

Experimental report

14/09/2026

Proposal: 5-25-289

Council: 4/2023

Title: Nickel Exsolution induced by the Phase Transformation from $n = 2$ to $n = 1$ Ruddlesden-Popper Manganite to catalyze Dry Reforming of Methane

Research area: Materials

This proposal is a new proposal

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Experimental team:

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Samples: $\text{La}_{1.5}\text{Sr}_{1.5}\text{Mn}_{1.5}\text{Co}_{0.25}\text{Ni}_{0.25}\text{O}_7$

Instrument	Requested days	Allocated days	From	To
D20	4	2	31/10/2023	02/11/2023

Abstract:

This study aims to investigate bimetallic exsolution involving a Ruddlesden-Popper manganite undergoing structure reconstruction. The exsolved system is used to activate the dry reforming of methane reaction.

In situ neutron diffraction data will be collected on D20 as a function of temperature

i) under H_2 at 750°C to monitor the exsolution process, phase reconstruction and phase stability

ii) under CH_4 / H_2O / He gas mixtures to monitor the evolution of the exsolution system in dry reforming conditions at different temperature, between 700°C and 900°C . Long data sets will be collected to track structural and composition changes of the bimetallic particles (oxidation?) and support (formation of intermediate phases, coking) to select the best conditions for the DRM reaction and get insight into key factors to guide the design of improved materials.

EXPERIMENTAL REPORT

***In situ* neutron diffraction of a Sr-rich Ruddlesden–Popper anode (LN3) during activation and dry reforming of methane**

Aim

To follow *in situ*, on $\text{La}_{0.5}\text{Sr}_{1.5}\text{Mn}_{0.7}\text{Ni}_{0.3}\text{O}_4$ (LN3), the competition that decides the working state of Sr-rich Ruddlesden–Popper anodes under CO_2 -containing feeds: Ni exsolution, which builds the metallic phase, against SrO carbonation, which attacks the host. The sample was reduced *in situ* under dilute H_2 at 700 °C and then held under $\text{CH}_4:\text{CO}_2:\text{He}$ through a ladder of isotherms up to 900 °C, with patterns collected continuously at two wavelengths.

Results

The experiment was successful and the dataset is fully refined. Carbonate is not supported at 750 °C, develops progressively through the 800 °C plateau and grows further at 900 °C, while the Ruddlesden–Popper framework is still the dominant phase after eight hours on stream.

The Pr analogue (PN3) behaves in the opposite way — massive carbonate formation and loss of the framework below 800 °C — but it was measured in an earlier experiment under a milder and different protocol, so the contrast between the two lanthanides cannot yet be attributed to the A-site cation rather than to the activation history. The present run also used a dry activation only, leaving open whether the CO_2 tolerance of LN3 is a property of the composition or something obtained through the activation atmosphere. A new proposal therefore requests the matched 2×2 series — Ln = La, Pr $\times$ dry or humidified activation — reproducing exactly the protocol used here, with the present dataset serving as the internal reference cell.