Supplementary Information

Acoustic shock wave induced sp^3-to-sp^2 type solid state phase transition: a case study of tetrahedral amorphous carbon

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Experimental section

Tetrahedral amorphous carbon (*ta*-C)

The test sample *ta*-C sample has been purchased from the Sigma Aldrich Company (Carbon purity - 99%, grain size ~10 μm, Porosity - 14%).

Shock wave loading experiment by shock tube

The required shock waves have been generated by an in-house tabletop semiautomatic shock tube which has three sections known as driver, driven and diaphragm sections that are made of seamless steel. The driver and driven sections consist of long tubes of 48 cm and 180 cm, respectively and both have an inner diameter of 1.5 cm. Atmospheric air is used as the input source for the required shock wave generation. The diaphragm section separates the driver section and the driven section. Carbonless paper diaphragms are fed into the diaphragm section with the help of a motor. While the atmospheric air is being compressed into the driver section, at the critical pressure, the diaphragm is ruptured such that the shock wave is generated and moves along the driven section.

For the shock loading, the sample pack with the dimension 10×10×1 mm^3 are rigidly fixed in the sample holder which is fixed 1cm apart from the open end of the shock tube. Totally, 3 equal amount of samples have been chosen such that one sample has been kept as the control sample. While the other two samples have been treated with 500 shocks with the Mach number of 2.2 and 4.7. Fig. S1 (a and b) shows the schematic diagram of the experimental setup of the shock tube and in Fig. S2, the acoustic wave signals are presented which are recorded in the driven section. Subsequently, 500 shock pulses have been loaded on the test sample with an interval of 5 sec between each shock pulse. For example, 500 pulses mean shock wave-exposed

on a sample 200 times with Mach number 4.7 (±0.1). For the present experiment, the shock waves of Mach number 4.7 with the reflected transient pressure of 4.7 MPa (P_5) and the transient temperature of 3171 K (T_5) have been used and the values have been calculated by the standard R-H relations as in equations.

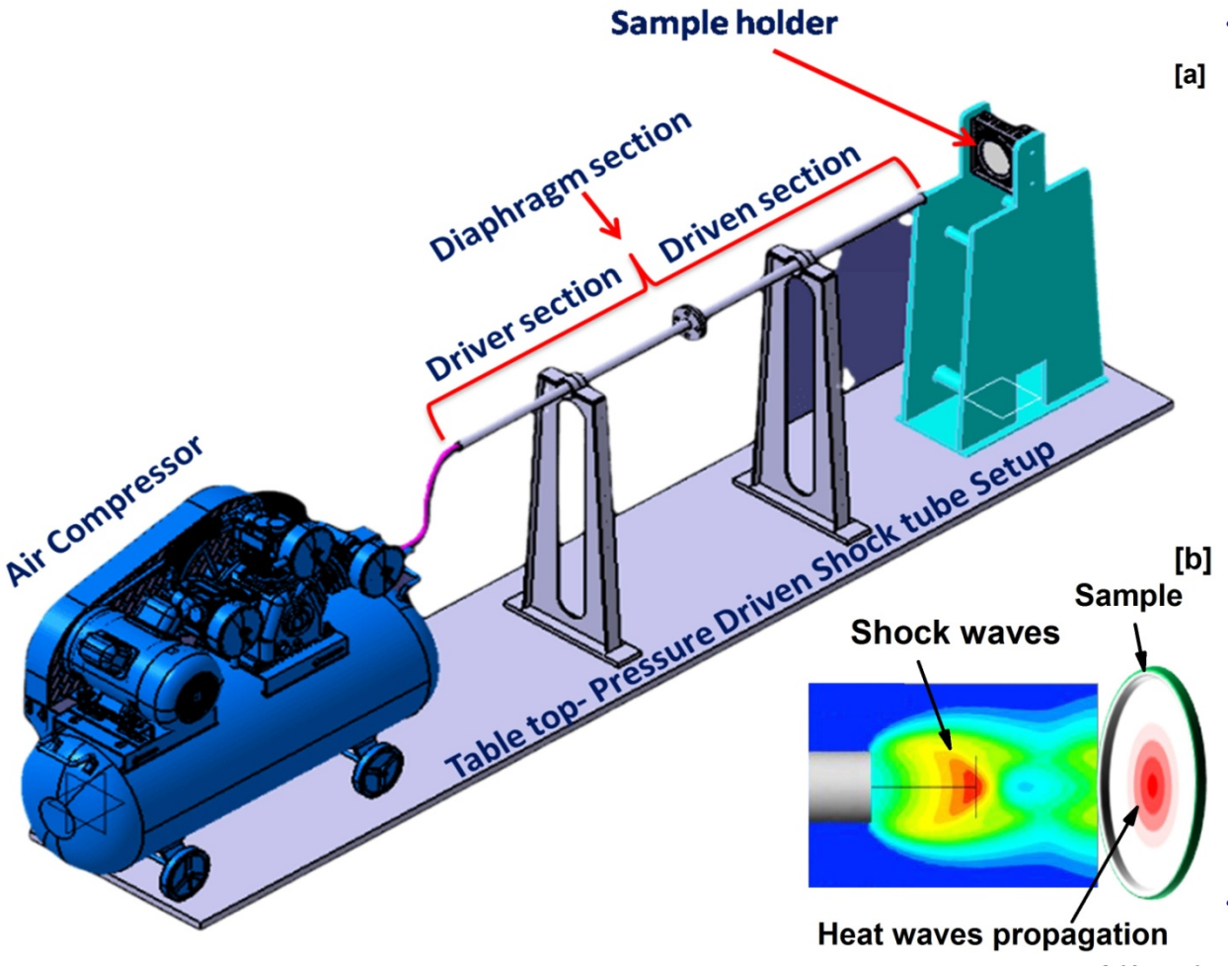

Fig. S1 (a) Schematic diagram of the table top pressure driven shock tube (b) shock propagation on the sample

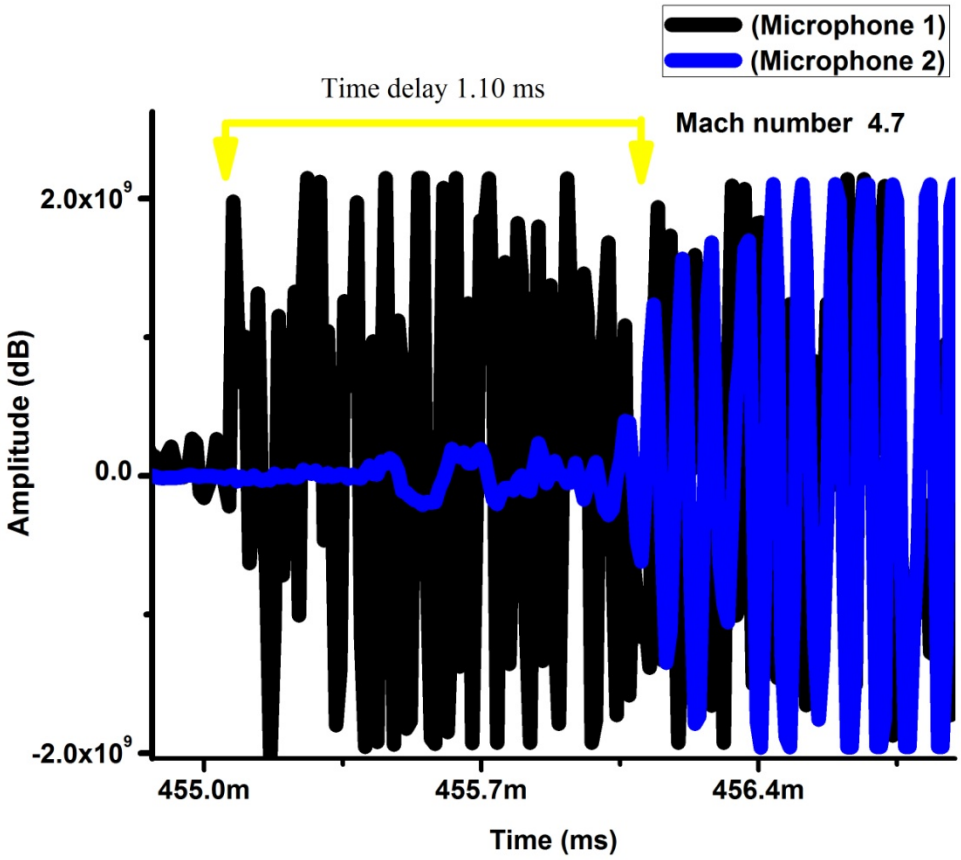

Fig. S2 Acoustic wave signals of the shock waves (recorded in the driven section)

R-H relations

$$P2 = P1\left[1 + \frac{2\gamma}{\gamma + 1}(M^2 - 1)\right] \ldots\ldots\ldots (1)$$

$$\frac{T_2}{T_1} = \frac{P_2}{P_1}\left[\frac{\left(\frac{\gamma + 1}{\gamma - 1}\right) + \frac{P_2}{P_1}}{1 + \left(\frac{\gamma + 1}{\gamma - 1}\right)\frac{P_2}{P_1}}\right] \ldots\ldots\ldots (2)$$

$$\frac{P_5}{P_2} = \frac{(3\gamma - 1)\frac{P_2}{P_1} - (\gamma - 1)}{(\gamma - 1)\frac{P_2}{P_1} + (\gamma + 1)} \quad \ldots\ldots\ldots (3)$$

$$\frac{T_5}{T_2} = \frac{P_5}{P_2}\left[\frac{\left(\frac{\gamma + 1}{\gamma - 1}\right) + \frac{P_5}{P_2}}{1 + \left(\frac{\gamma + 1}{\gamma - 1}\right)\frac{P_5}{P_2}}\right] \quad \ldots\ldots\ldots (4)$$

The initial fixed values are P_1 = 1 bar, γ =1.4 , T= 300 K, where M- Mach number, P_5 and T_5 are the reflected transient pressure and temperature at the end of the driven tube.

After the completion of the shock wave loading, the control and shock wave loaded samples have been analyzed by powder Raman, XPS, VSM, HR-TEM, XPS and electrical properties to understand the structural and functional properties of the shocked samples.

Analytical experiments

Raman Spectroscopy Experiment

We investigated the Raman spectra of the control and shocked *ta*-C using a Renishaw 2000 micro confocal Raman spectrometer coupled with a 532 nm argon ionic excitation source. Single-crystal silicon with a characteristic Raman peak at 520.0 cm^{-1} was utilized to calibrate the Raman spectroscopy system prior to measurement. Raman spectra of the control and shocked CdO NPs were gathered within the wavenumber range of 100−3500 cm^{-1} in the backscattering geometry with the spectral resolution of 1.0 cm^{-1} and the acquisition time was 120 s. The laser spot size was 50 μm^2 and the optical microscope's objective lens was used such that an X50 long working distance objective lens (WD = 10.6 mm) was utilized and the value of numerical

aperture was 0.5 while the Raman data was collected by Renishaw Wire 5.1 instrument control and the data acquisition software. We processed the obtained Raman spectra with a Lorentzian-type function in Origin 9.0 software to extract the Raman peak position and its corresponding FWHM.

X-ray Photoelectron Spectrometer Experiment

We investigated the carbon bonding patterns of the control and shocked *ta*-C by XPS analysis. For this study, PHI - VERSAPROBE III – X-ray Photoelectron spectrometer was utilized. The typical characteristics of the spectrometer are as follows: Monochromatic X-ray Beam area is 15 μm and Al Kα radiation (1486.6 eV) and the data was recorded over the input energy from 0-1350 eV. The control and 200 shocked CdO NPs were utilized as it is to record the XPS data. XPS spectra were deconvoluted with CasaXPS 2.3.12 software, using a non-linear least squares fitting routine after a Shirley-type background subtraction and the peaks were interpreted using a combination of Gaussian/Lorentzian functions. The adjusted parameters were FWHM, binding energy and peak area. To correct possible deviations caused by the electric charge of the samples, the C1s band at 284.6 eV was taken as the internal standard. Surface atomic percentages were calculated from the corresponding peak areas upon spectra deconvolution and using the sensitivity factors provided by the manufacturer.

HR-TEM Experiment

Microscopic structural characterizations for the control and shocked *ta*-C NPs were investigated using HRTEM, which was operated at the State Key Laboratory of Environmental Geochemistry, Institute of Geochemistry, Chinese Academy of Sciences, Guiyang, China. A small amount of sample was homogeneously distributed onto a carbon-film-coated copper grid

for the HRTEM observation, which was performed through a Tecnai G2 F20 S-TWIN TMP with an acceleration voltage of 200 kV. We precisely measured the interplanar spacing of the samples through the Image J software.

Vibrating-Sample Magnetometer (VSM)

A vibrating sample magnetometer (Lake Shore, Cryotronics, model -7407-S) was employed to monitor the magnetic properties of the control and shocked *ta*-C at room temperature.

Electrochemical measurement

An electrochemical workstation (SP-150, BioLogic Science Instruments, France) at ambient temperature has been used for the present experiment. The working electrode has been prepared by bending 80 wt% of active material (*ta*-C), 10 wt% polyvinylidene fluoride (PVDF) binder with 0.3 mL of N-methylproline (NMP) solution and 10 wt% conductive carbon black. The prepared uniform slurry has been painted onto pre-cleaned 1 × 1 cm^2 nickel foam and allowed to air dry for 24 hrs under ambient conditions. The above-mentioned method has been used to fabricate the activated charcoal for the negative electrode as well. A three-electrode cell has been used for the measurement (electrolyte–3 mol KOH solution, working electrode–the prepared material, counter electrode–Pt wire, reference–Ag/AgCl in the saturated KCl). Shock wave-loaded sample electrode has been also prepared by employing the above-mentioned procedure. The cyclic voltammetry (CV) tests have been carried out for the control and post-shocked samples for different applied potentials under the applied voltage ranging from 0 V to 0.5 V.

Results

Raman results

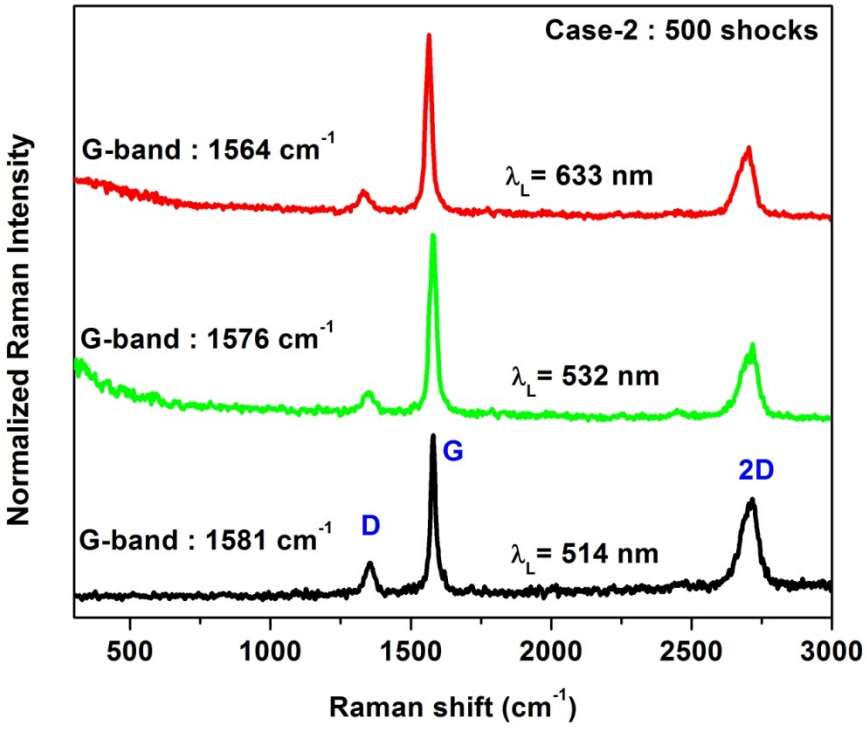

Fig. S3 laser wavelength dependent Raman spectra of case-2 500 shocked sample

XPS results

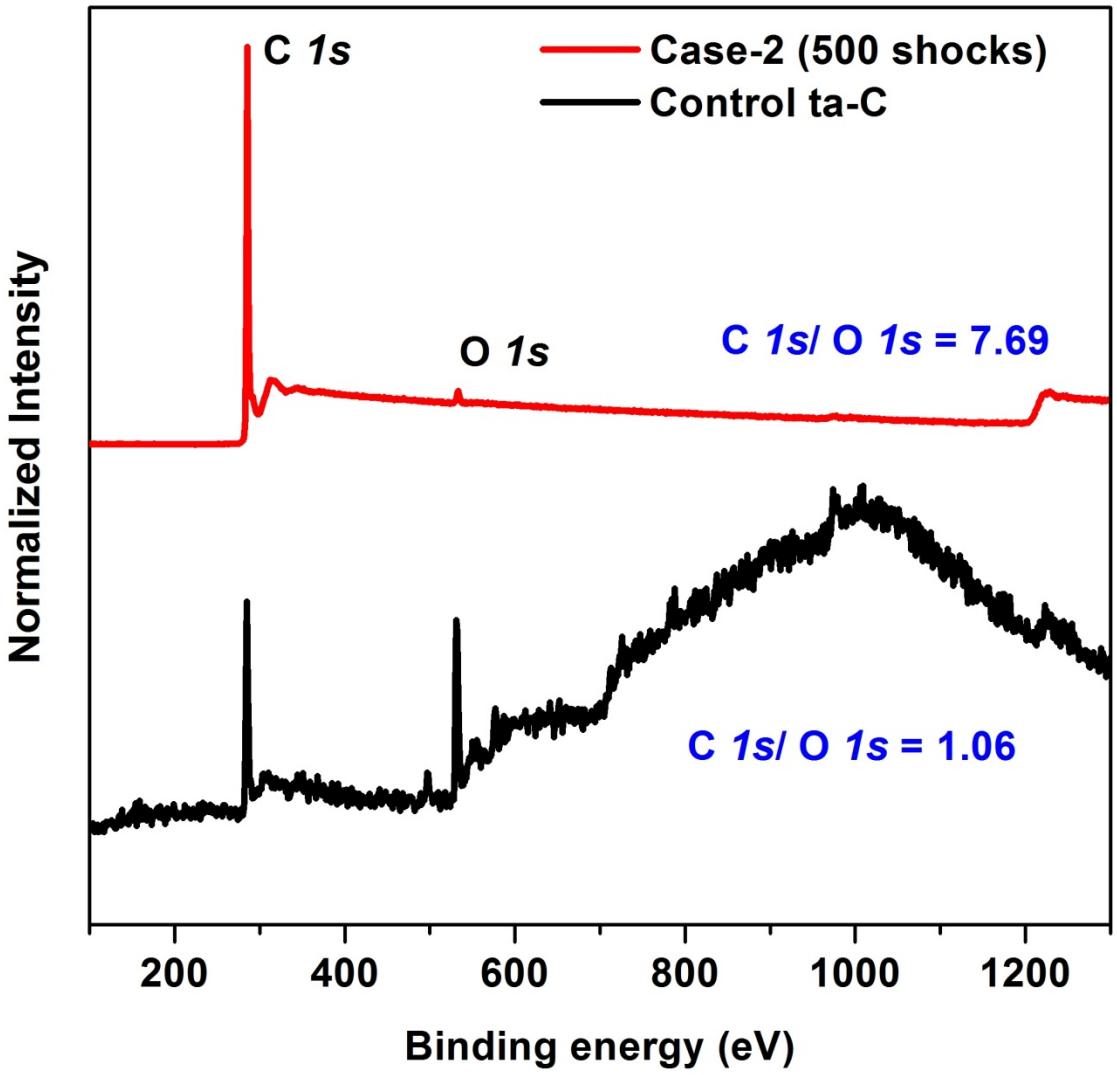
C 1s
Case-2 (500 shocks)
Control ta-C
O 1s
C 1s/ O 1s = 7.69
C 1s/ O 1s = 1.06
Normalized Intensity
200
400
600
800
1000
1200
Binding energy (eV)

Fig. S4 Survey spectra of control and case-2 500 shocked *ta*-C samples