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## A Tunable Route to Oxidative and Eliminative [3+2] Cycloadditions of Organic Azides with Nitroolefins: CuO Nanoparticles Catalyzed Synthesis of 1,2,3-Triazoles under Solvent-Free condition

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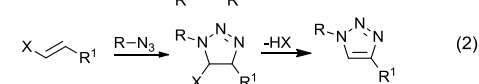
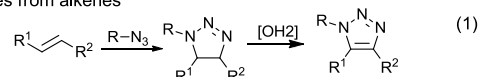
### ABSTRACT

A regioselective and tunable synthesis of 1,5-disubstituted 1,2,3-triazoles from oxidative and eliminative [3+2] cycloadditions of nitroolefins and organic azides under solvent-free condition in the presence and absence of commercially available CuO nanoparticles catalyst is described. In the presence of the catalyst under solvent-free condition, nitro group is retained in the product while it gets eliminated in the absence of the catalyst.

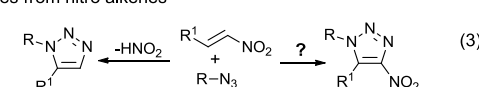
1,2,3-Triazoles have gained paramount importance for the past two decades owing to their versatile applications in various fields such as drug discovery,<sup>1</sup> medicinal chemistry,<sup>2</sup> supramolecules,<sup>3</sup> functional coatings,<sup>4</sup> material science and polymers.<sup>5</sup> In addition, they contribute to the chemical synthesis by serving as synthetic precursors to many valuable compounds.<sup>6</sup> Particularly, 1,2,3-triazoles bearing NO<sub>2</sub> group gain additional importance since they are the starting materials for nitrogen group substituted 1,2,3-triazoles. Apart from acting as nitrogen source, NO<sub>2</sub> group can be derivatized further to access diverse molecules.<sup>7</sup> In spite of the conventional Huisgen cycloaddition,<sup>8</sup> major breakthrough was achieved in this area only after the discovery of copper catalyzed azide-alkyne cycloaddition (CuAAC)<sup>9</sup> which amassed numerous publications encapsulating a wide spectrum of applications in various fields. Even though CuAAC epitomises ideal route to access 1,4-disubstituted 1,2,3-triazoles, it is limited to terminal alkynes only. Further advancement in this area was achieved by the development of Ruthenium catalyzed azide-alkyne cycloaddition (RuAAC)<sup>10</sup> which is stunningly selective to the complementary 1,5-disubstituted 1,2,3-triazoles and it works out for internal alkynes as well. As an alternative approach to azide-alkyne cycloaddition, olefins were envisaged in the place of alkynes since olefins are easily accessible and economically viable than alkynes. This type of azide-olefin cycloaddition was pioneered by Huisgen and Labbe<sup>11</sup> with organic azides and electron deficient olefins. The product of azide-olefin

cycloaddition is triazolone which is an unstable compound and it readily decomposes into different products as implicated by the reaction conditions.<sup>12</sup> At this juncture, several methods were sought out to transform this unstable triazolone into the stable triazole. In this regard, two ingenious strategies have been reported in the literature. First strategy is oxidative azide-olefin cycloaddition (OAOC) where the triazolone formed by the cycloaddition of azide and olefin will subsequently be oxidized into the triazole [Eq. (1), Scheme 1].

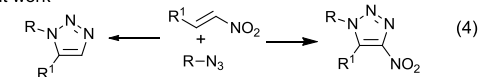
#### Triazoles from alkenes



#### Triazoles from nitro alkenes



#### Present work



Scheme 1. Background of [3+2] cycloaddition of nitro alkenes and azides.

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