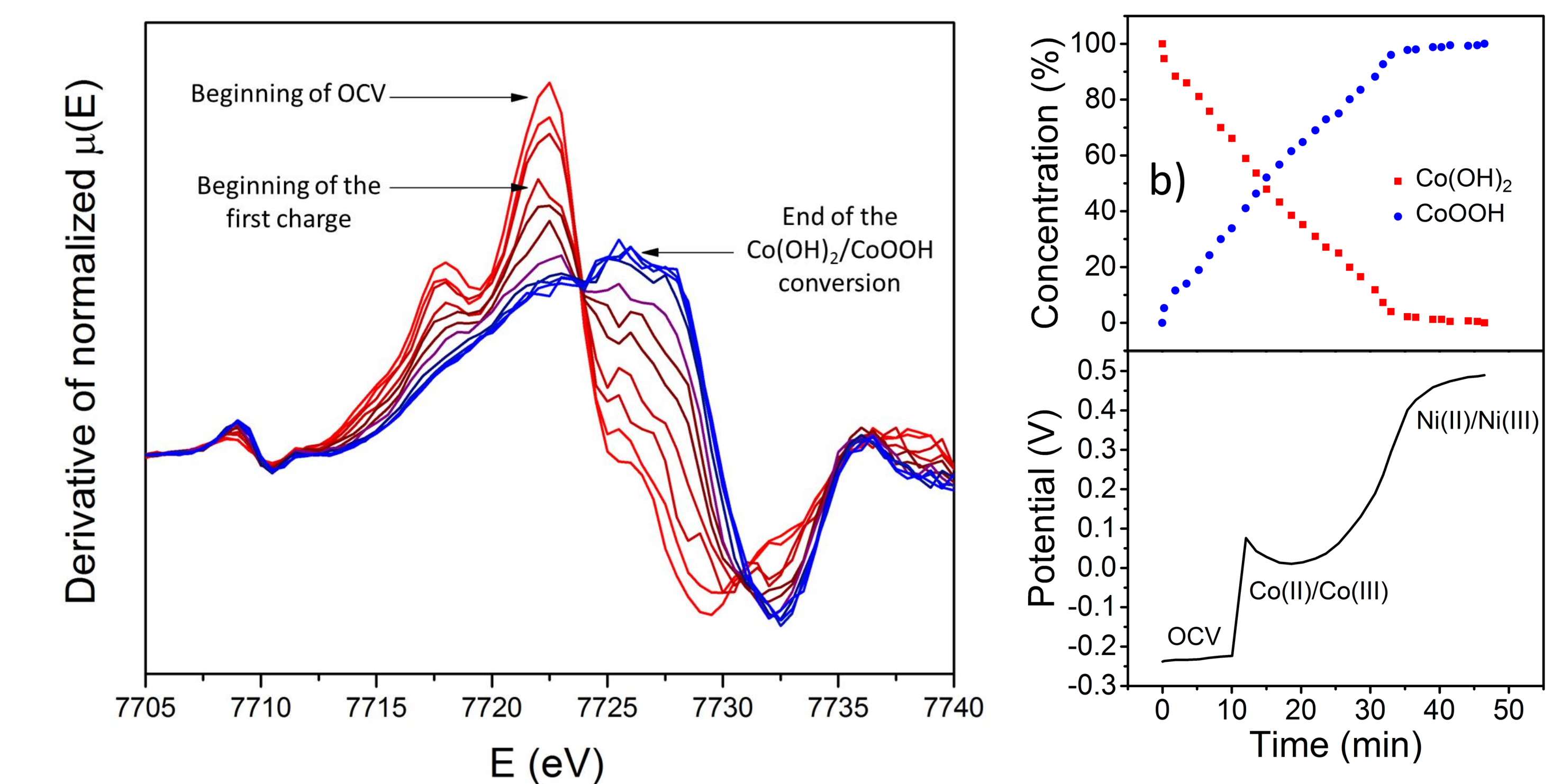
Ex situ XAS

The lattice templating effect was confirmed by XAFS results showing that Co-M and Co-O distances are close to those of the Ni(OH)₂ lattice in thin shells and shift gradually towards values of bulk Co(OH)₂ as the shell thickness increases



In situ XAS

In situ synchrotron XAS experiment was performed on a solid casted pouch cell during the first two charge and discharge cycles to investigate changes in local Co environment.



Conclusion

A systematic investigation of the effect of coating thickness on the electrochemical performance of core/shell NPs revealed a dependence of the reversibility of the Co(II)/Co(III) redox transformation on the shell thickness. Co(OH)₂ shells facilitate discharging of NiOOH through galvanic coupling between the core and the shell, and at the optimal shell thickness (1.9±0.3 nm) complete engagement of the cores and the shells is achieved with nearly maximum theoretical capacity of the composite cathode with 99.6% utilization of both core and shell material and a 10% improvement over the capacity of unmodified Ni(OH)₂ material. It is suggested that such thickness dependent Co(OH)₂ shell behavior is due to the lattice templating effect and strained growth of the Co(OH)₂ phase on Ni(OH)₂ platelets.

