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Relationship between Carbon Electrode Materials and Electrolytes in Capacitive Energy Storage

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This dissertation aims to explore the relationship between the electrolyte and electrodes in capacitive energy storage systems. Specifically, ionic liquid electrolytes were investigated for their performance in different carbon electrode-based supercapacitors. First, the effect of cation size on capacitive performance was observed with onion-like carbon electrodes, which suggests that choosing the right ionic liquid is important for optimizing the system. Additionally, it was shown that ionic liquids have a limited potential window due to the asymmetric performance between the two ions. This can be corrected by simply mixing two ionic liquids together at the correct ratio, expanding the operating window by 1 V and therefore the energy density. This concept of ionic liquid mixtures was also applied to more complex porous carbon electrode materials. By mixing two ionic liquids with different sized cations, the rate performance of a porous carbon material can be improved, taking advantage of the larger ion at low rates and the smaller ion at high rates. The same increase in potential window was shown for the porous carbon as well, suggesting that mixing ionic liquids can improve the performance of many different types of carbon electrodes. Finally, this method of mixing ionic liquids was applied to the development of a model for predicting the ideal mixture concentration for a given carbon materials, specific to its pore size distribution and surface area. The model suggests that by adding 0.1 mole fraction of a smaller anion, the capacitance of the system could be increased by 30%. It can also be extended to different ionic liquids with different sized ions. When experimentally verified, the predicated capacitance was within 6% of the experimental value. This can be further improved with more development of the model and the parameters, including the surface area and pore size distribution of the given material. This model is useful, however, for screening the best electrolytes to test with new materials which are being developed for energy storage every day.