Structure and morphology of thermally reduced graphene oxide under high temperatures

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This work presents a detailed study of the structural and morphological evolution of thermally reduced graphene oxide (trGO) [1, 2] subjected to high-temperature treatment at 500 °C and 700 °C. The resulting materials were characterized by Transmission electron microscopy, X-ray diffraction and Raman spectroscopy.

Transmission electron microscopy (TEM) demonstrates that thermal reduction induces pronounced morphological evolution of graphene oxide. At 300 °C, trGO retains a highly functionalized and loosely packed structure containing numerous oxygen-containing groups distributed over graphene sheets. Annealing at 500 °C results in significant deoxygenation accompanied by wrinkling, folding, and defect formation. Further heating to 700 °C produces a more compact and partially graphitized structure characterized by closely stacked graphene layers, reduced interlayer spacing, and the formation of corrugated ("rippled") graphene sheets. These observations indicate that thermal treatment simultaneously promotes significant structural changes and defect generation.

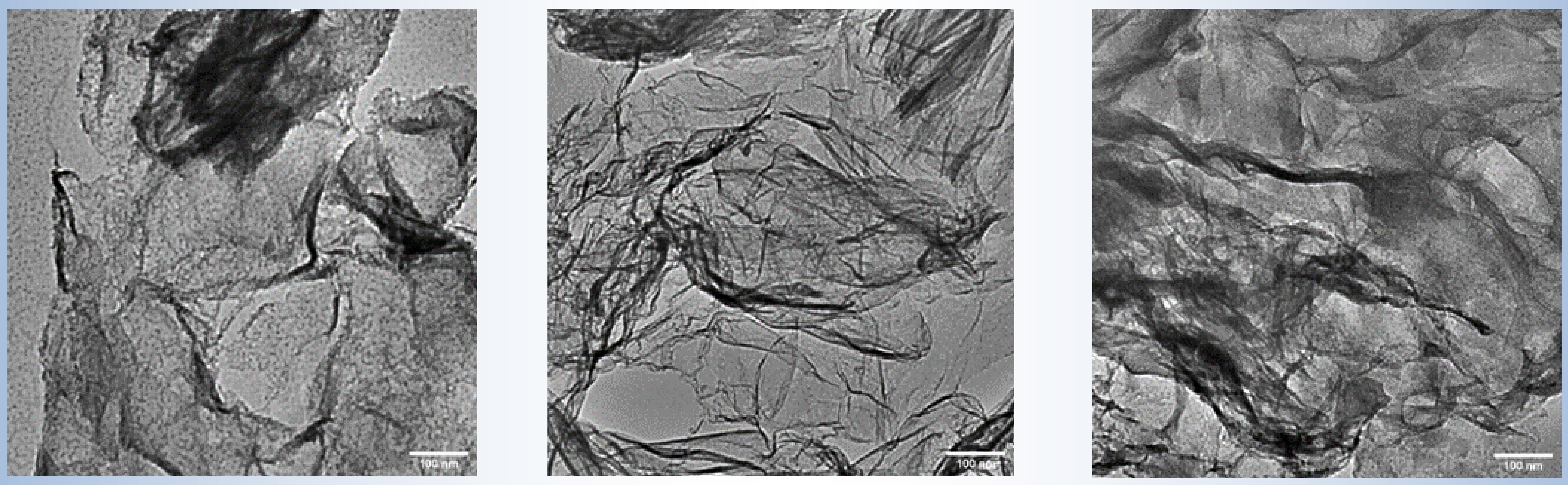

Fig. 1. TEM images of powder sample trGO_300 (a), trGO_500 (b) and trGO_700 (c), which were obtained by thermal treatment at different temperatures: 300°C, 500°C, and 700°C, respectively. [3]

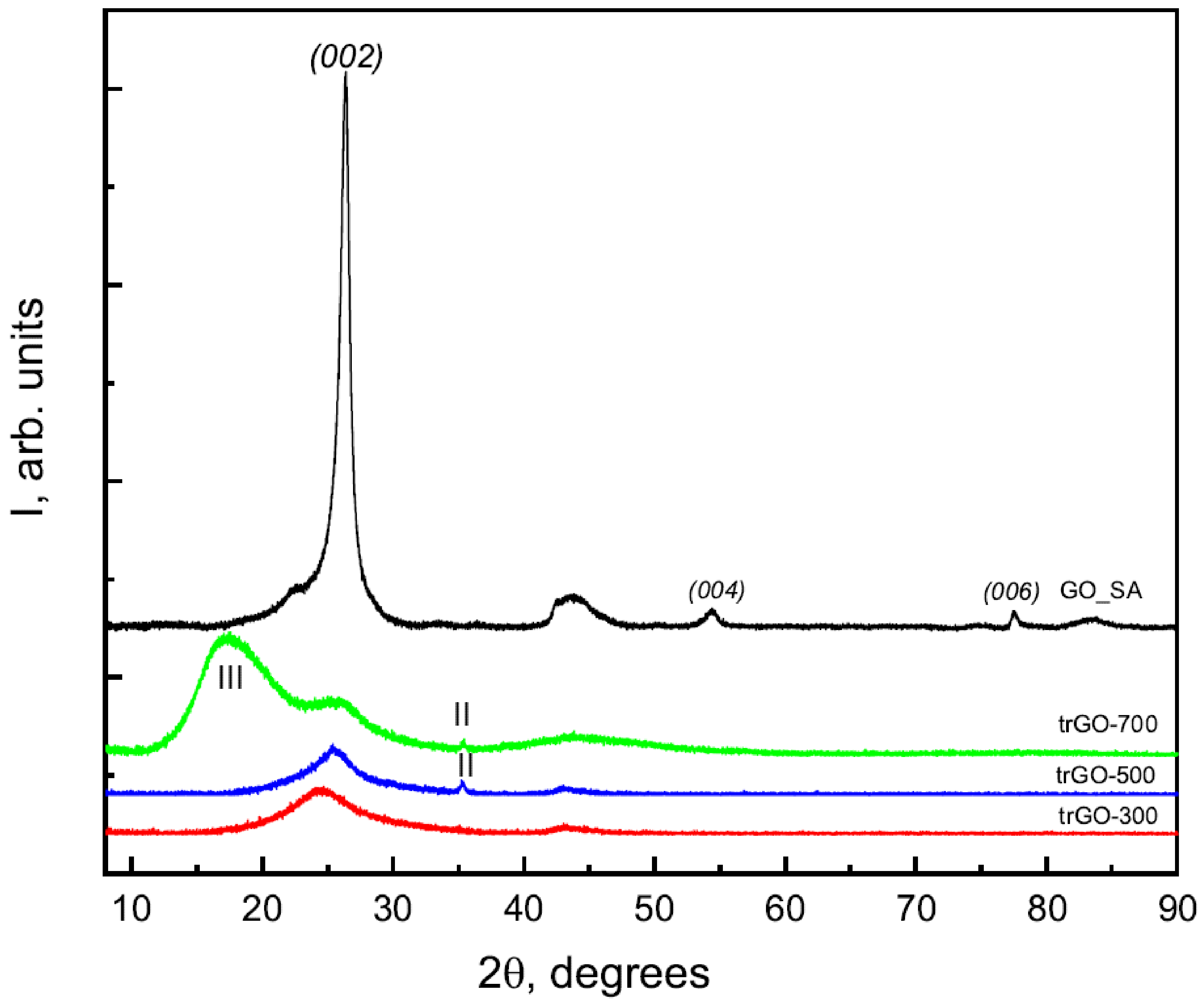

Fig. 2. X-ray diffraction patterns obtained from the GO SA, trGO 300, trGO 500, and trGO 700 samples (shown from bottom to top). XRD pattern II indicates the formation of a minor impurity phase. XRD pattern III shows the appearance of "ripples" and the wrinkles in the trGO. The patterns are vertically offset for clarity.

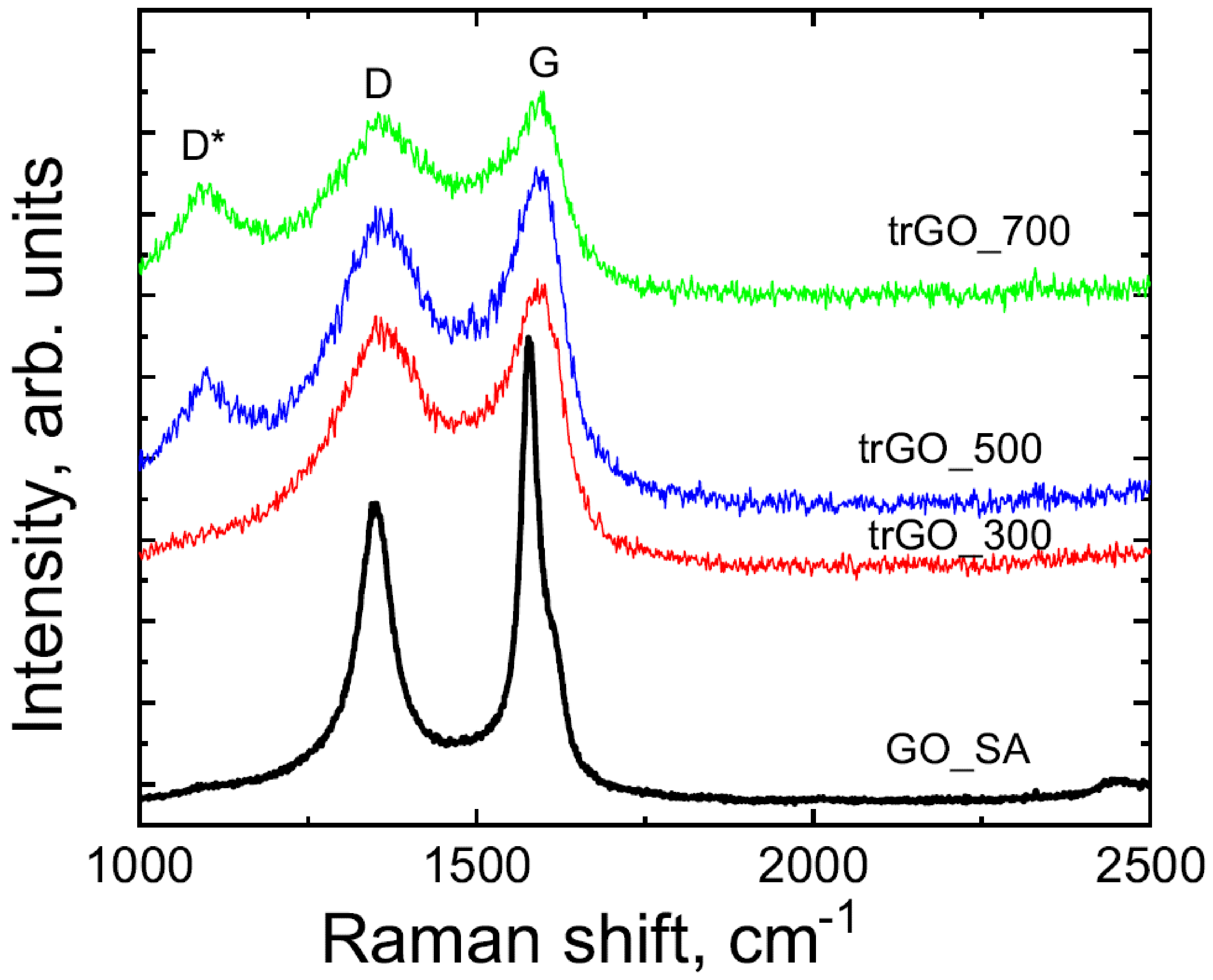

Fig. 3. Raman spectra of GO SA, trGO 300, trGO 500, and trGO 700 samples (shown from bottom to top). The spectra are offset from each other for clarity.

XRD analysis demonstrates a gradual shift of the broad (002) diffraction maximum toward higher diffraction angles with increasing temperature. The corresponding decrease in interlayer spacing from 3.63 Å for trGO300 to 3.49 Å for trGO700 indicates the removal of intercalated water and oxygen-containing groups, accompanied by closer stacking of graphene layers along with increasing disorder. An additional diffraction feature observed in trGO700 suggests the formation of thermally induced lattice distortions associated with corrugated graphene structures (formation "ripples").

Raman spectroscopy confirms the defect-mediated structural evolution of trGO. The characteristic D and G bands are present in all samples, while the increase in the intensity ratio ID/IG from approximately 3.98 for trGO300 to 5.21 for trGO700 indicates a significant increase in point-defect concentration generated during high-temperature reduction. Simultaneously, the evolution of Raman spectra reflects the gradual restoration of sp^2-carbon domains and partial graphitization of the material.

The combined TEM, XRD, and Raman results demonstrate that thermal annealing above 500 °C promotes two competing processes: restoration of graphitic ordering and generation of structural defects. As a result, trGO700 exhibits the highest degree of structural changes, characterized by reduced interlayer spacing on *(002)* plane, increased defect density, and the formation of corrugated graphene layers.

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[2] A. V. Dolbin et. al., Low Temp. Phys. 46, 293 (2020); https://doi.org/10.1063/10.0000701.
[3] A.I. Krivchikov, A. Jeżowski, M.S. Barabashko et al., Nanoscale and Microscale Thermophysical Engineering, 29 (4), P. 219-233 (2025), https://doi.org/10.1080/15567265.2026.2635099.

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