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Pyromellitic diimide-based copolymers and their application as stable cathode active materials in lithium and sodium-ion batteries

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Organic molecules are emerging candidates for the next generation of cost-effective active materials of Li-ion batteries. Small diimide building blocks such as pyromellitic diimide (PMDI) have attracted much attention due to their high theoretical capacity. Many strategies have been undertaken to limit the well-known phenomenon of dissolution of the active material in the electrolyte. Such strategies include the preparation of salts and the synthesis of polyimides or macrocycles. Since dibromopyromellitic diimide exhibits almost no sp² cross-coupling polymerization reaction by conventional synthetic routes (Suzuki-Miyaura or Migita-Stille), we used PMDI as an aromatic C-H bond-bearing unit for direct (hetero)arylation polymerization (DHAP) with 1,4-dibromobenzene as comonomer as a new stabilization strategy. DHAP proved to be an effective tool in the preparation of this polymer, yielding a number average molecular weight of up to 31 kDa. We studied the effect of side-chain engineering using variable chain lengths, cross-linked structures and thermocleavable functional groups. Practical potential limits of 1.65 to 2.50V vs. Li/Li⁺, wherein two distinct redox phenomena appear, and a galvanostatic high rate limit of 2C were determined. Galvanostatic measurements at C/20 show a starting normalized capacity of 0.94 decreasing to 0.48 after more than 80 days (50 cycles). A maximum discharge capacity of 73 mAh/g as a first cycle was obtained for a polymer of this family at C/10. Density functional theory calculations were applied to understand the higher corrected redox potentials obtained by cyclic voltammetry for sodium ion over lithium ion batteries.