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The most common belief still claims the quinone contributes to the redox activity of the multi-walled carbon nanotubes (MWCNTs). However, from our experiments, the redox reaction from MWCNTs do not shown the same sharp as the quinone/carbonyl redox. Since the redox activity plays an important role during electrochemical applications, a clear study on the relationship of functional groups and redox activity is critical for various research fields. Here, the redox reaction dependence on the functional groups has been studied by introducing/removing functional groups using various oxidation treatment and argon annealing process. A weak oxidant treatment was applied to specifically introduce the functional groups on the original defective structures of MWCNTs at slow temperature, while maintaining the other carbon structures intact. In comparison, more defects on sidewall were first introduced by the chemical drilling process, then more functional groups formed on those sidewall defects after further acid treatment. Furthermore, to investigate the redox reaction of the functional groups, two oxidation processes were designed to introduce functional groups on MWCNTs with and without additional chemical drilled defective structure, respectively. The influence on redox of various functional groups ratios has also been studied. The impact of different OFGs on redox reaction can be revealed by thermal annealing with different temperatures. By gradually removing the functional groups on MWCNTs with different thermal annealing temperatures, carboxyl/anhydride and phenol groups were recognized to play essential roles in the redox reaction. These results provide new insights for the study of the MWCNTs' electrochemical properties towards supercapacitors and batteries.

Moreover, several experiments suggest that removing the OFGs would lead to the carbon atoms reconstruction on the surface of MWCNTs, which would form topological defective structures. In order to compare the difference electrochemical influences from topological defective structures with our original hold defective structures. By carefully choosing the combination and sequence of oxidants, a mild oxidizing atmosphere is established for the convenience of carbonyl construction. Results indicate that topological defective structures would bring benefits to ORR activity of MWCNTs compared with our traditional chemical drilling hole defect making procedure. From the results, the original hole defective structures would not improve the ORR activity directly, while the topological defects form by removing OFGs will benefit the ORR activity. The results also suggest that removing carboxyl, carbolic anhydride, phenol and also quinone/carbonyl will improve the ORR activity.

In order to improve the hole defective making procedure, different chemical drilling processes have been investigated in order to increase the ORR activity of MWCNTs directly. The results suggested that the defective structures can be adjusted by changing the parameter in oxidation process.

In this study, beside the electrochemical measurements, further characterization of MWCNTs has been conducted by using FTIR, Raman, TPD, TG/DTA, BET, CV/LSV and XPS to study the roles of the oxygen containing functional groups to investigate their effects on the reactions. In order to study the property of MWCNTs in the real cell application, a serious of and PEMFC power generation testing has also been studied. The MEA fabrication procedures have been well studied to form desirable layer structures. Moreover, the MWCNTs have been proved could work as cathode catalyst in PEMFC. MWCNTs also have the ability that could help to reduce the Pt/C content while at the same time increase the cell output and stability anti-extreme situation.