Biomass Derived High Porous Carbon via CO_2 Activation for Supercapacitor Electrodes

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The choice of the appropriate activation method, reagents, temperature and other parameters is a determining factor for obtaining high-quality, environmentally friendly and cost-effective porous carbons for use as an active electrode mass in energy storage systems. The chemical activation method using potassium hydroxide is the most effective and common method for producing activated carbon (AC) from biomass and is widely described in the literature. However, its use poses certain dangers to the environment and has technological drawbacks. KOH is a strong alkali and can cause serious irritation of the skin, eyes, respiratory tract, and other mucous parts of the body. Another drawback is the impact on the environment both during production and during disposal after use.

Alternative methods, such as physical activation using carbon dioxide (CO_2), can offer a safer, cheaper, faster, and more economical approach to the production AC from various biomass materials. Along with the advantages of the method, the study showed that CO_2 activation has some disadvantages. One of the main drawbacks is the lower high specific surface area (SSA) with a narrower range of pore sizes compared to the chemical activation methods. This observation could potentially be associated with the relatively low reactivity of CO_2 towards the oxidation of inorganic components in the raw material, particularly when compared to the efficacy of KOH. Therefore, there is a need to search for cheap and widely available biomass with the most suitable chemical composition and morphological characteristics for activation using CO_2. Based on a review of numerous experimental data, we have established that the use of biomass such as rice husk (RH) or walnut shell (WS), as raw material for the production of AC, is a comparatively reliable and effective choice for obtaining product with the best characteristics such as high SSA, developed porous structure, mechanical and chemical stability.

It was determined that the electrochemical characteristics of AC derived from RH and WSh, activated via KOH and through CO_2 are comprehensively different. Cyclic voltammetry (CV) curves acquired at 10 and 100 mV s^{-1} present a near-rectangular shape, indicative of electric double-layer capacitor behavior, with no peaks for Faradaic reactions observed. It was established that at a low scan rate of 10 mV s^{-1}, RH_KOH and RH_CO_2 reach high specific cell capacitances of 74.7 and 24.3 F g^{-1}. It is also noteworthy that at all scan rates between 5 and 500 mV s^{-1}, the cell capacitance of RH_KOH is significantly higher than that of RH_CO_2. In a general comparison of the RH and WSh, it is easily noticeable that the specific capacity of the for RH electrode activated with CO_2 is the lowest at all scanning speeds and charge-discharge currents, while the electrodes activated KOH exhibit the highest specific capacity. Contrary to rice husk, the characteristics of WSh activated with CO_2 do not lag significantly behind KOH.

We associate these features with the initial composition of the bio-precursors. For instance, the composition of rice husk contains an estimated 15-25% SiO_2 and other inorganic compounds, while the ash content in walnut shell averages 0.1-3.4%. Given that ¼ of the RH composition consists of inorganic components that are less susceptible to the effect of CO_2, the formation of new pores does not take place, ultimately leading to significantly lower SSA and electrochemical characteristics of RH_CO_2. In the sample treated with KOH, the ash and silicon present in the raw material are completely washed away, thereby increasing the total SSA and consequently improving the electrochemical characteristics of RH_KOH. Electrochemical impedance spectroscopy tests were conducted to investigate the resistive behavior of the materials. Similarly to RH, WSh samples exhibit nearly identical contact resistances (Rs ~ 0.36 Ohm). However, the charge transfer resistance of the electrode material derived from the CO2-activation process (Rct ~ 0.1 Ohm) is smaller than that derived from the KOH-activation process (Rct ~ 0.24 Ohm). Although both electrodes have dominant diffusion-controlled processes in the low-frequency region, ion diffusion is faster in WSh_CO_2_2h than in WSh_KOH (relaxation time is 1277ms).

In conclusion, the superior performance of the CO_2-activated material in comparison to the KOH-activated counterpart can be ascribed to key differences in their physicochemical properties. The hysteresis loop scanning analysis of the N_2 adsorption-desorption isotherms disclosed that CO_2 activation of resulted in a material with generally enhanced pore network connectivity.

Dr. Meiram Atamanov is currently acting associate professor at Al-Farabi Kazakh National University and Institute of Combustion Problems working in the field of chemical engineering. This research direction includes experimental study on thermal analyses, combustion, synthesis of carbon nanomaterials and their application.

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