Supporting information

Discovery of Natural Few-Layer Graphene on the Moon

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I. Some background for the CE-5 sample

Recent studies have identified Basalt (*1*), ilmenite ($FeTiO_3$) (*2*), pentlandite (*3*), amorphous features (*4*), and even photosynthetic catalysts(*5*) in CE-5 lunar soil.

(1) H. Cao, C. Wang, J. Chen, X. Che, X. Fu, Y. Shi, D. Liu, Z. Ling, L. Qiao, X. Lu, Xiaobin Qi, C. Yin, P. Liu, C. Liu, Y. Xin, J. Liu, A Raman spectroscopic and microimage analysis perspective of the Chang'e-5 lunar samples. *Geophys. Res. Lett.* **49**, e2022GL099282 (2022).

(2) J.-H. Li, Non-destructive identification and quantification of ilmenite from a single particle of the Chang'e-5 lunar soil sample. *At. Spectrosc.* **43**, 283-290 (2022).

(3) X. Tang, H. Tian, S. Sun, L. Gu, Q. Li, X. Li, J. Li, Origin and implication of pentlandite in Chang'e-5 lunar soils. *Lithos* **458-459**, 107342 (2023).

(4) J. Xi, L. Ma, H. Xian, G. Wang, J. Xing, J. Wei, J. Zhu, H. He, In situ micro-XRD methods for identifying glass and minerals in extraterrestrial samples. *At. Spectrosc.* **43**, 19-27 (2022).

(5) Y. Yao, L. Wang, X. Zhu, W. Tu, Y. Zhou, R. Liu, J. Sun, B. Tao, C. Wang, X. Yu, L. Gao, Y. Cao, B. Wang, Z. Li, W. Yao, Y. Xiong, M. Yang, W. Wang, Z. Zou, Extraterrestrial photosynthesis by Chang'E-5 lunar soil. *Joule* **6**, 1008-1014 (2022).

II. Materials and Methods

Abbreviation of Terminology

SEM: scanning electron microscopy
TEM: transmission electron microscopy
HRTEM: high-resolution transmission electron microscopy
HAADF: high-angle annular dark-field
STEM: scanning transmission electron microscopy
EDS: energy dispersive X-ray spectroscopy
EELS: electron energy loss spectroscopy
TOF-SIMS: time-of-flight secondary ion mass spectrometry

Laser scanning confocal microscopy (LSCM) image was performed on an OLS 5100 LSCM. TESCAN All-in-One System (TESCAN S9000G) was used to obtain the information via SEM (including secondary-electron and backscattered electron) images, EDS (Oxford Instruments), Raman spectra (WITec alpha300 R) and the TOF-SIMS result. GeminiSEM 360 was employed to analyze the morphology.

TEM and EDS were obtained with a TEM (JEM-2100F). HAADF-STEM, HRTEM, and EELS characterizations were performed on a spherical aberration (Cs) corrected TEM (JEM ARM300F Grand ARM), with an accelerating voltage of 300 kV. The low-order aberrations have been tuned to an acceptable level that A1 is <2 nm, Cs is <0.5 μm, A2 is <10 nm. The TEM samples were acquired by scraping the lunar sample, followed by the deposition process on a holey carbon film for microscopy observation. Since electron beam radiation damage is unfavorable for the analysis of the original structure of samples in TEM characterization, Fischione's Cryo-holder 2550 was employed to reduce the radiation damage as much as possible.

Supplementary figures

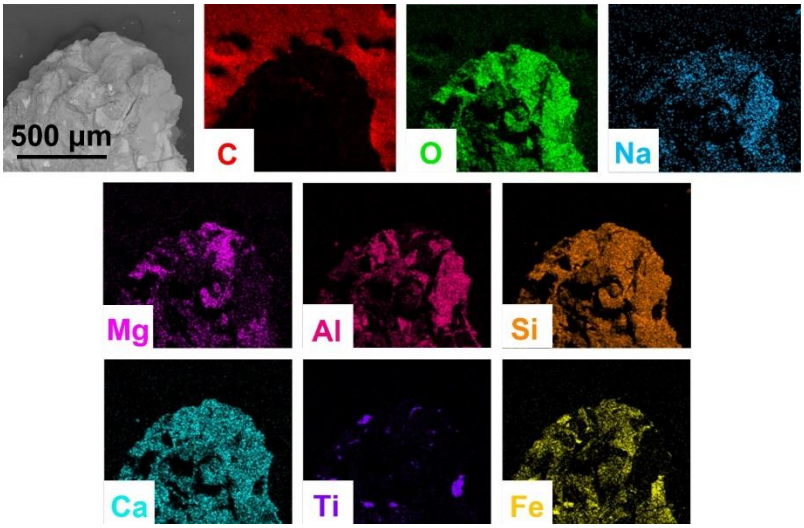

Figure S1 SEM image and the corresponding EDS elemental mapping analysis of the CE-5 lunar soil sample.

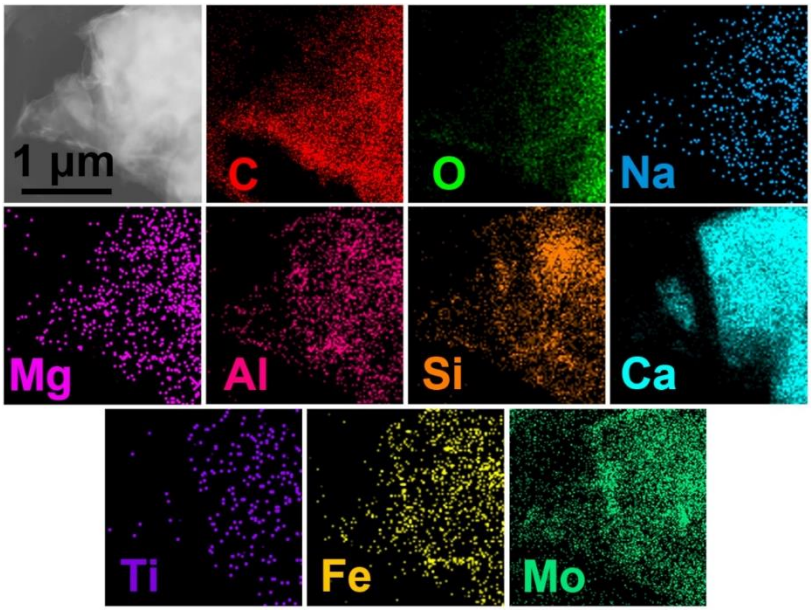

Figure S2 STEM image and the corresponding EDS elemental mapping analysis of the CE-5 lunar soil sample.

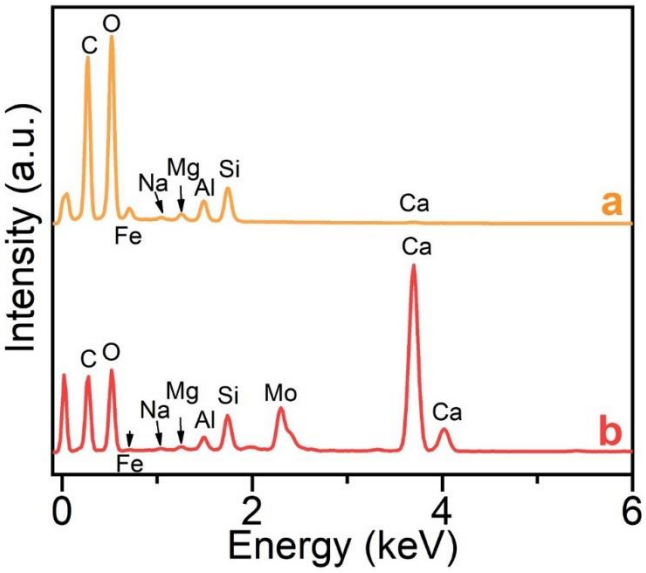

Figure S3 EDS spectra of different areas in (a, Fig. S1) and (b, Fig. S2) of the CE-5 lunar soil sample.

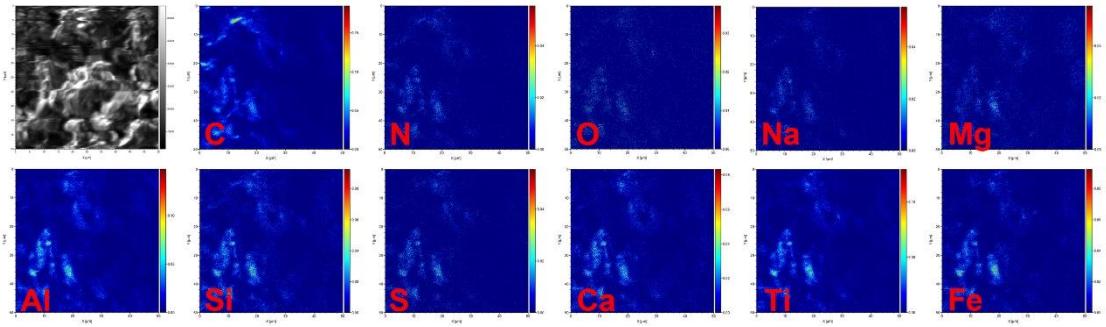

Figure S4 Correlative SEM/TOF-SIMS (time-of-flight secondary ion mass spectrometry) result of the CE-5 lunar soil sample for the distribution of elements.

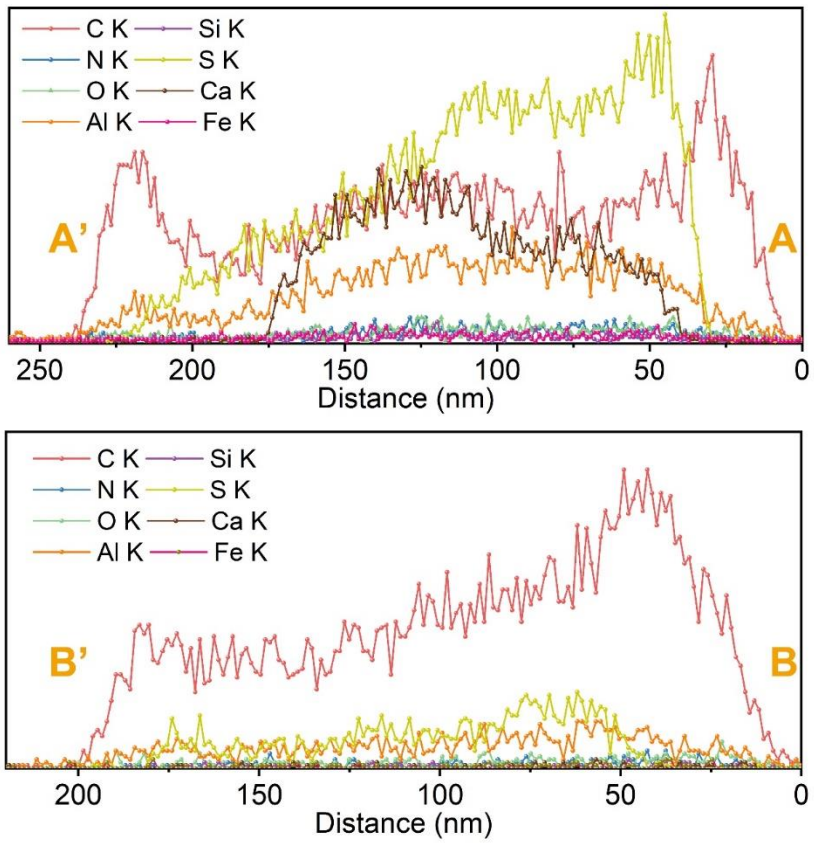

Figure S5 EDS line profiles of the core-shell structure, showing fluctuated spatial distribution of the elements.

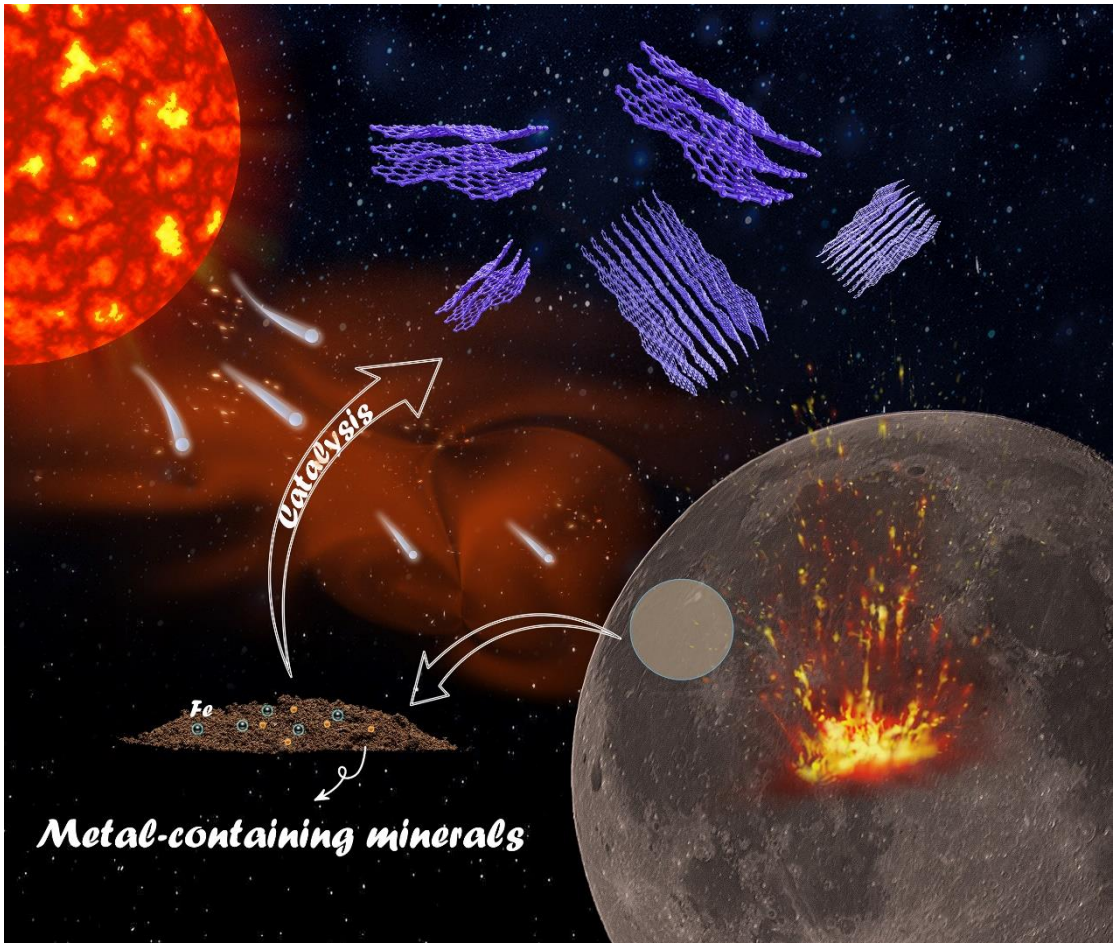

Figure S6 Illustration of the possible formation process of graphene on the Moon.