Syntheses and Properties of Cycloamidines Based on 4*H*-Imidazoles*

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Employing three different syntheses a broad spectrum of 4*H*-imidazoles **3a** – **3s** has been synthesized. In the course of the two-fold aminolysis reaction leading to derivatives **3q** – **3s**, deeply colored byproducts could be isolated and structural characterized.These novel donor-acceptor derivatives of type **7** consist of an 1*H*- and 4*H*-imidazole which are connected by a nitrogen bridge and rearrange *via* rapid 1,3-/1,5-hydride shifts. Using ^{1}H NMR experiments the aminolysis product **3p** shows prototropic isomers which could be detected in equilibrium for the first time. Cyclovoltammetric measurements of a series of substituted 2-aryl derivatives **3d** – **3i** displayed two reversible single electron transfer steps with relatively small semiquinone formation constants between 10^2 and 4×10^3. The 4*H*-imidazole **3d** was successfully converted into boratetraaza-pentalene **8a**, which showed two well separated reduction potentials. The value of semiquinone formation constant of **8a** (1.8×10^{15}) is even higher than those reported for similar derivatives. 4*H*-imidazoles can also be employed for the efficient complexation of catalytically important metals as exemplified by copper complexes **11** and **12**. Derivative **3m**, which possesses an additional chelating pyridine substructure, formed a stable complex of structural composition Zn(**3m**)$_2$ with diethyl zinc.

Key words: Fulvenimines, 4*H*-Imidazoles, Aminolysis, Diazaborolidines, Metal Complexes, Redox Systems