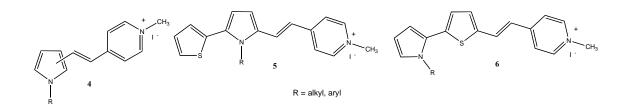
Synthesis, redox and quadratic nonlinear optical properties of novel thienylpyrrole derivatives containing pyridinium electron-acceptor group

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Ouaternized pyridine derivatives have attracted interest for many years from various perspectives, especially for potential applications in electronic/optical devices.¹ Pyridinium salts are readily synthesized, highly stable, and often combine intense visible absorptions with reversible electrochemistry. Notable recently proposed applications of these compounds range from advanced telecommunications to fluorescent probes for bioimaging.² As part of an ongoing program to develop strong charge transfer donor-acceptor thienylpyrrole systems with enhanced nonlinear optical (NLO) properties³ we have explored the potential of pyrrole **4** and thienylpyrrole-based chromophores 5-6 as efficient π -electron donor moieties functionalized with *N*-methylpyridinium acceptor group substituted at thiophene or pyrrole rings (Figure). Compounds 4-6 were prepared, in good yields, through Knoevenagel condensation of the precursor pyrrole 1 or thienylpyrrole $2-3^4$ aldehydes with 1-methylpyridinium salt. On the other hand, formyl-pyrroles 1 were prepared through Paal-Knorr synthesis followed by Vilsmeier formylation. The molecular structure, especially the degree of planarity and the relative orientations of the auxiliary donor and acceptor entities, is found to strongly influence their electrochemical and NLO properties. In this communication we report on the synthesis, redox properties, thermal stability and first order nonlinear optical hyperpolarizabilities for the novel cationic chomophores 4-6. These results indicate that these compounds have good potential for a variety of NLO applications.



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