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Oscillating structural transformations in the electrochemical synthesis of graphene oxide from graphite

Bartosz Gurzęda, P. Jeżowski, N. Boulanger, A. V. Talyzin

Coupling the in-situ XRD analysis with electrochemical oxidation of graphite to monitor structural changes in its crystal lattice

25 June 2026, Gliwice, Poland

Institute for Resource Ecology | b.gurzeda@hzdr.de

In-situ synchrotron radiation XRD investigation of the structural transformations of graphite

Graphite intercalation

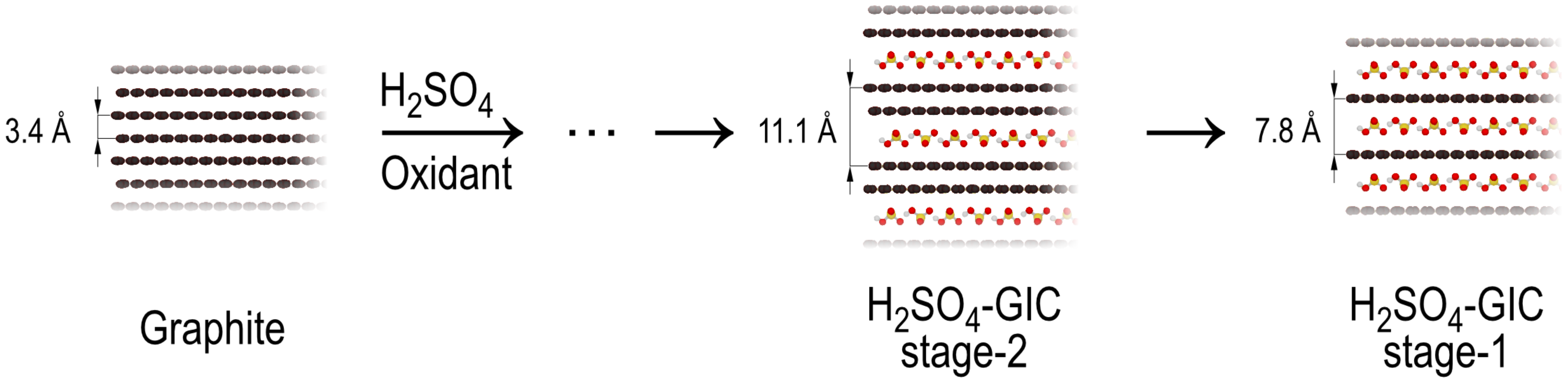

Fig. 1 Scheme of sulfuric acid intercalation into graphite.

GIC – graphite intercalation compound

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Electrochemical graphite oxidation in aqueous sulfuric acid

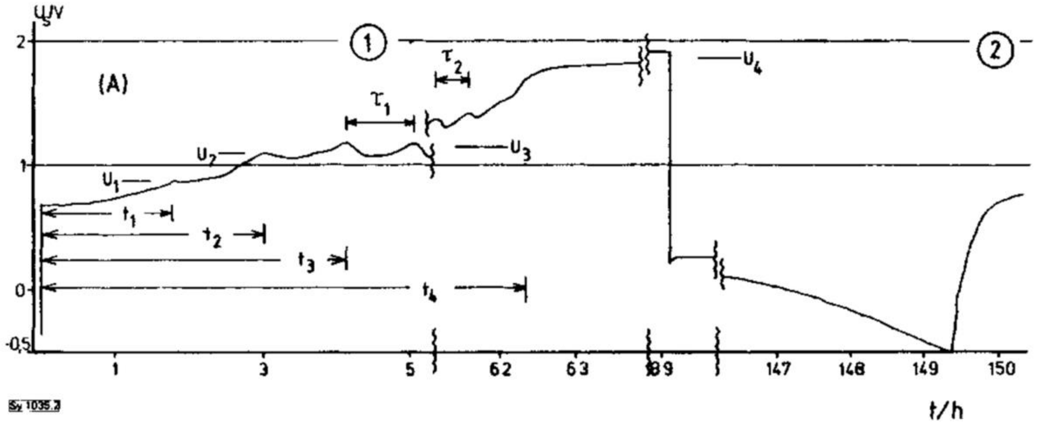

Abb. 2. Verlauf des Potentials U_s bei einer Festbett-Elektrode aus Naturgraphit: 1. Zyklus bei einem Versuch mit hohen Umsätzen (1600%), entsprechend einer formalen Stöchiometrie $C_{1,5}A$; $j = 5$ mA/cm^2 in 12 M H_2SO_4.

H. Krohn, F. Beck, R. Herrmann,
Chemie Ingenieur Technik 1982, **54**, 530–531.

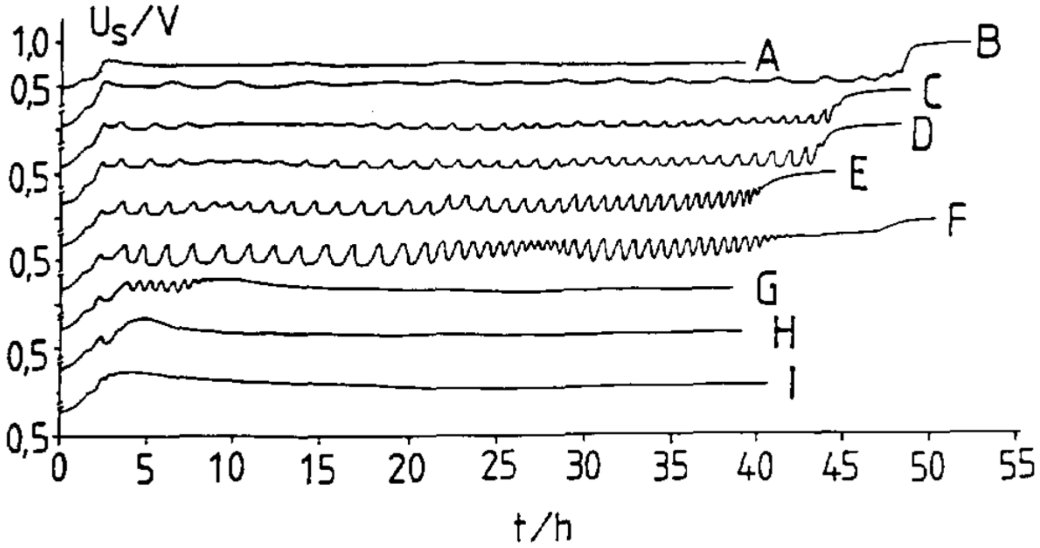

Fig. 1. Potential oscillation curves of natural graphite during anodic charging in aqueous sulphuric acid at various concentrations: (A) 13 M; (B) 12.5 M; (C) 12 M; (D) 11.5 M; (E) 11 M; (F) 10.5 M; (G) 10 M; (H) 9.5 M; (I) 9 M. Electrode, Pt mesh + graphite ($C = 99.95\%$, $m = 0.0430$ g, $A = 0.5$ cm^2); current, 1.0 mA.

F. Beck, J. Jiang, H. Krohn,
Journal of Electroanalytical Chemistry 1995, **389**, 161–165.

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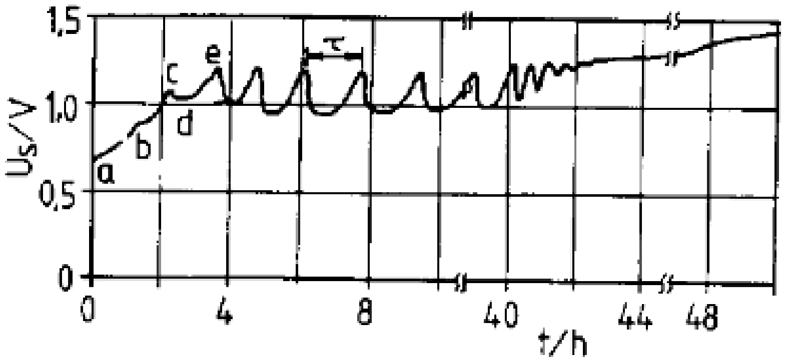
Electrochemical graphite oxidation in aqueous sulfuric acid



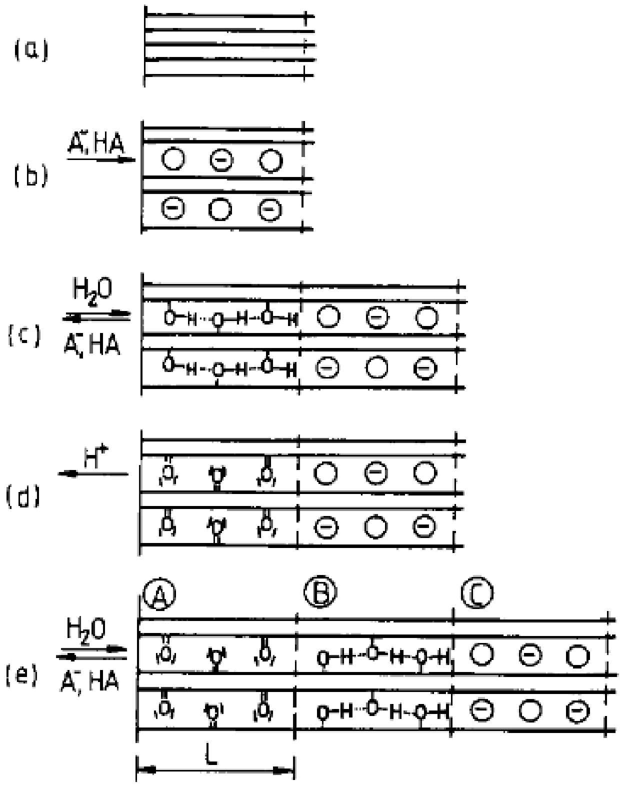


Fig. 3. Zone model of the formation of C–OH groups with hydrogen bonds and their conversion to C=O groups in the intercalate layers during the galvanostatic charge of graphite in aqueous sulphuric acid (– HSO_4^- anion; ○ H_2SO_4 molecule): (a) graphite; (b) graphite intercalation; (c) first potential maximum; (d) first potential minimum; (e) next potential maximum.

F. Beck, J. Jiang, H. Krohn, *Journal of Electroanalytical Chemistry* 1995, **389**, 161–165.

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Electrochemical setup for studies of graphite in-situ structural transformations

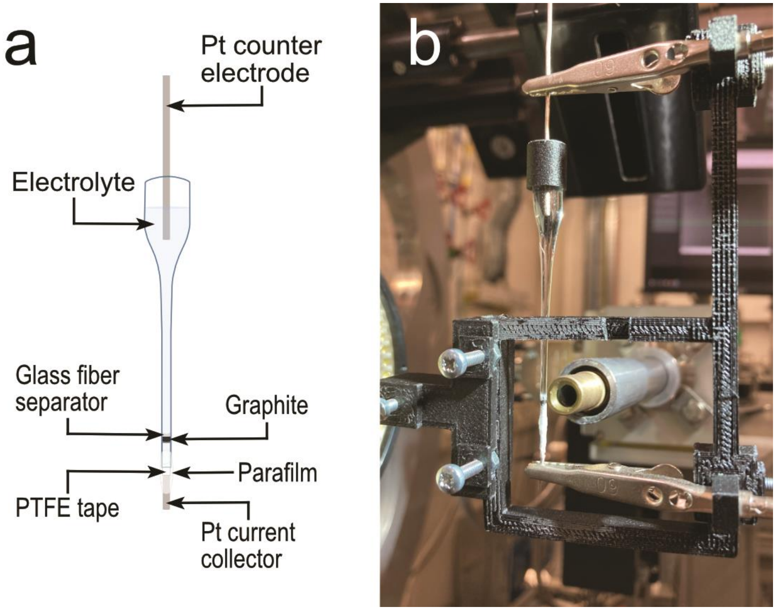


Fig. 2 a) Scheme of the micro-electrochemical reactor. b) Photo of the installed reactor aligned in synchrotron radiation beam.

Mass of graphite electrode: ~280 µg
Current: 40 µA
Electrolyte: 11 M H_2SO_4

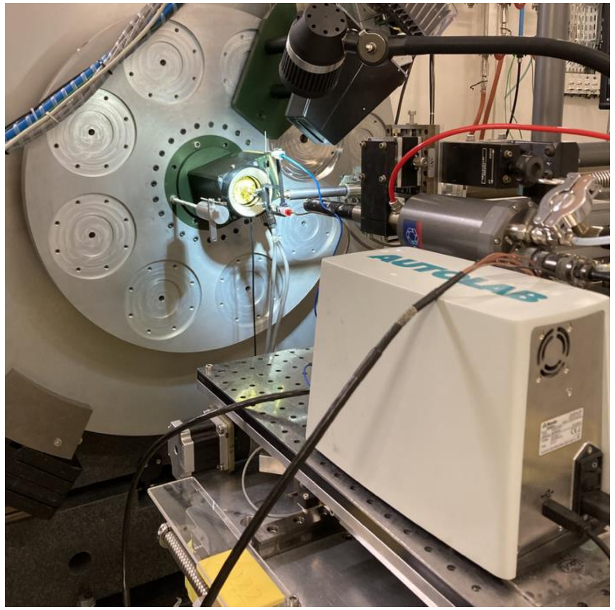


Fig. 3 Photo of the installed reactor connected to the potentiostat-galvanostat at beamline ID22 (ESRF).

B. Gurzęda, P. Jeżowski, N. Boulanger, A. V. Talyzin, *Angewandte Chemie International Edition* **2024**, e202411673.

In-situ synchrotron radiation XRD investigation of the structural transformations of graphite

Electrochemical oxidation of graphite in the capillary-size reactor – X-ray beam damage

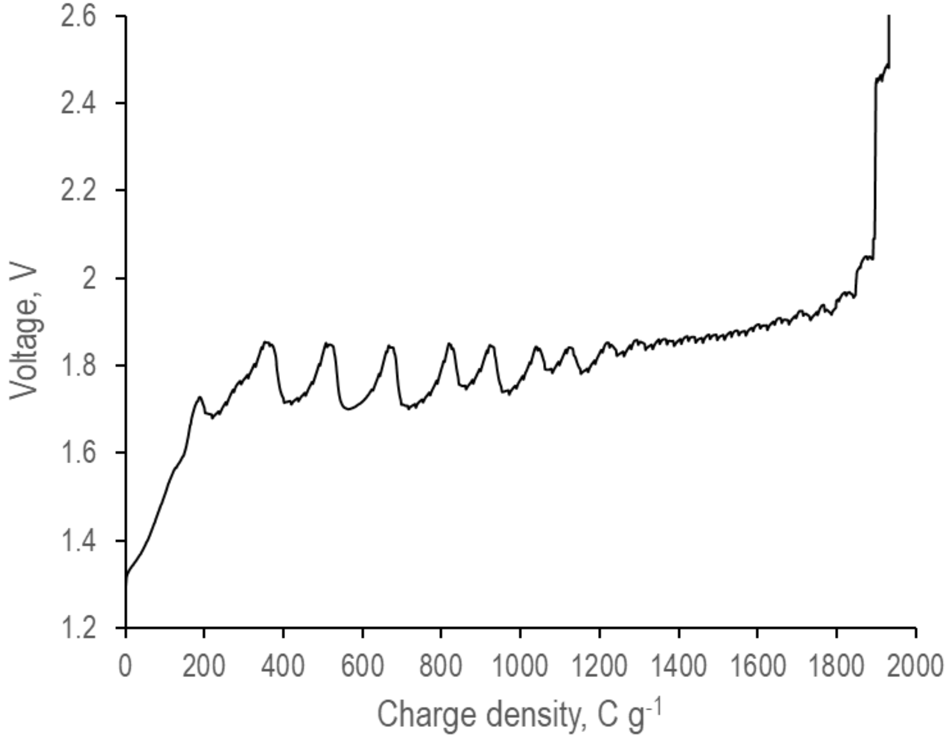


Fig. 4 The galvanostatic curve showing the X-ray beam damage of formed H_2SO_4-GIC.

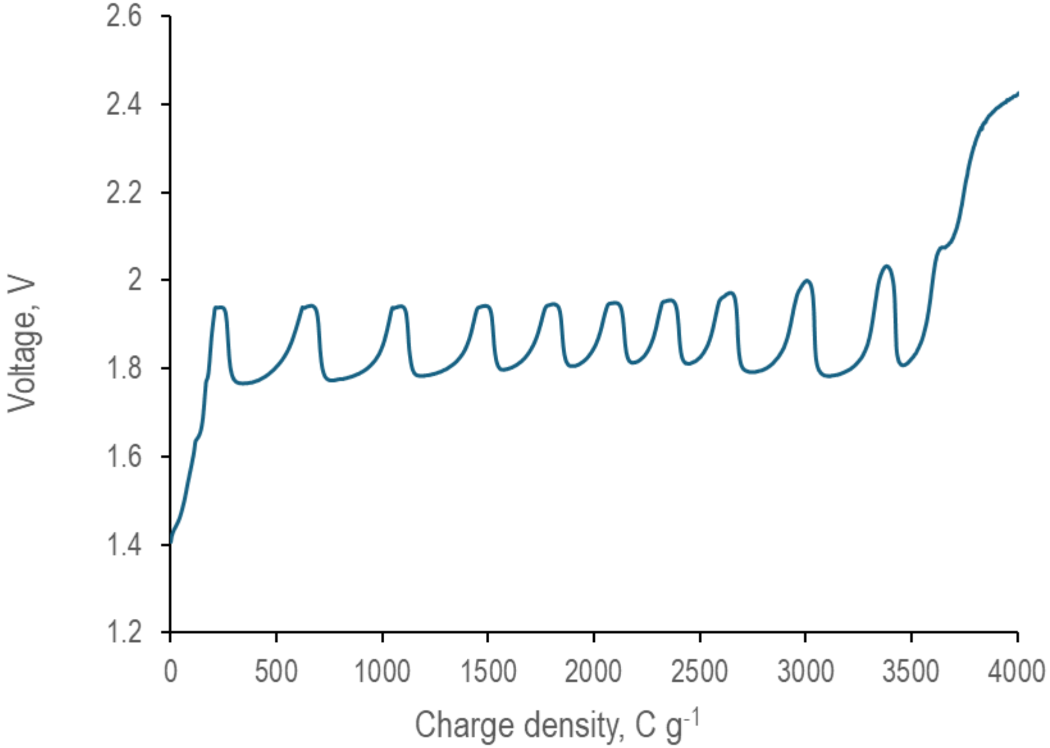


Fig. 5 The galvanostatic curve recorded during in-situ XRD analysis of graphite oxidation with cooling of the electrochemical reactor (no X-ray beam damage were observed).

In-situ synchrotron radiation XRD investigation of the structural transformations of graphite

Electrochemical oxidation of graphite in the capillary-size reactor

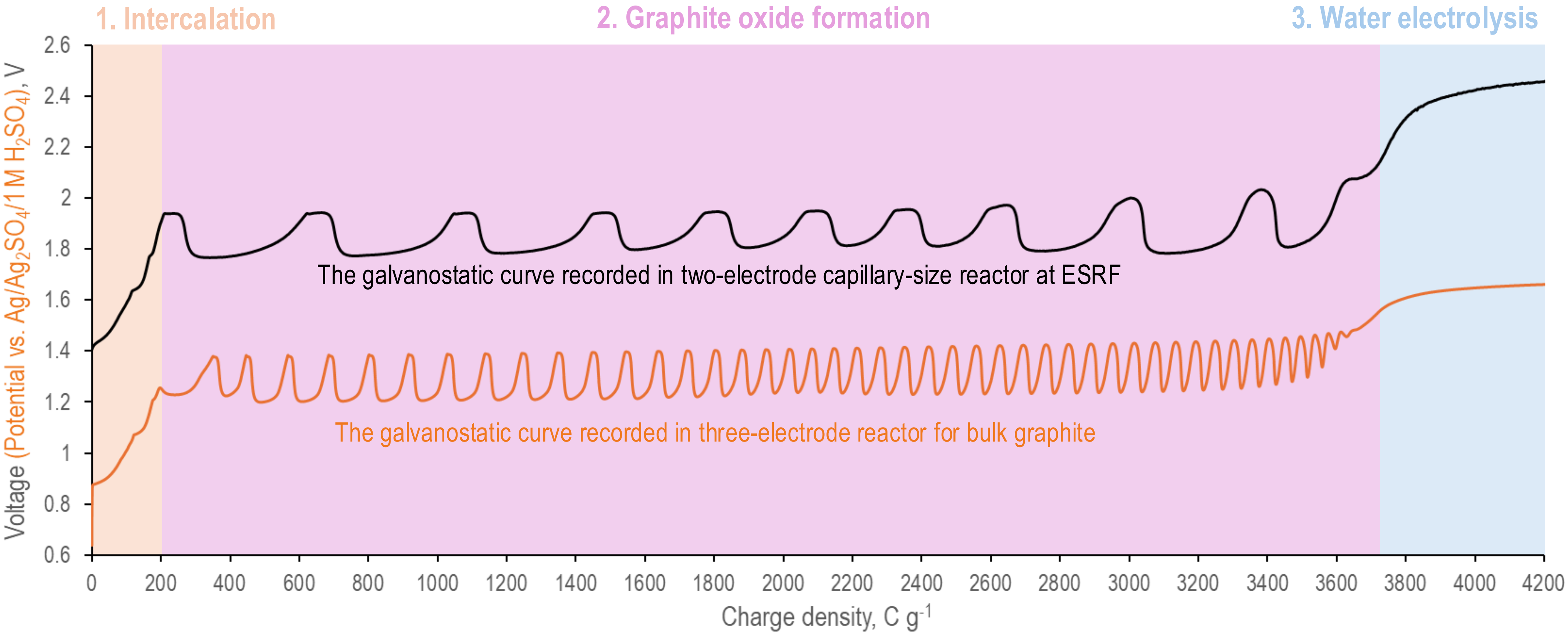


Fig. 6 The galvanostatic curves recorded during the electrochemical oxidation of graphite in 11 M H_2SO_4.

In-situ synchrotron radiation XRD investigation of the structural transformations of graphite

Electrochemical formation of H_2SO_4-GIC

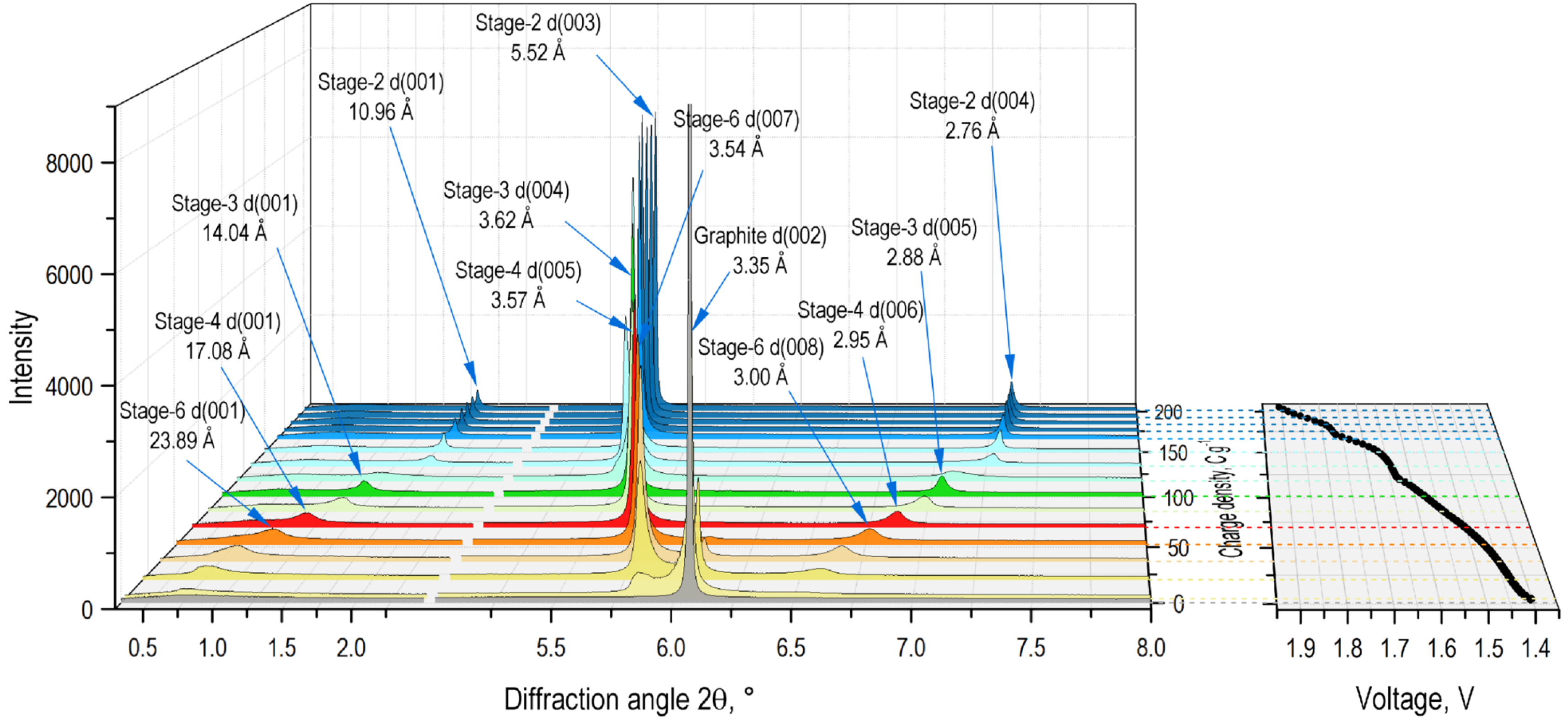


Fig. 7 Synchrotron radiation XRD patterns recorded during the beginning of graphite oxidation showing the changes in graphite intercalation compound (GIC) staging until pure stage-2 GIC is formed. Interlayer d-spacings 23.89, 17.08, 14.04, and 10.96 Å are assigned to stage-6, stage-4, stage-3 and stage-2 GIC, respectively (λ=0.35435 Å).

In-situ synchrotron radiation XRD investigation of the structural transformations of graphite

Electrochemical formation of graphite oxide

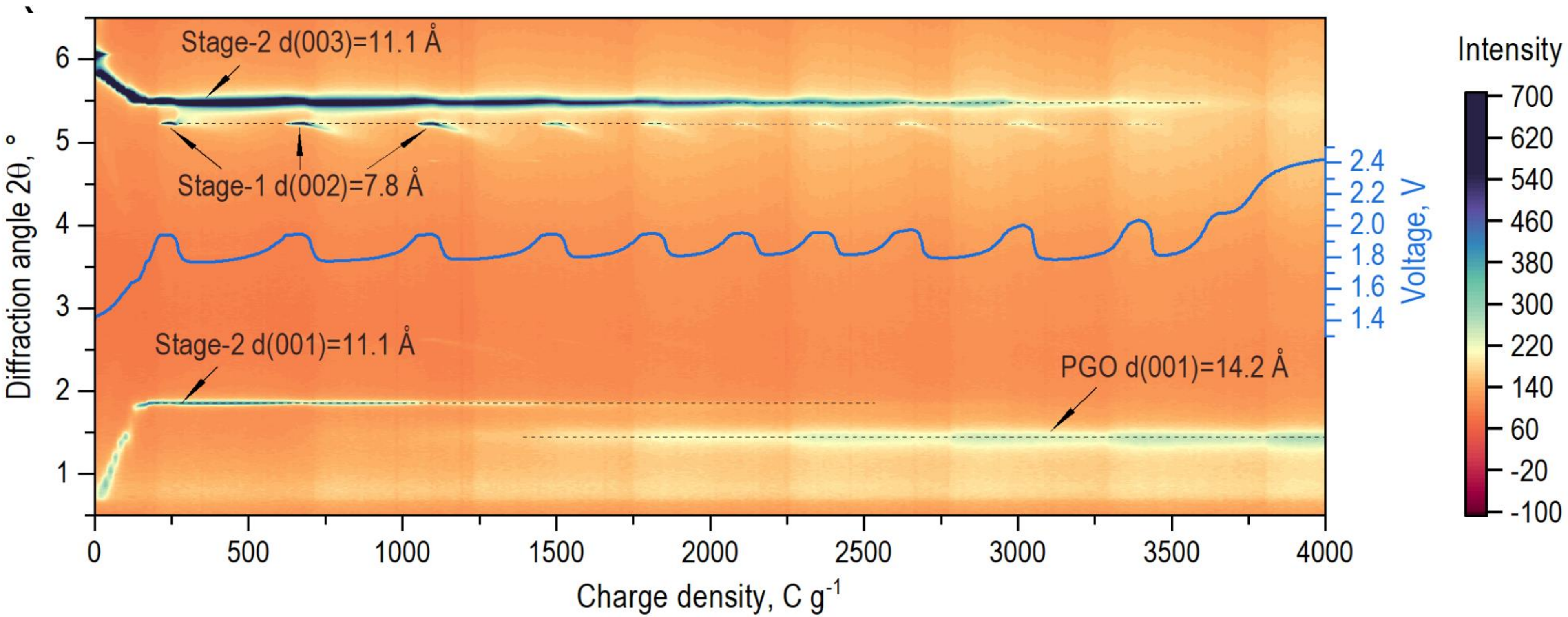


Fig. 8 “Heat map” constructed using XRD patterns (λ=0.35435 Å) recorded during the electrochemical graphite oxidation combined with the galvanostatic curve showing voltage oscillations.

B. Gurzęda, P. Jeżowski, N. Boulanger, A. V. Talyzin, *Angewandte Chemie International Edition* **2024**, e202411673.

In-situ synchrotron radiation XRD investigation of the structural transformations of graphite

Electrochemical formation of graphite oxide –changes in the graphite surface

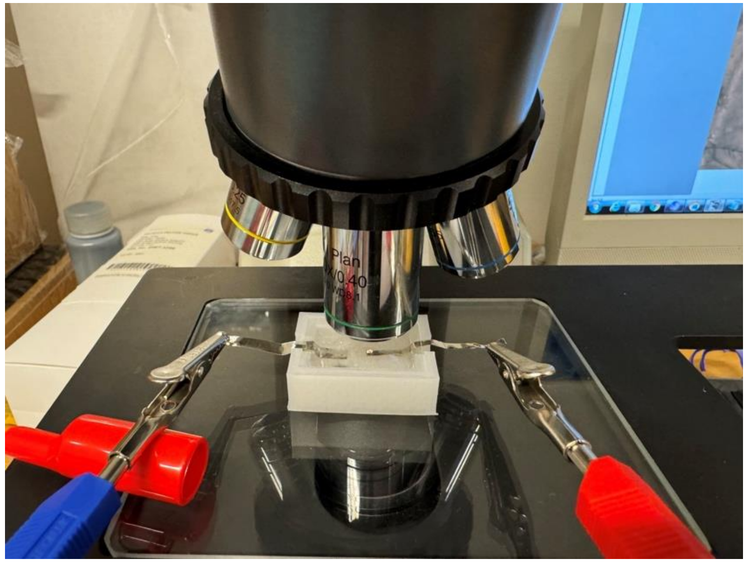
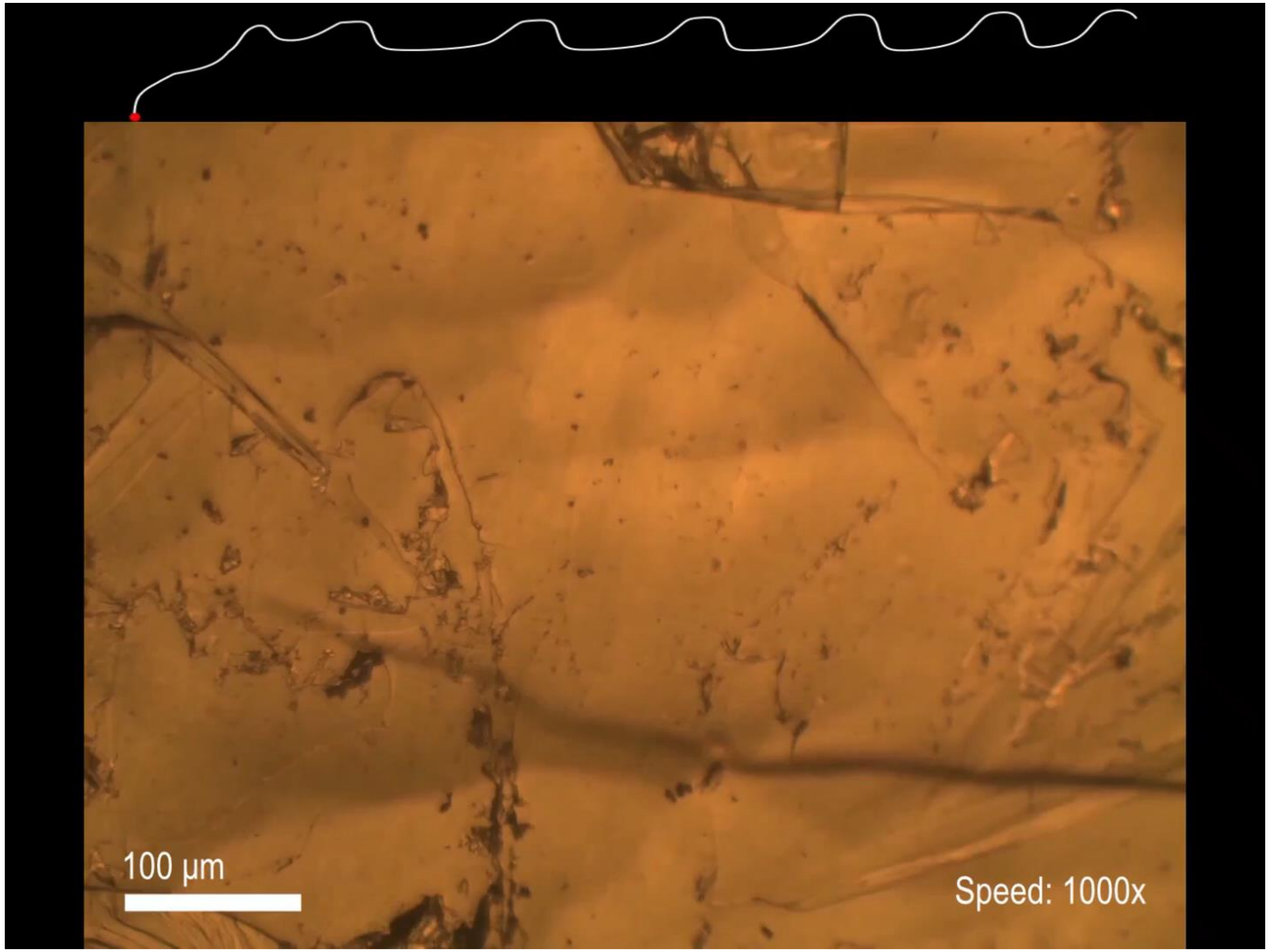


Fig. 9 Photo of 3D printed PE reactor used to observe the changes in the graphite surface caused by electrochemical oxidation in 11 M H_2SO_4.



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Mechanism of the electrochemical graphite oxide formation

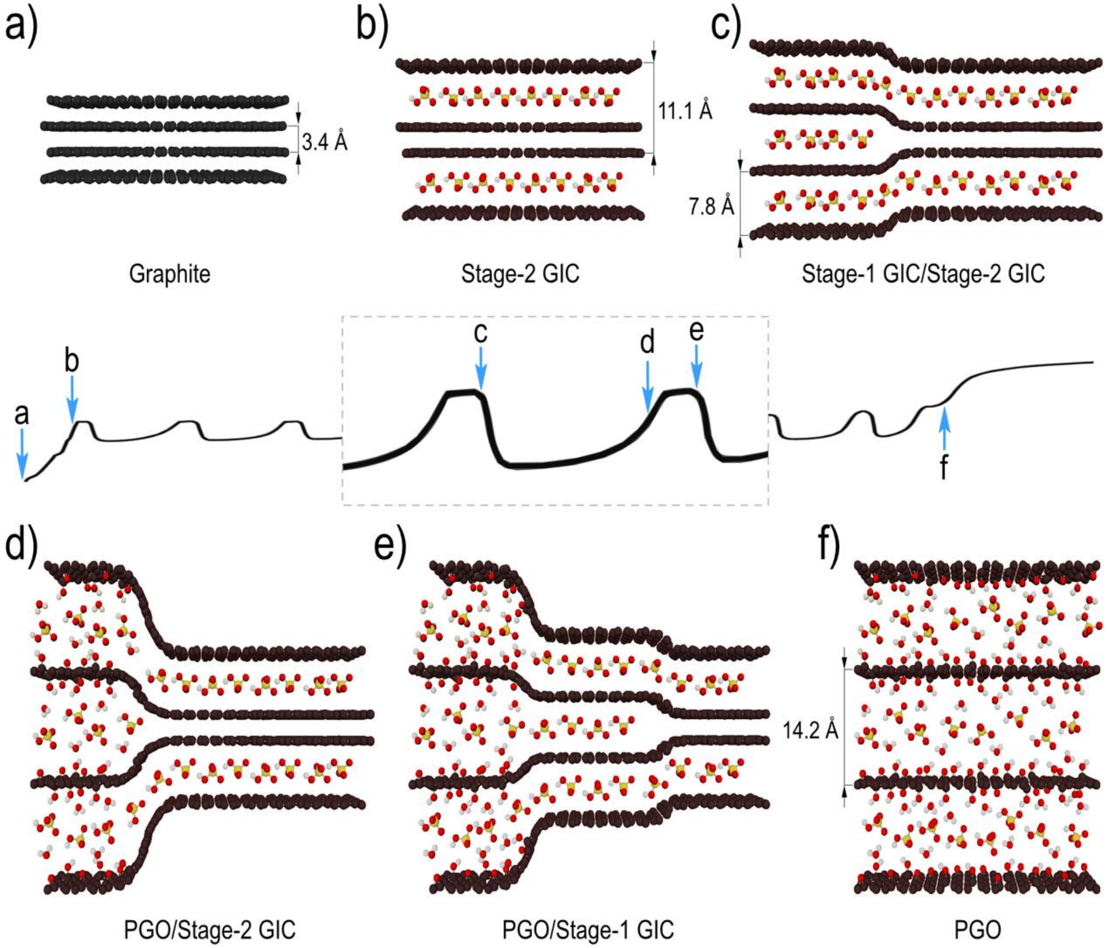

Fig. 10 The Scheme illustrating mechaism of GO formation through the periodic structural transformations observed during electrochemical oxidation of graphite.

In-situ synchrotron radiation XRD investigation of the structural transformations of graphite

Conclusions

- **In-situ synchrotron XRD analysis of the electrochemical oxidation of graphite in aqueous sulfuric acid reveals the periodic appearance and disappearance of the solid phase recognized as stage-1 H_2SO_4-GIC. The observed phenomenon can be considered as an oscillating reaction.**

- **The potential oscillations are caused by the competition between two reactions:**
  1. **Formation of stage-1 H_2SO_4-GIC**
  2. **H_2O co-intercalation and formation of oxygen functional groups on graphene layers surface.**

- **The mechanism of GO electrochemical formation in aqueous sulfuric acid was updated.**

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Thank you for your attention!

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