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## **Electronic Supplementary Information (ESI)**

## Exploring few-layer graphene and graphene oxide as fillers to enhance the oxygen-atom corrosion resistance of composites

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## **Experimental**

12.5 g PVA (polyvinylalcohol, Beijing Yili Fine Chemicals Co., Ltd.,

 $M_w$ =77000±2200 g/mol) was dissolved in 250 mL water (~90 °C) and magnetically stirred for 8 h to form homogeneous solution with a PVA concentration of 50 mg/mL. Then 1.5 g crystal graphite flakes (Alfa Aesar,  $\leq$ 300 mesh) were added into the PVA solution and sonicated for 30 h in a sonic bath (Kunshan, 100 W, 40 KHz) with water cooling. The sonicated dispersions were further centrifuged at 2000rpm (×1024g) for 45 min. The top 90% supernatant was collected for further characterization and use. According to the Lamber-Beer law,  $A/l=\alpha C$ , where A is the absorbance measured at 660 nm, l=0.01m is the path length, and C is the concentration, the dispersed fewlayer graphene (FLG) concentration was measured (taking  $\alpha$ ~3620 mL/mg/m<sup>1</sup>) to be ~0.27 mg/mL. These dispersions were then mixed with PVA solution (50 mg/mL) to give a range of composite dispersions with FLG mass fractions from 0 to 0.5 wt%.

After the centrifugation at 2000rpm ( $\times$ 1024g), the sediment was expected to contain larger FLG flakes and unexfoliated graphite. So 100 mL PVA solution (50 mg/mL) was added to dissolve the sediment, followed by sonication for another 2 h. Then 500rpm ( $\times$ 64g) was used to centrifuge this dispersion and the supernatant was collected. By the method mentioned above, the FLG concentration of this dispersion was measured to be ~0.8 mg/mL. These dispersions were also then mixed with PVA solution (50 mg/mL) to give a range of composite dispersions with FLG mass fractions from 0 to 1.0 wt%. The above composite dispersions were further formed into films for atomic oxygen corrosion test. To form films, after sonication for 20 min, these dispersions were drop cast into high density polyethylene trays and left for 24 h in a vacuum over at 60 °C with a pressure of 0.9 bar. Composite films were peeled off and cut into  $15 \times 15$  mm<sup>2</sup> samples using a die cutter. These films are measured as ~80 µm by micrometer (micrometer screw gauge).

FLG oxide (GO) was prepared by mild sonication of graphite oxide for 30 min in water. Graphite oxide was prepared from natural graphite flake by a modified Hummers method which has been widely used.<sup>2</sup> The GO dispersion was diluted to 2 mg/mL. These GO dispersions were then mixed with PVA solution (50 mg/mL) to give a range of composite dispersions with GO mass fractions from 0 to 1.0 wt%. The composite dispersions were then sonicated for 1 h. Then the homogeneous mixtures were drop cast into high density polyethylene trays and left for 24 h in a vacuum over at 60 °C with a pressure of 0.9 bar. Composite films were peeled off and cut into  $15 \times 15 \text{ mm}^2$  samples using a die cutter.

Oxygen-atom exposure experiments were carried out in low earth orbit (LEO) environment ground-based atomic oxygen effect simulation facility in Beijing University of Aeronautics and Astronautics,<sup>3, 4</sup> under a condition of pressure of 0.15 Pa, filament discharge voltage of 120 V, and filament discharge current of 140 mA. The oxygen atom is generated by a plasma-type ground-based atomic oxygen effects simulation facility designed by ourselves (*J. Phys. D: Appl. Phys. 2001,34,2308; Acta Aeronautica Et Astronautica Sinica 2000,21,425*). This facility is with the filament

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discharge and bound of magnetic field. The temperature in the test is low, less than 50 °C. The mass loss of Kapton in the atomic oxygen exposure experiments was used as a criterion to calculate the atomic oxygen flux. The calculation formula is  $Ft=\Delta M/(\rho AE_y)$  in which *F* is the effective flow rate of atomic oxygen onto the sample surface and  $\Delta M$ ,  $\rho$ , *A*, *t*, *E<sub>y</sub>* are mass loss, density, surface area, exposure time, and erosion yield, respectively. *Ft* is the accumulative atomic oxygen flux exerting on the sample surface. For Kapton, *E<sub>y</sub>* is equal to ~3.0×10<sup>-24</sup>cm<sup>3</sup>/atom. In this study, the accumulative atomic oxygen flux was about  $4.7 \times 10^{20}$  atoms/cm<sup>2</sup>, which was approximately equivalent to the accumulative atomic oxygen flux of ~140 days at an orbit of 300 km in LEO.<sup>5</sup> For each mass fraction of PVA/FLG or PVA/GO composite films, at least three samples were tested and the mass loss averaged.

UV-Vis absorption spectroscopy was performed with a Purkinje General TU1901 (1cm cuvettes). Scanning electron microscopy (SEM) images were collected by a LEO 1530VP. Atomic force microscopy (AFM) images were captured with a Multimode 8 AFM (Bruker) in ScanAsyst Air mode. The Raman measurements were made by a Renishaw Rm2000 using a 514 nm laser. Thermogravimetric (TGA) analysis was performed on a TGA/SDTA851 (Mettler Toledo, USA) at 10°C/min in a nitrogen atmosphere. The mass of samples before and after oxygen-atom exposure was measured using a DT-100 balance with a sensitivity of 0.05 mg. X-ray photoelectron spectroscopy (XPS) was obtained by an ESCALAB-250 spectrometer.

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**Fig. S1** Additonal AFM images of (a), (b) large (size selected) FLG flakes after centrifugation of 500rpm (×64g) and (c), (d) small FLG flakes after centrifugation of 2000rpm (×1024g).



**Fig. S2** Representative stress-strain curves for the films of PVA, PVA/0.5 wt% FLG composite. The maximum tensile stress increases from 58 MPa in PVA film to 74 MPa in PVA/0.5 wt% FLG composite.



**Fig. S3** (a) Surface SEM image and (b) mass loss of PVA composite with a graphite loading of ~1 wt% after oxygen-atom corrosion.

## References

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