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Phosphotungstate Nanoclusters Enhance the Capacitance and Energy Density of Activated Carbon in Organic Electrolyte Supercapacitors

Jun-Jie Zhu; Raul Benages-Vilau; Pedro Gomez-Romero
Catalan Institute of Nanoscience and Nanotechnology (ICN2), CSIC and BIST, Campus UAB, Bellaterra, 08193 Barcelona, Spain.
pedro.gomez@icn2.cat

ABSTRACT

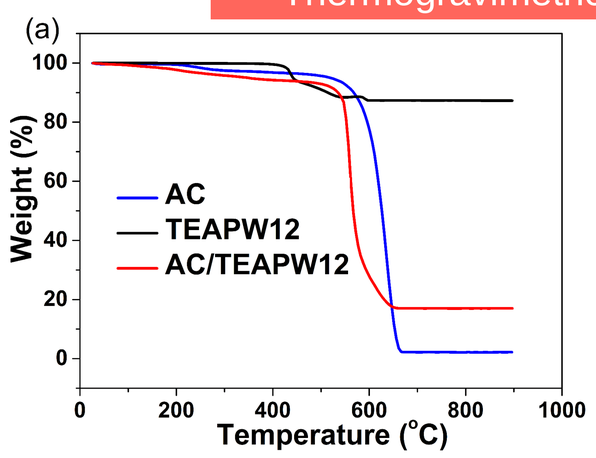
Phosphotungstic acid is a solid acid with reversible electroactivity and as such, we have previously shown how it can work as a faradaic additive to activated carbon (AC) in acidic aqueous electrolytes[1]. Yet, their use in organic media allows not only for added capacity but also higher voltage. We will present our recent work showing how the tetraethylammonium derivative of phosphotungstate $[PW_{12}O_{40}]^{3-}$ (TEAPW12) can be homogeneously distributed throughout the pores of activated carbon (AC) in organic solvents such as N,N'-dimethylformamide (DMF) and demonstrate the use of this hybrid electrode material in an organic electrolyte (1 M TEABF4 in acetonitrile) supercapacitor.

EXPERIMENTAL PROCEDURE

- TEAPW12 was synthesized through metathesis reaction. Typically, 200 mL of a 20 mM aqueous solution of phosphotungstic acid mixed with 200 mL of a 70 mM aqueous solution of tetraethylammonium chloride led to a white precipitate of TEAPW12 ($[(C_2H_5)_4N)]_3PW_{12}O_{40}$)
- The hybrid material (AC/TEAPW12) was prepared by mixing activated carbon and TEAPW12 in DMF in bath sonication.
- For comparison, hybrid material (activated carbon with phosphotungstic acid, AC/HPW12) was prepared following the previous literature methodology. [1]

RESULTS

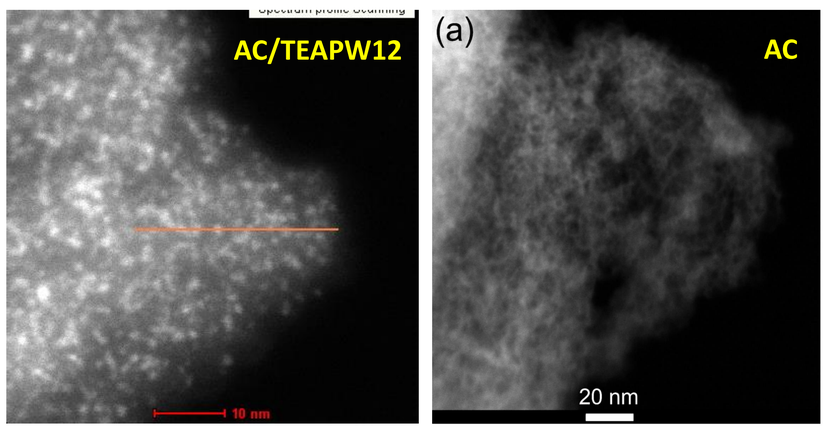
Thermogravimetric analysis

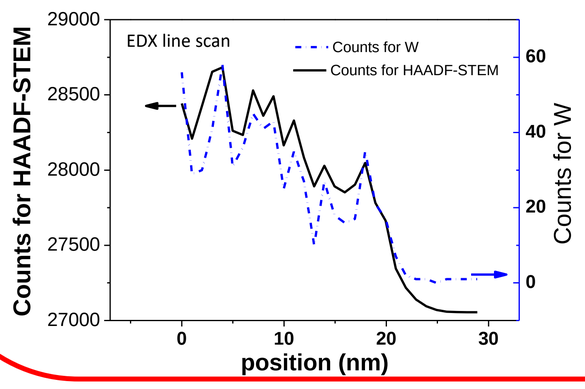


TGA confirms:

- TEAPW12 accounts for 17.5 wt% in the hybrid materials.
- Thermal stability of AC/TEAPW12 (ca. 550ºC) intermediate between its components

HADDF-STEM





- EDX line scan proves bright dots correspond to PW12 clusters.
- The image of AC/TEAPW12 show PW12 clusters are homogeneously dispersed on AC in nanoscale

Cyclic voltammograms (CV)

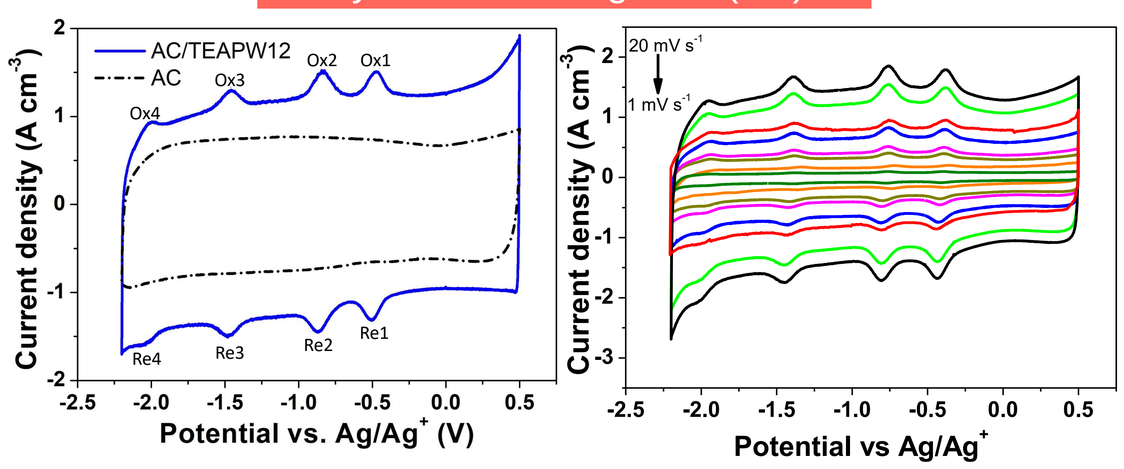
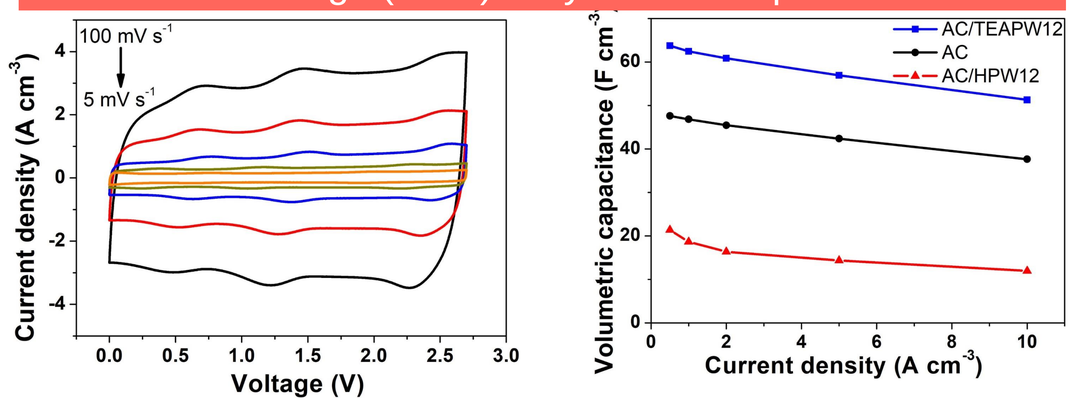


Table 1 Values of *b* (eqs. $i = av^b$) and regression coefficients (R^2) derived from fitting current of reduction peaks (Re) and oxidation peaks (Ox) at various scan rates.

	Re1	Ox1	Re2	Ox2	Re3	Ox3	Re4	Ox4
b	0.99	1.0	1.0	1.0	1.0	0.98	1.0	0.95
R^2	0.9996	0.9787	0.999	0.9978	0.9994	0.9996	0.9924	0.9819

CVs at 20 mV s^{-1} show that AC/TEAPW12 can deliver high volumetric capacitance.
CVs of AC/TEPW12 at various scan rates were carried out to analyze the contribution of surface-controlled process and diffusion-controlled process to energy storage. The linear proportionality of i vs scan rate (v) (b=1 in Table 1), even at potentials associated to redox peaks, imply non-diffusion-limited process(b=1).

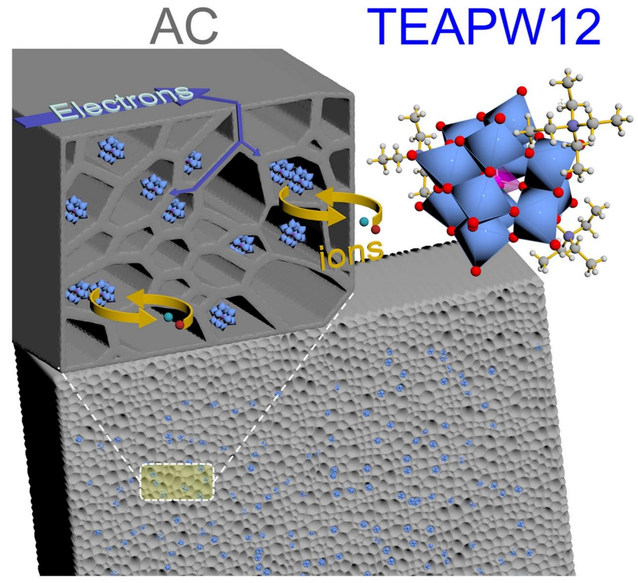
Cyclic polarization (CP)and Galvanostatic Charge-Discharge (GCD) in symmetric capacitor



CP curves of AC/TEAPW12 symmetric capacitor (left) show three pairs of redox waves.
Capacitance of AC/TEAPW12 is 36% larger than that of AC at the whole range.

CONCLUSIONS

- TEAPW12 clusters anchored on AC homogeneous in nanaoscale
- AC/TEAPW12 shows an increase (36%) in volumetric capacitance with respect to pristine AC;
- This increase is predominantly from non-diffusion-limited processes thanks to the utterly dispersed nature of POMs.



CONTACT PERSON

Pedro Gomez-Romero
pedro.gomez@icn2.cat

REFERENCES

[1] J. Suarez-Guevara, V. Ruiz and P. Gomez-Romero, Journal of Materials Chemistry A, 4 (2014) 1014-1021
[2] J.-J. Zhu, R. Benages-Vilau, P. Gomez-Romero, Electrochimica Acta, 362 (2020) 137007.

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