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Communication

"Soft" graphene oxide-organopolysulfide nanocomposites for superior pseudocapacitive lithium storage

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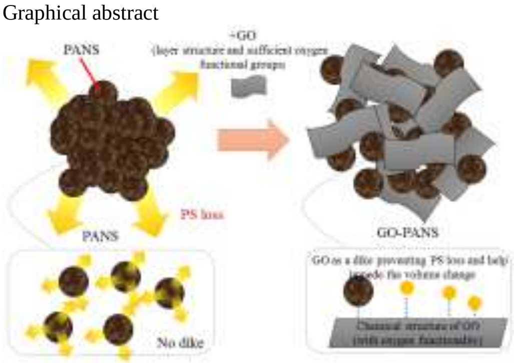
Graphical abstract



We report a "soft" graphene oxide-organopolysulfide nanocomposite with improved *pseudo*capacitive performance for high-potential (1-2.8 V *vs.* Li^0/Li^+), high-capacity (278 mA h g^{-1}) and stable (500 cycles) lithium storage.

ABSTRACT
We report a "soft" graphene oxide-polymeric organosulfide nanocomposite with improved pseudocapacitive performance for high-potential (1-2.8 V *vs.* Li^0/Li^+), high-capacity (278 mA h g^{-1}) and stable (500 cycles) lithium storage.

Keywords:
Lithium ion capacitor
Graphene oxide
Organic cathode
Polysulfide
*Pseudo*capacitor

Unlike the versatile electrochemical uses of grapheme [1-4], the present electrochemical applications of graphene oxide (GO), a typical graphene derivative abundant with polar surface functionalities, are devoted to catalysis, luminescence and sensing [5]. Exploring the emerging potential of GO in electrochemical energy storage is thus of appreciable significance. Our group recently demonstrated the exceptional reversible lithium ion storage by both epoxide and carbonyl groups on GO cathodes in the potential range of 1.5~4.5 V *vs.* Li^0/Li^+ [6]. Graphene oxide stores lithium ions *via* a surface process which involves the opening and closing of C-O-C/C=O bonds upon lithium uptake and release. Fast reaction kinetics is permitted for lithium storage in this case compared to the slow bulk intercalation in between sp^2 carbon layers at low potentials (< 0.5 V). Another feature of this surface lithium ion storage is that as the discharge/charge capacity increases, the potential decreases/increases monotonously suggesting a pseudocapacitive lithiation of GO, which is distinguished from the plateau potential behaviour for lithium intercalation/de-intercalation.

Despite the high lithiation activity, the GO nanosheets are prone to restack; and aggregation often reduces the available surfaces for lithium storage which results in capacity loss. Similar problems were associated with graphene electrodes; and the nanopillaring concept was developed to solve them [7]. By mixing graphene with nanoparticles, the graphene nanosheets are physically separated. It is therefore feasible to adopt a similar concept to design GO-nanoparticle composites to cope with the aggregation issue. To reach this target, it is important to preserve the *pseudo*capacitive property for the composites. Therefore, the practical challenge is to find a nanoparticle material that 1) stores lithium ions at the medium-high potential ranges (1~4.5 V *vs.* Li^0/Li^+), and 2) exhibits *pseudo*capacitive behavior for lithiation and delithiation. A nanosized particulate organosulfide polymer that is from the vulcanization

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