

## Experiment Report



**Experiment title: In-situ structural investigation of the mechanism of electrochemical graphite transformation into graphite oxide**

**Experiment number:**  
CH-6716

<b>Beamline:</b>	<b>Date of experiment:</b> from: 12 September 2023 to: 15 September 2023	<b>Date of report:</b> 10 November 2023
<b>Shifts:</b>	<b>Local contact(s):</b> Andy Fitch	<i>Received at ESRF:</i>

**Names and affiliations of applicants** (\* indicates experimentalists):

Bartosz Gurzeda,\* Department of Physic, Umeå University, Sweden

Pawel Jezowski,\* Faculty of Chemical Technology, Poznan University of Technology, Poland

Alexandr Talyzin,\* Department of Physic, Umeå University, Sweden

### Report:

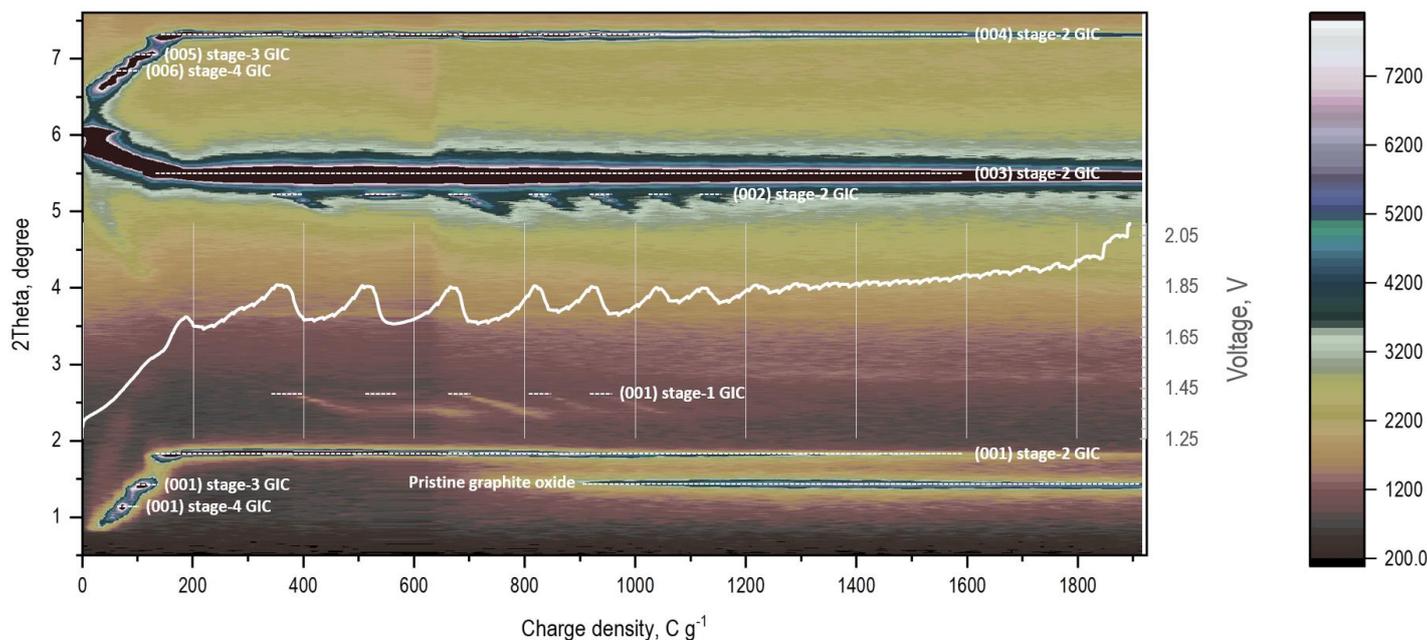
The goal of this project was to study of the mechanism of electrochemical method of graphite oxide synthesis. *In-situ* study of electrochemical GO formation in aqueous H<sub>2</sub>SO<sub>4</sub> and HClO<sub>4</sub> electrolytes was performed. The new findings and conclusions will provide the fundamental understanding of electrochemical oxidation of graphite and contribute to more wide use of electrochemical oxidation to produce high quality graphite/graphene oxide.

Experiments were performed at ID22 beamline using “micro” electrochemical reactor made of glass capillary (Fig. 1). Graphite flakes were placed on the platinum current collector and hold by glass fibre membrane. Reactor was filled with the electrolyte (11 M H<sub>2</sub>SO<sub>4</sub> or 8 M HClO<sub>4</sub>) and platinum wire was used as a counter electrode. Graphite was electrochemically oxidized using galvanostatic method.

The electrochemical transformation of graphite into graphite oxide was successfully performed in sulfuric acid electrolyte. Galvanostatic curve shows that sample was sensitive to X-ray beam (Fig. 2 – white curve).



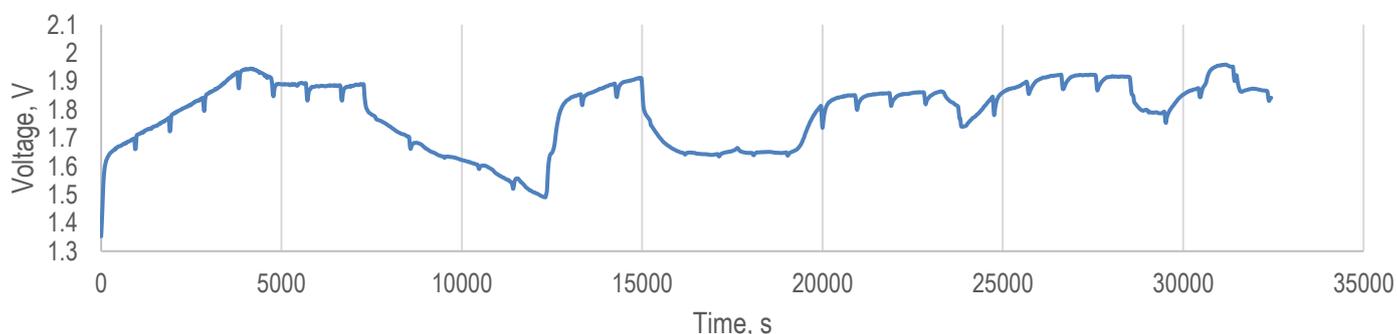
**Fig. 1.** Electrochemical reactor



**Fig. 2.** In-situ XRD data recorded during the electrochemical oxidation of graphite in 11 M  $\text{H}_2\text{SO}_4$  electrolyte. Presented XRD data is combined with galvanostatic curve recorded during graphite electrochemical oxidation.

At the beginning of electrooxidation of graphite, the formation of second stage graphite intercalation compound (GIC) was observed. Next, potential start to oscillate around voltage 1.85 V. As can be seen from the XRD data presented in Fig. 2, When the maximum of voltage oscillation was reached the formation of stage-1 GIC with sulfuric acid started. After partial formation of stage-1 GIC, voltage decreased and simultaneously the maxima of peaks assigned to stage-1 GIC started to shift to lower  $2\theta$  angles (the interlayer distances increased). Such a behaviour is most likely caused by water co-intercalation between the graphene layers occupied by sulfuric acid and the partial formation of graphite oxide. When the charge density reached  $900 \text{ C g}^{-1}$ , the signal assigned to pristine graphite oxide (interlayer spaces of graphite oxide are occupied by sulfuric acid) appeared. With the increase of the oxidation time, the intensity of signal originating from graphite oxide increases and at the same time the intensity of the signals assigned to stage-2 GIC decreases. Full transformation of GIC to graphite oxide was not achieved, however the recorded data allows us to propose the mechanism of electrochemical graphite oxide formation in aqueous sulfuric acid.

In the case of graphite electrochemical oxidation in 8 M  $\text{HClO}_4$ , the system was too sensitive to the beam. The potential was changing dramatically during the electrooxidation of graphite and no voltage oscillations can be recorded (Fig. 3). In summary, the registration of structural changes during graphite electrochemical oxidation in aqueous perchloric acid was unsuccessful.



**Fig. 3.** Galvanostatic curve recorded during the electrochemical oxidation of graphite in 8 M  $\text{HClO}_4$ . Drops in the voltage were caused by the exposure of graphite to the beam.

More detailed analysis of gathered XRD data will be performed at the end of 2023. The recorded data reveals that the mechanism of graphite oxide electrochemical formation in aqueous sulfuric acid is different than the one proposed in the literature. Although the electrochemical oxidation of graphite in aqueous perchloric acid was not achieved, we consider that our experiment was successful, and the data recorded during the electrochemical oxidation of graphite in 11 M  $\text{H}_2\text{SO}_4$  will be used for preparation of scientific publication.