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Value added products from renewable esters and lactones by implementing green chemistry and biocatalysis

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β-Ketonitriles are versatile starting materials, not only for the preparation of a myriad of heteroaromatic compounds (e.g. pyrimidines, pyrazoles, thiophenes and quinolones), but also for their potential to readily afford enantiomerically enriched β-hydroxy carboxylic acid derivatives, with applications for the synthesis of valuable Active Pharmaceutical Ingredients such as non-proteinogenic amino acids for peptidomimetics, a variety of natural products, as well as statins. Traditionally, β-ketonitriles have been prepared by using a variety of often harsh, toxic, environmentally unfriendly and expensive metal-mediated methods. More recently, two greener methods have emerged which generally entail mild, base (KtAmyl and KtBuO)-promoted acylation of alkyl nitriles under ambient conditions. Importantly, these methods were also applicable to enolizable esters, although in some cases the use of excess reagents, excess base and high temperatures were necessary to obtain the desired products in moderate yields. A significant solvent dependency was also observed when applying these milder reaction conditions. Inspired by these methods, we sought to develop similar, preferably more efficient reaction conditions, not only for the preparation of simple aliphatic and aromatic β-ketonitriles, but also for the direct and atom economical ring-opening of lactones to afford chemically versatile, "trifunctionalised" hydroxylated β-ketonitriles. Furthermore, we have demonstrated that these and related compounds can be successfully applied as substrates for the green, and often enantioselective preparation of important β-hydroxy and β-amino acid derivatives through nitrile hydratase, nitrilase, amidase and carbonyl reductase facilitated biotransformations. We are currently investigating various potential applications of the lactone-derived β-hydroxynitriles, including the stereoselective synthesis of heterocyclic compounds such as alkaloids through ring closure.

Biography

Dr. Daniel Pienaar grew up in Pretoria, South Africa, where he obtained three degrees with distinction (in Biochemistry and Organic Chemistry) at the University of Pretoria, after which he migrated to the University of the Witwatersrand (WITS), Johannesburg, to complete a PhD degree in Natural Product Organic Synthesis in 2005. Daniel started his professional career as a part-time lecturer at WITS, soon followed by a contract R&D position at Industrial Distillers & Refiners in Johannesburg, where he successfully developed a proprietary superplasticiser product with applications in the Concrete Industry. Subsequently, in January 2007, he was appointed as the first employee of Oxyrane, a Council for Scientific and Industrial Research spin out Biotechnology company, which is still doing ground breaking R&D work in the Life Sciences today. He continued to work as a pioneering R&D Scientist for the company for almost nine years (in the UK and in Belgium), leading the development of proprietary chemo-enzymatic routes to a variety of high-value Active Pharmaceutical Ingredients (APIs). In September 2014, he took up a Senior R&D Scientist role within the Biotechnology (Sustainability) Group of Invista, a Koch Industries subsidiary. During his time at Invista, he developed chemo-enzymatic routes to obtain biological pathway intermediates (for enzyme screening purposes) and novel two-step (fermentation, chemical) processes for the production of olefins (monomers used in the materials industry). The work included the development of medium- to high-throughput Biochemical assays and he also served as the Analytics Technical Group Leader here. Recently, he returned to South Africa and is currently employed as a Research Fellow in Green Chemistry at the Molecular Sciences Institute, School of Chemistry at WITS.

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