

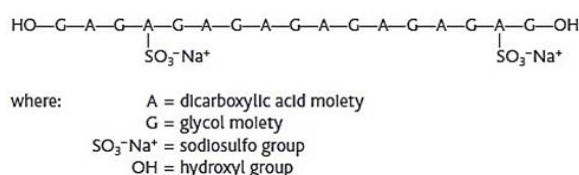
## A Green Fabrication Process for Lithium Ion Battery Using An Efficient, Low Cost and Environmentally Benign Polymer Binder

Min Ling<sup>a,b</sup>, Gao Liu<sup>b\*</sup>, Shanqing Zhang<sup>a\*</sup>

<sup>a</sup> Centre for Clean Environment and Energy, Environmental Futures Centre, and Griffith School of Environment, Gold Coast Campus, Griffith University QLD 4222, Australia.

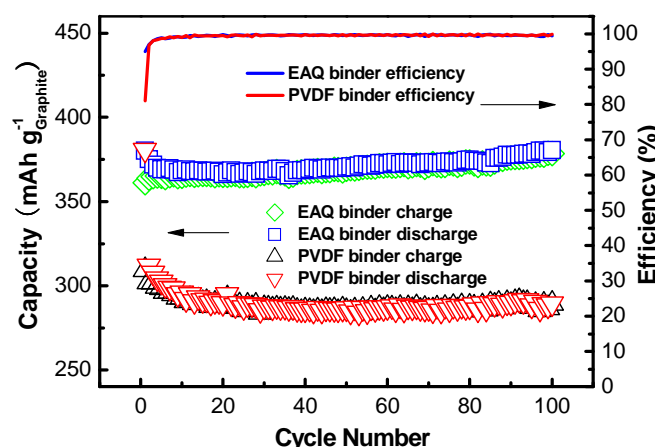
<sup>b</sup> Environmental Energy Technologies Division and Advanced Light Source, Lawrence Berkeley National Laboratory, Berkeley, California 94720, USA.

Polyvinylidene fluoride (PVDF), carboxymethyl cellulose (CMC), styrene-butadiene copolymer (SBR) binders have been the dominant binders of state-of-the-art electrode manufacturing in Li-ion batteries (LIBs) industry. However, PVDF requires the use of large amount of organic compounds organic solvents, typically N-methyl-2-pyrrolidinone (NMP), that are often volatile and toxic. In fact, collection, purification and recycling of these solvents results in substantial manufacturing cost to battery production process [1]. Furthermore, in order to address the ever-increasing concerns of human and environment, we need to convert the tradition LIBs production process from organic solvent and binder based process into a green and water-based production technologies. To achieve this goal, water soluble synthetic and low cost Eastman AQ<sup>TM</sup> 55S polymer (EAQ) is proposed here for the first time as an alternative to the conventional PVDF, CMC and SBR binders.



**Fig. 1** The molecular structure of the EAQ polymer.

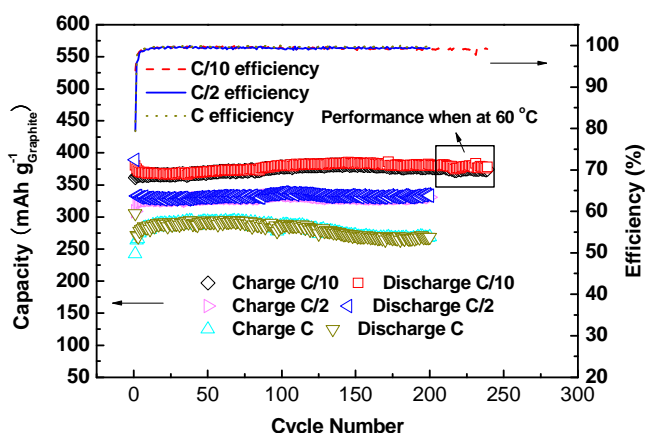
The EAQ polymer is cheap, water soluble, and nontoxic. Originally, it was produced by Eastman Chemical Company as an ingredient for cosmetics and personal care applications. The AQ polymer has a high viscosity in water with the inherent viscosity of 0.29-0.37 dL/g. Also, this polymer has abundant active hydroxyl groups on its molecular backbone (See Fig. 1) that can make it a promising material to interact with a large variety of surfaces basing on these advantages [2], we exploited the use of this polymer as an LIB binder using graphite as electrode active materials in an aqueous suspension.



**Fig. 2** Comparison of the galvanostatic charge discharge profiles of the oxidized graphite using PVDF and EAQ polymer (graphite/binder=9) as the binder between a cycling voltage of 3 V and 0.01 V.

Firstly, in order to strengthen the adhesion of the binder to

the graphite surface, the graphite was modified by being oxidized in concentrated HNO<sub>3</sub> at 60 °C for 1 h to increase the amount of the active groups on its surface that will result in the enhanced chemical bond between the graphite and the EAQ. To compare the performance of EAQ with that of PVDF, the oxidized graphite with carboxyl group on its surface was used as the anode material as illustrated in Fig. 2. As expected, the performance of EAQ polymer binder is better when using the oxidized carboxyl enrichment graphite, the specific capacity is stable at 370 mAh/g and closed to the theoretical capacity of graphite compare to 260 mAh/g when use PVDF binder. The enhancement of the reversible capacity and stability can be attributed mainly to the chemical bonds between EAQ polymer binder and oxidized graphite and the structural imperfections elimination through the oxidation treatment.



**Fig. 3** Galvanostatic charge discharge profiles of the oxidized graphite oxidized using EAQ polymer (oxidized graphite/binder=9:1, weight ratio) as a composite binder between a cycling voltage of 3 V and 0.01 V.

To investigate the higher current discharge-charge performance, as illustrated in Fig. 3, the reversible capacity of modified graphite combined with EAQ polymer can be stable at 370 mAh/g during more than 200 cycles when at C/10, the coulombic efficiency also keep stable up to 99%. When the discharge-charge current increased to C/2 the capacity is about 330 mAh/g and a furthermore 280 mAh/g when at 1 C. According to Fig 2, we can also find that the higher temperature performance of the EAQ polymer binder/graphite electrode is quite well since there is no decrease after the temperature of the testing environment increased to 60 °C. [3,4]

In conclusion, better performance of the EAQ polymer binder was obtained in comparison with PVDF binder, we demonstrate that the formation of the chemical bonds between the carboxyl group on the oxidized graphite surface and the hydroxyl group on the EAQ polymer surface is the key factor. The water soluble Eastman EAQ polymer binder have the potential to convert the tradition LIBs production process from organic solvent and binder based process into a green and water-based production technologies.

### Reference

1. J.B. Goodenough, Y. Kim, Chemistry of Materials, 22 (2010) 587-603.
2. I. Kovalenko, B. Zdyrko, A. Magasinski, B. Hertzberg, Z. Milicev, R. Burtovyy, I. Luzinov, G. Yushin, Science, 334 (2011) 75-79.
3. G. Liu, S. Xun, N. Vukmirovic, X. Song, P. Olalde-Velasco, H. Zheng, V.S. Battaglia, L. Wang, W. Yang, Adv Mater, 23 (2011) 4679-4683.
4. G. Liu, H. Zheng, X. Song, V.S. Battaglia, Journal of The Electrochemical Society, 159 (2012) A214.