SUPPORTING INFORMATION

An aqueous rechargeable and high-capacity zinc ion battery using novel rGO-V$_2$O$_5$-SiO$_2$ hybrid nanocomposite as cathode material

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Materials

Graphite flakes were purchased from Graphene Supermarket, USA. Other chemicals like sodium metasilicate (Na_2SiO_3) hydrochloric acid (HC), sulfuric acid (H_2SO_4), potassium permanganate ($KMnO_4$), hydrogen peroxides (H_2O_2), ethanol, acetone, sodium hydroxide (NaOH), vanadium (V) oxychloride, 1-Butyl-3 methyl-imidazolium-trifluoro-methane sulfonate, and poly(vinylidene-fluoride-co-hexafluoropropylene) (PVDF) were purchased from Sigma Aldrich. Acetylene black, Zn sheet (0.02 mm) were procured from Mumbai, India. Polypropylene separator film (20-micron thickness) was obtained from Aero Incorporation, Chennai.

rGO-V_2O_5-SiO_2 nanocomposite synthesis

SiO_2 nanoparticles were obtained from Na_2SiO_3 (854 mg), which was dissolved into 5 mL deionized (DI) water and stirred for 30 minutes. Then, acid-base titration of the above solution was performed with HCl (2 M, 8 mL) by slowly adding it into the above solution under constant stirring until a white colloid formed. Next, the mixture was filtered and washed continuously with deionized water by centrifugation (till the pH of the solution reached 7.0). In another set, 50 mg of graphene oxide powder was added to a mixture of ethanol and water (1:1 ratio by vol.) and ultrasonicated for 30 min. Next, V_2O_5 (182mg) and the prepared SiO_2 (420 mg) were added to the graphene oxide solution and stirred for 2 hours. The mixture was later transferred into a Teflon-lined autoclave and kept at 200°C for 30 hours. After 30 hours, the solution was filtered out through centrifugation, and then the obtained solution was dried at 55°C to be used as the cathode material.

Fabrication of Electrode and Pouch Cell

The cathode was fabricated by using rGO-V_2O_5-SiO_2 nanocomposite. For this purpose, a slurry was prepared by mixing the nanocomposite (80%), acetylene black (10%), and PVDF solution (10%) by weight. The slurry was then coated on a carbon-coated aluminum foil (thickness 16 μm, coating 1 μm) of 16 cm^2 area with an applicator with a thickness of ~100 μm. The mass loading of the active material (rGO-V_2O_5-SiO_2 + acetylene black + PVDF) was 0.45 mg cm^{-2}. The cathode was then kept in the hot air oven at 80°C for 12 hours. After drying, the deposited film was calendaring to attain uniform coating. Zinc foil (thickness, 0.1 mm) was used as the anode, and the polypropylene membrane acted as the separator. The electrolyte solution was added to the separator-electrode assembly and kept in a laminated aluminum pouch.

The electrochemical performance was tested using pouch-type cells with an active single electrode area of 16 cm^2. The aqueous Zn-rGO-V_2O_5-SiO_2 battery was fabricated in ambient conditions. The

aqueous electrolyte (0.5 M) was prepared by mixing 0.545 gm of Zn (CF_3SO_3) powder in 3 ml of organic solvent 1-Butyl-3-methylimidazolium trifluoro methane sulfonate and stirring for 12 hours. The single pouch cell was tested between 0.1 and 1.5 V using the battery testing instrument (30 V, 10A, Neware, China). The specific capacity was calculated by considering higher active mass loading, i.e., ~7.3 mg/cm^2.

Characterizations

The crystallinity information of the samples was characterized using powder X-ray diffraction (PXRD) (Panalytical Empyrean, Cu Ka, λ=1.5406 Å). The morphology of the samples was studied using field emission scanning electron microscopy (FESEM, JEOL, JSM 7900F). The energy dispersive spectroscopy (EDS) mapping was analyzed using an EDS detector (Oxford instrumentation) equipped with FESEM. Raman spectra has been recorded on alpha300 R – Raman Imaging Microscope. Electrochemical measurements, including cyclic voltammetry (CV), The cyclic voltammograms (CV) were performed at ambient temperature (30 °C) using an electrochemical workstation (Kanopy tech, Kanpur, India) with the conventional three-electrode system. The developed material was investigated with three-electrode (3E) cells and two-electrode (2E) asymmetric cell configurations by 8-channel battery testing instrument (NEWARE, China). The pouch was sealed using a vacuum sealing machine (Winner electronics, India).
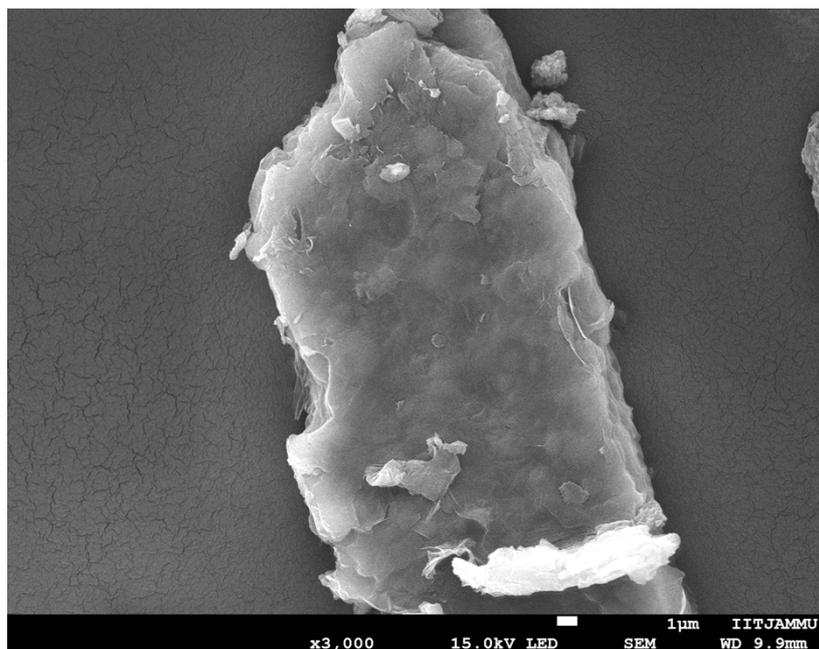
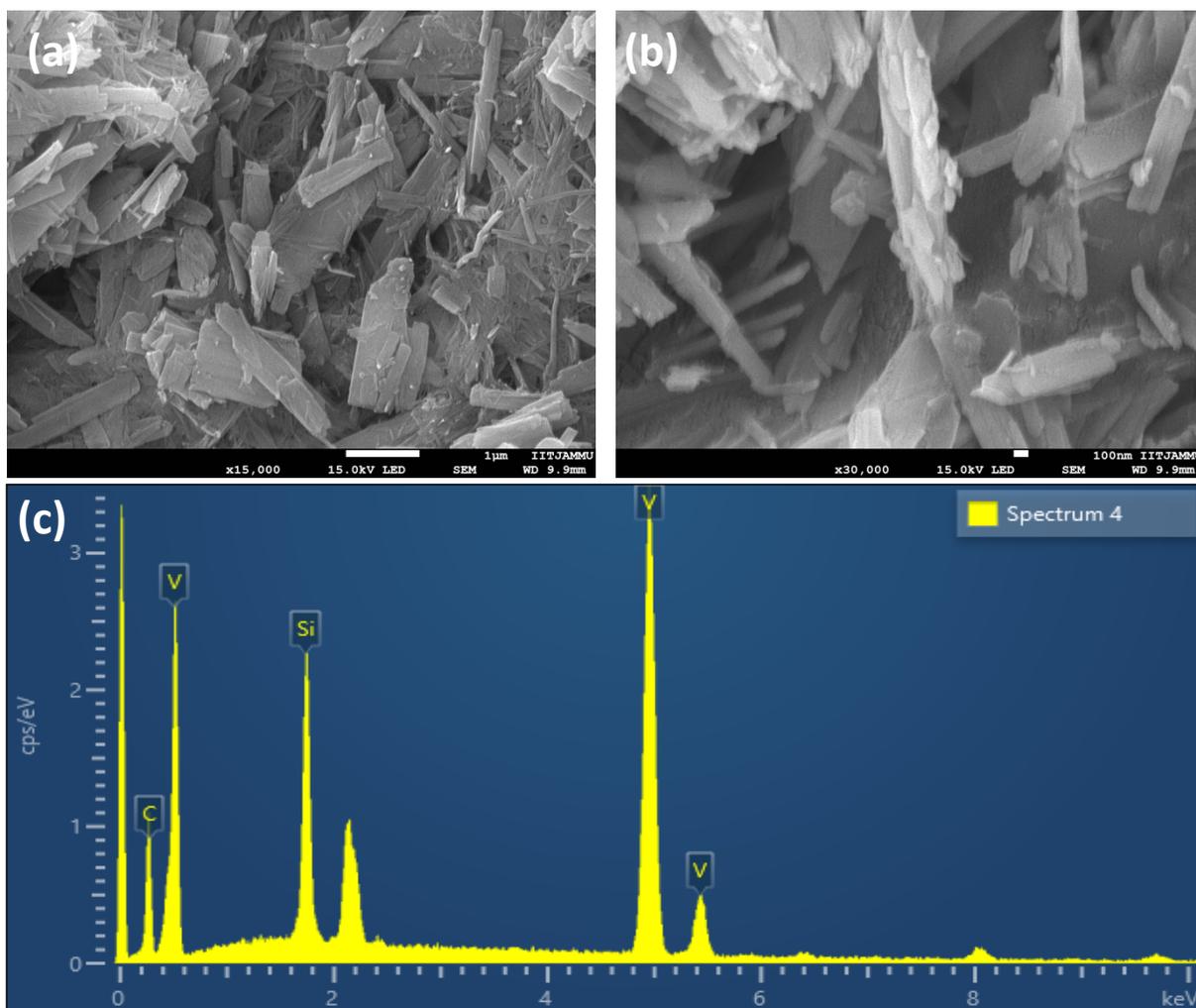**Fig. S1** FESEM images of GO sheet at 1 μm magnification showing a crumpled paper like morphology with wrinkled edges.

Fig. S2 (a) V$_2$O$_5$ Nanostructure with perfect plated sheet a different magnification level. (b) At higher magnification level of 30000x, a nano spherical morphology can be observed at the surface of the V$_2$O$_5$, (c) Elemental analysis of the pristine composite with quantitative analysis of the synthesized nanocomposite.
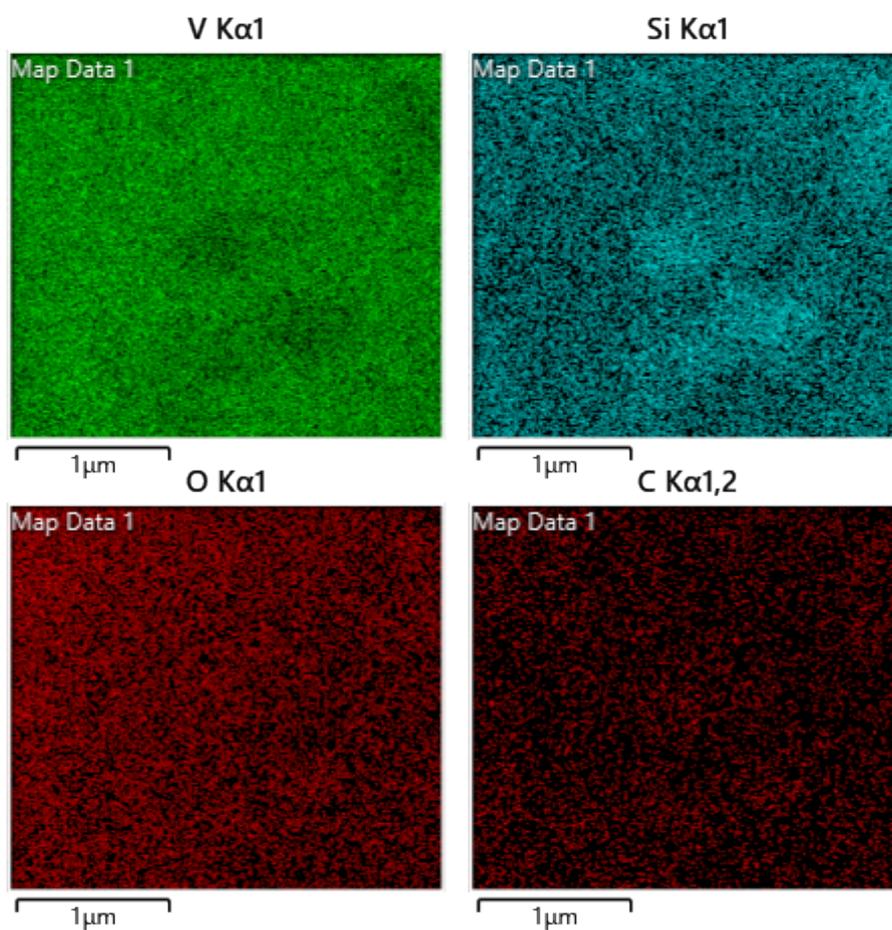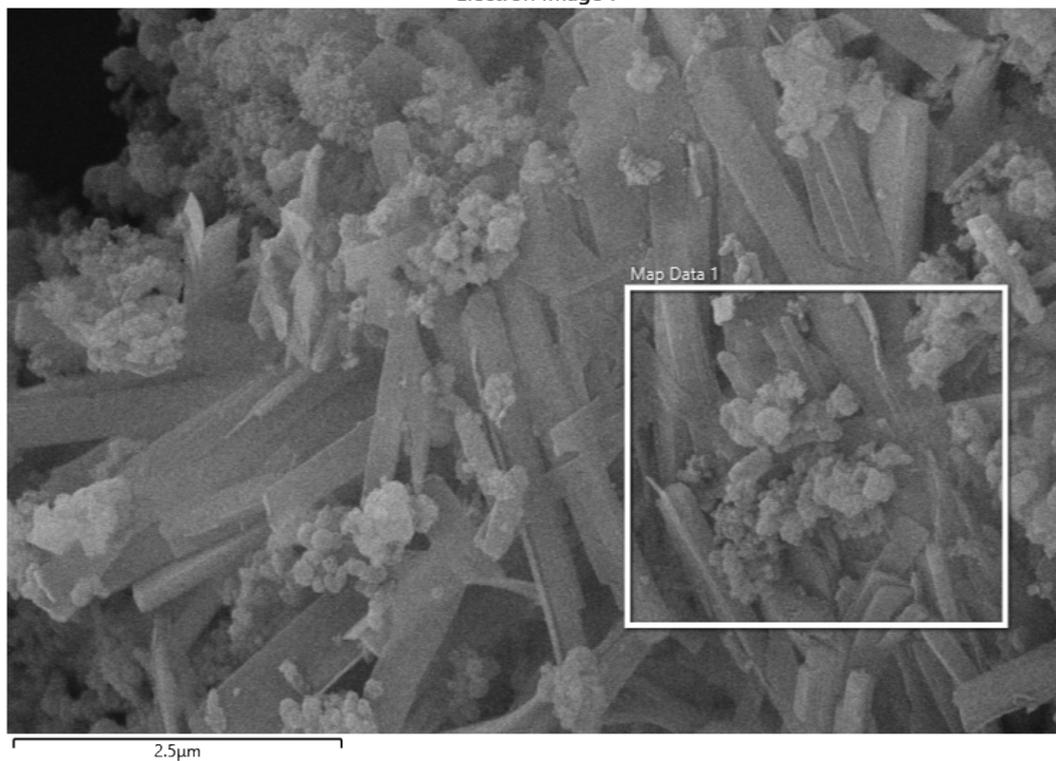
Electron Image 7

2.5μm

V Kα1

Map Data 1

1μm

Si Kα1

Map Data 1

1μm

O Kα1

Map Data 1

1μm

C Kα1,2

Map Data 1

1μm

Fig. S3 EDS mapping of nanocomposite (rGO-V_2O_5-SiO_2) for a large area and a clean even dispersion of the participating materials can be observed in the images.
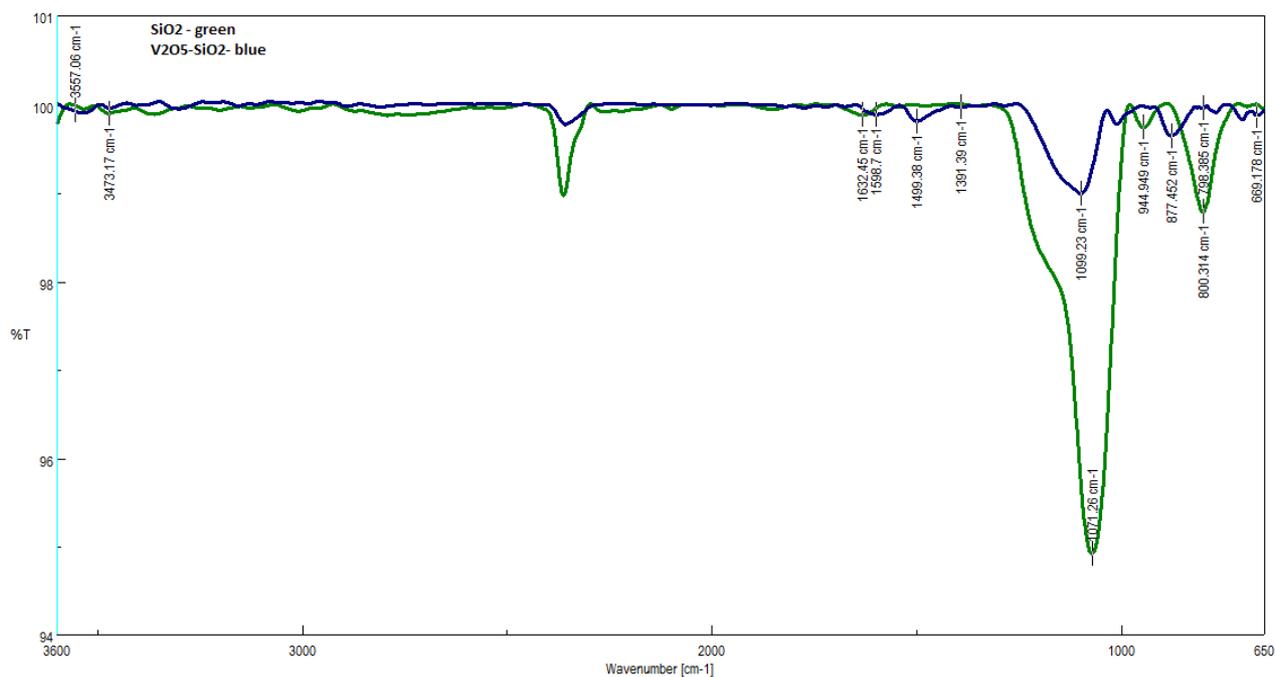
Fig. S4 FTIR spectra of the cathode material, asymmetric might be attributed to the V=O stretching band at about 1010 cm^{-1} (blue spectra).

Fig. S5 assemble the 10 pouch cells of 1.2V and 890 mAh in series connection. All specific sizes can be clearly seen through these optical images, which directly affect the storage and charge exchange mechanism.
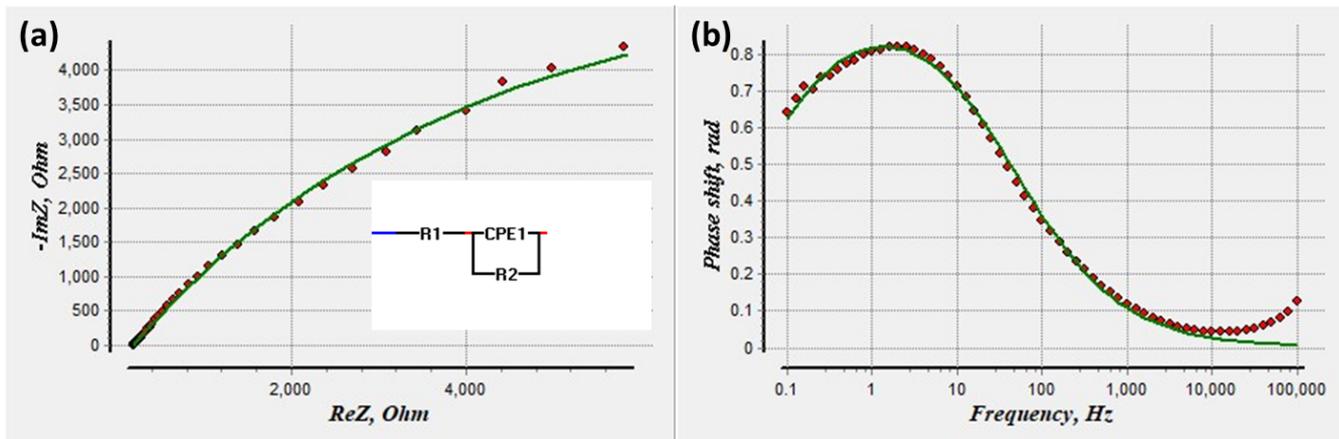
Fig. S6 (a, b) EIS test recorded at 1.4 V after charged state in the voltage window of 0.2 to 1.4 V. (a) Nyquist plot and (b) the corresponding Bode plot

Table S1 Resultant parameters of EIS analysis

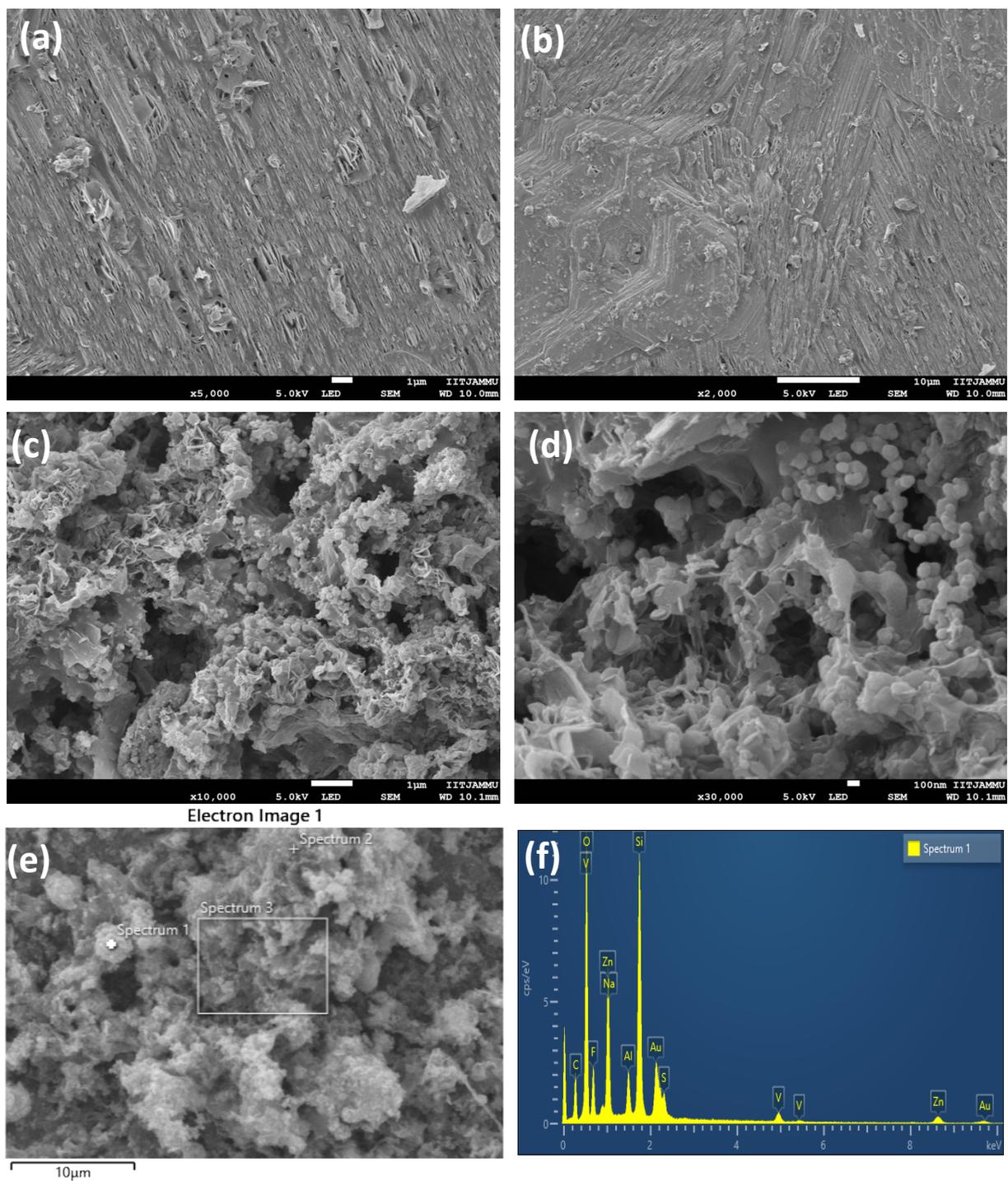| Parameters	Values of the parameters	Errors (%)
R1	205.83	1.1349
R2	17301	7.5657
P1	0.0001369	1.4475
n1	0.63214	0.6016

Fig. S7 Material characterizations of the Zn electrode and rGO-V$_2$O$_5$-SiO$_2$ coated on Aluminum foil (carbon coated) after 120 cycles (a) Zn sheet surface area with the scale of 1 micron (b) Surface area of Zn sheet over a large area (c) Surface morphology of the composite present with the distinct phases (d) At nanoscale visualizing morphology of rGO sheets, V$_2$O$_5$ nanoplatelets along with the SiO$_2$ nanoparticles. (e) EDS mapping area of rGO-V$_2$O$_5$-SiO$_2$ (f) Mapping data with all available elements in the composite.

Table S2 Comparison of different parameters of Zn-ion battery with reported literature

Cathode	Electrolyte	Operating Voltage V	Current Density	Capacity	Cycling Performance	Ref
V_2O_5-EGO	3M Aqueous $Zn(CF_3SO_3)_2$	0.4–1.6V	0.2 A/g	462 mAh/g	3000 cycles performed for different specific capacity	[1] (cc)
Zn- V_2O_5	3M Aqueous $Zn(CF_3SO_3)_2$	0.4–1.6V	0.2 A/g	470 mAh/g	91.1% retention capacity after 4000 cycles	[2] (cc)
$V_2O_5 \cdot nH_2O$	3M Aqueous $Zn(CF_3SO_3)_2$	0.2–1.6V	60 mA/g	381 mAh/g	71% retention capacity after 900 cycles	[3] (cc)
V_2O_5/rGO	3M $ZnSO_4$	0.4–1.4V	0.1 A/g	135 mAh/g	200 cycles	[4] (cc)
V_2O_5	3M $ZnSO_4$	0.4–1.4V	0.3 A/g	372 mAh/g	400 cycles	[5] (cc)
$V_3O_7 \cdot H_2O$	0.25 M Aqueous $Zn(CF_3SO_3)_2$	0.4–1.1V	375 mA/g	400 mAh/g	99.5% retention capacity after 200 cycles	[6] (cc)
VO_2	3M Aqueous $Zn(CF_3SO_3)_2$	0.3- 1.5V	100 mA/g	357 mAh/g	100 cycles with different C rates	[7](Not mentioned)
rGO- V_2O_5-SiO_2	1M Aqueous $Zn(CF_3SO_3)_2$	0.4- 0.8V	200 mA/g	640mAh/g	98% retention capacity after 100 cycles	This work (PC)

CC – Coin cell

PC- Pouch cell

Note 1: In this work a significant increment has been recorded in the field of Zn ion batteries, we obtained up to 98% Cyclic retention of 100 cycles for the segment of pouch cells, which is very difficult to achieve t a large scale. And also delivered a good specific capacity in numbers in comparison existing coin cells.

References:

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