



The pyrrole methodology for the functionalization of sp^2 carbon allotropes. From the reinforcement of elastomer composites to single atom catalysis

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Natural sources and waste are nowadays the first choice for preparing innovative materials inspired by sustainability. In our research group, C-3 and C-6 building blocks from vegetable oils and lignocellulosic materials, such as glycerol and aldaric acids, and waste proteins were selected for the preparation of functional materials. A pyrrole compound, 2-(2,5-dimethyl-1H-pyrrol-1-yl)-1,3-propanediol (Serinol Pyrrole, Figure 1) was prepared from a glycerol derivative, serinol, through the Paal Knorr reaction with 2,5-hexanedione, performed in the absence of solvents and catalysts, with high yield and water as the only co-product. Serinol pyrrole is a Janus molecule, with two moieties having different chemical behaviour. This result paved the way for the preparation of a large variety of pyrrole compounds (PyC).

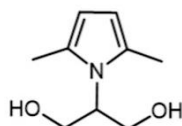


Figure 1. 2-(2,5-dimethyl-1H-pyrrol-1-yl)-1,3-propanediol

The PyC were used for the functionalization of sp^2 carbon allotropes, that occurred with the help of either thermal or mechanical energy, through a domino reaction: the carbocatalytic oxidation of the pyrrole compound followed by the Diels Alder reaction with the carbon substrate. Such a functionalization procedure, named pyrrole methodology, was scaled up hundreds of kilograms, at a semi-industrial scale. The adducts were extremely versatile. The adducts of PyC with carbon black were used as reinforcing fillers in elastomer composites suitable for tyre compounds, improving the vulcanization kinetics and the dynamic-mechanical reinforcement and were used as well as the conductive fillers in protein based bionanocomposites. The adducts of PyC with graphene layers were used for anchoring transition metal atoms, as an easy process for the preparation of single metal catalysts. When silver was the transition metal, antimicrobial inks were prepared. When the transition metals were ruthenium and palladium, high selectivity was achieved in chemical reactions such as isotope exchange and C-O Ullmann coupling.

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