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METAL-FREE C–H/N–H CROSS-COUPLING OF CYCLIC ALDONITRONES WITH NH-AZOLES

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Abstract. Cyclic nitrones are the class of compounds widely known not only as efficient spin traps and potential therapeutics but also as versatile chemical synthons, affording one to achieve azaheterocyclic derivatives of diverse architecture. Hereby, the direct C(sp^2)–H functionalization of aldonitrone moieties belongs to one of the most viable strategies towards their modification due to the inherent step-efficiency of the methodology mentioned.

Using model cyclic aldonitrones, 2*H*-imidazole 1-oxides **1**, we have demonstrated that such substrates can be involved into the cross-dehydrogenative couplings with both carbon- and nitrogen-centered synthons. In particular, a series of NH-azoles **2** have proved themselves beneficial in terms of the reaction, the process being tolerant to various functional groups. Besides, it turned out to be not necessary to employ transition metal catalysis to carry out the coupling; instead, the green reagent Selectfluor® has been found to mediate the reaction successfully. Moreover, the substantial yield improvement has been achieved through the exposure of the reaction media to blue light irradiation. Using the described approach, we have obtained 25 azolyl-substituted cyclic ketonitrones **3** in yields up to 94%. Further investigations of the reaction scope are currently underway in our lab.

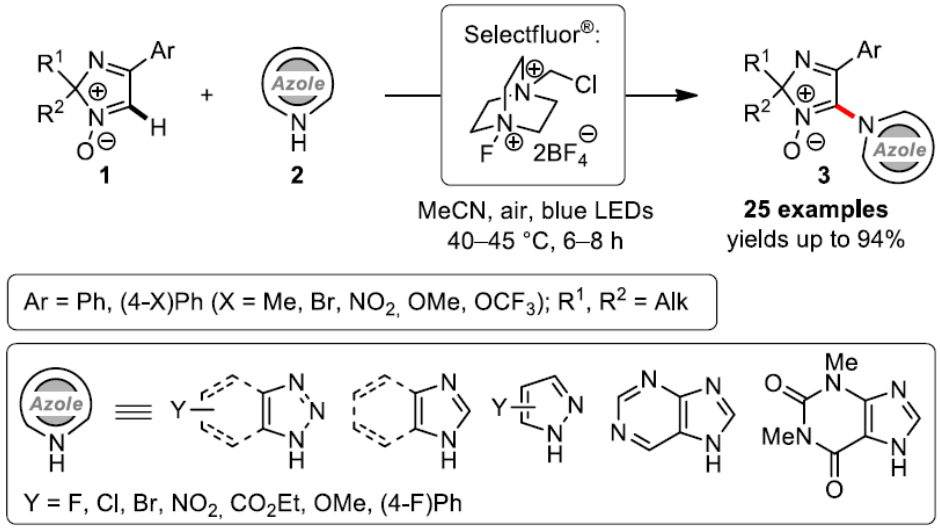

Figure 1. Cross-coupling of 2*H*-imidazole 1-oxides **1** with NH-azoles **2**

References

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