

Sulfur as pollutant species on the cathode side of a SOFC system

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Sulfur poisoning in a strontium doped lanthanum cobaltite (LSC) cathode current collection layer was revealed in a solid oxide fuel cell (SOFC) tested in repeat-element and stack configuration. Sources of sulfur contamination, other than trace SO_x in air, were identified. Strontium sulfate (SrSO_4) and strontium chromate (SrCrO_4) enriched with sulfur were found at the interface between LSC and air channels.

Understanding degradation mechanisms is a major issue in the development of SOFCs. In particular, in repeat-element and stack configuration, an important coupling of different degradation processes exists, with internal sources at the stack level and exogenous sources coming from system components. A specific diagnostic test station was developed to allow locally-resolved measurements of electrochemical performance and degradation in a repeat-element. Large differences in local degradation behavior were observed, affecting different electrochemical processes. Post-mortem analysis, mainly done by scanning electron microscopy (SEM) and energy-dispersive X-ray spectroscopy (EDX), allowed to identify the pollutant species responsible for degradation. Complete results are published elsewhere [1-2].

In this study, an anode-supported cell, with an active strontium doped lanthanum manganite (LSM) - yttria stabilized zirconia (YSZ) composite cathode and a LSC current collection layer, was tested over 1900 h around 1073 K. Chromium and sulfur were found as major pollutant species on the cathode side; both are dependent on upstream conditions and components. While chromium poisoning is well known for SOFC cathodes, sulfur contamination of the cathode has received more attention recently.

Sulfur poisoning is a major limitation for wide use of perovskite catalysts for treatment of auto exhaust gas [3]. Sulfur pollution from trace SO_x in air and involving sulfate formation was reported by Xu *et al.* [4], where perovskite-type ceramics were used for oxygen permeation membranes. Yokokawa *et al.* identified sulfur as impurity after long-term operation of SOFC stacks in [5], and Xiong *et al.* reported and studied sulfur poisoning of different cathode materials just recently [6].

In the present experiment, a commercial vulcanized polymer tube for air inlet and an insulating high temperature sealing paste were identified as potential sulfur sources. Figure 1 shows the LSC microstructure before and after exposure to sulfur containing air. Strontium sulfate growth on porous LSC surface leads to an almost dense sulfate layer. Strontium sulfate was found over the whole LSC thickness without affecting the LSM/YSZ cathode as shown in figure 2. The more pronounced strontium sulfate formation in LSC compared to LSM is explained by the higher activity of strontium oxide (SrO) in LSC [7].

Chromium on the LSC surface was only found at the air-inlet, indicating that Cr was principally evaporated from upstream system components. SEM/EDX analysis allowed to identify sulfur-rich strontium chromate with a composition close to $\text{Sr}(\text{Cr}_{0.85}\text{S}_{0.15})\text{O}_4$. The chromate structure is changing along the airflow direction, from an almost dense structure

(see figure 2) to isolated particles, while maintaining the same composition. Further analyses are ongoing to confirm sulfate and chromate crystalline structures.

1. Z. Wuillemin et al., ECS Transactions **25** (2009) in press.
2. A.J. Schuler et al., ECS Transactions **25** (2009) in press.
3. L. Wan, Properties and application of perovskite-type oxides, Marcel Dekker Inc., New-York (1993) p145.
4. N. Xu et al., AiChE Journal **45** (1999) p2519.
5. H. Yokokawa et al., ECS Transactions **7** (2007) p133.
6. Y. Xiong et al., J. Electrochem. Soc. **156** (2009) pB588.
7. H. Yokokawa et al., J. Alloys and Compounds **452** (2008) p41.
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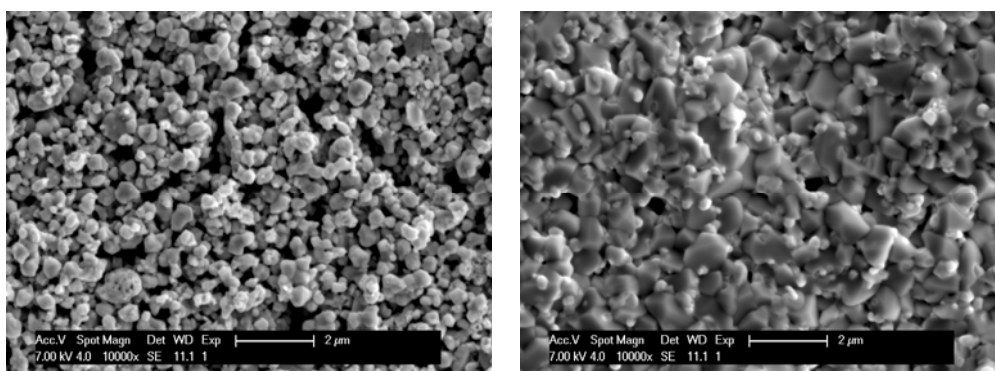


Figure 1. Strontium doped lanthanum cobaltite (LSC) current collector layer surface before (left) and after (right) 1900 h exposure to sulfur containing airflow, where strontium sulfate, SrSO_4 , is formed.

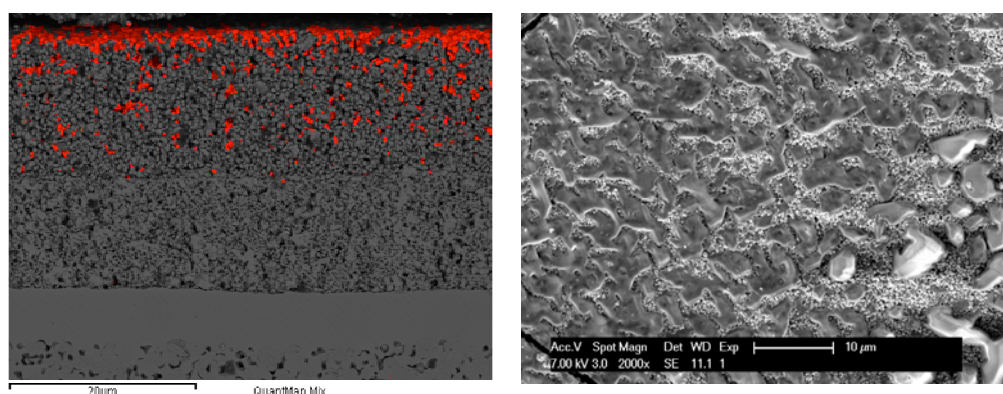


Figure 2. Left: EDX mapping on a polished cathode cross-section reveals strontium sulfate (red spots) formation in the LSC layer, whereas LSM-YSZ composite cathode is free from sulfur poisoning. Right: Sulfur-rich strontium chromate is formed on the LSC surface in an airflow channel at the air inlet.