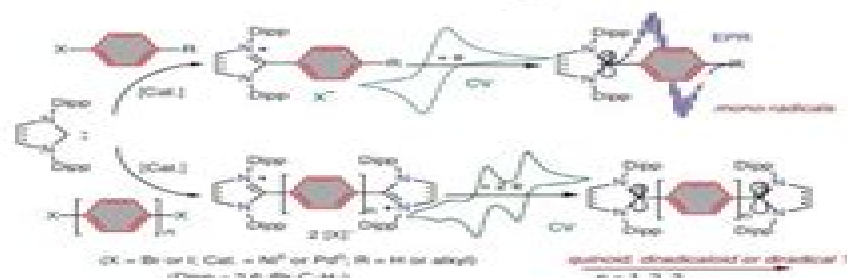


Stable Carbon-Centered Radicals Based on N-Heterocyclic Carbenes

Rajendra S. Ghadwal*

Anorganische Molekülchemie und Katalyse, Anorganische Chemie und Strukturchemie, Centrum für Molekulare Materialien, Institut für Chemie, Universität Bielefeld, Universitätsstraße 25, 33615 Bielefeld, Germany
rghadwal@uni-bielefeld.de



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Abstract Carbon-centered radicals and diradicaloids based on classical N-heterocyclic carbene (NHC) scaffolds are readily accessible in crystalline solids. The presence of an aryl (Ar) substituent at the C2-position is the key to the remarkable stability of these open-shell species as it provides appropriate room for the spin-density delocalization. Two catalytic as well as high-yielding protocols have been developed to install a suitable aryl group at the C2-position of NHCs. The spin-density in mono-radicals (NHC[•])⁺ is mostly located on the parent carbene-carbon (C2) atom. The bridging of two NHCs through a phenylene spacer (C₆H₄), enables the isolation of various p-quinodimethane (p-QDM) derivatives, which may be considered as open-shell Kekulé diradicaloids. The diradical character of these NHC-analogues of Thiels (n = 1), Chichibabin (n = 2), and Müller (n = 3) hydrocarbons [(NHC)(C₆H₄)_n(NHC)] can be tuned by a rational choice of the size and/or the topology of spacers. In this account, the synthesis, structure, and properties of this new class of radical hydrocarbons is presented.

- 1 Introduction
- 2 NHC-Monomericals
- 3 NHC-Diradicaloids
- 4 Conclusion

Key words: radical, diradical, diradicaloid, carbene, hydrocarbon, heterocycle, open-shell, paramagnetic

1 Introduction

Radicals are fundamental intermediates¹ and they play crucial roles in numerous biological² and industrially important processes.³ In general, radicals are short-lived species and, therefore, in a condensed phase, spontaneously react with surrounding (solvent, precursor, or by-product) molecules, undergo self-decomposition or dimerization to eventually form closed-shell products.⁴ Consequently, syn-

thetic access to thermally stable⁵ radicals for detailed structural investigations has been a subject of intense fundamental interest for augmenting our understanding about chemical bonding and electronic structure.⁶ In addition, stable radicals are also highly promising class of compounds for their potential applications in materials science and emerging areas.⁷ This is because of their unique optical, electronic, and magnetic properties.^{8a,b} The potential of stable radicals as unique building blocks for the design of next-generation advanced functional materials for applications in optoelectronics, spintronics, and solar cells is substantial.⁹ The incorporation of stable radicals into artificial molecular machines (AMMs) is very appealing because an electron's spin can be much easier and more precisely controlled, even with a smaller energy input, than a charge.¹⁰ In this context, carbon-centered radicals have special advantages because of their planar trivalent nature, which promises structural diversity and elaborate designs. Stable radicals are also of immense interest as active materials in organic radical batteries (ORBs).¹⁰ These so-called 'eco batteries' have the potential to be cheaper, safer, and longer-lasting than current metal-based batteries and, thus, are of ever-growing importance in our modern technology-based society.¹¹ This becomes evidently more appealing if such radicals are derived from 1,3-imidazoli(n)ium precursors, which are promising electrolytes for battery applications. In recent years, radical catalysis has also emerged as an important tool in organic synthesis.¹²

Stable Radicals and Radicaloids

In 1900, Gomberg isolated the first stable radical, the triphenylmethyl I (Scheme 1).¹³ This seminal report marked the dawn of stable radical chemistry. I, however, undergoes self-association in the condensed phase, yielding the α -dimer II. Although the delocalization of the unpaired electron

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