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Rearrangement of 4-oxobutane-1,1,2,2-tetracarbonitriles to the penta-1,3-diene-1,1,3-tricarbonitrile moiety as an approach to novel acceptors for donor-acceptor chromophores

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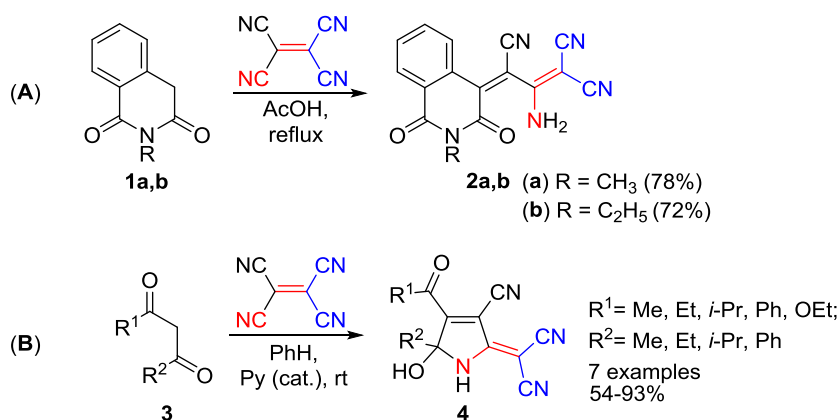
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Abstract – The rearrangement of 4-oxobutane-1,1,2,2-tetracarbonitriles to give the penta-1,3-diene-1,1,3-tricarbonitrile moiety, accompanied by elongation of the carbon chain *via* introduction of the cyano group carbon atom to the carbon skeleton, is described. The rearrangement in acetic acid in the presence of ammonium acetate allows the synthesis of 2-(3-cyano-7a-hydroxy-5,6,7,7a-tetrahydro-1*H*-indol-2(4*H*)-ylidene)malononitriles, which are structural analogs of practically significant tricyanofuran (TCF) and tricyanopyrrole (TCP) acceptors for donor-acceptor chromophores.

Keywords: Rearrangement; 4-oxobutane-1,1,2,2-tetracarbonitriles; pyrroles; cyano groups; donor-acceptor chromophores.

Rearrangements are an important group of reactions which have the advantage, in comparison to other reactions, of the possibility of a one-step transformation of the carbon skeleton. In recent years a number of reviews describing the use of rearrangements in organic synthesis, including for the preparation of practically important products, have been reported.^{1a-f} In this regard, the study of new rearrangement reactions of organic compounds is an important task.

A prerequisite for this work was the report of an unexpected reaction pathway of tetracyanoethylene (TCNE) with some methylene-active compounds: imides **1** (Scheme 1, A),^{2a} 1,3-diketones and ketoesters **3** (Scheme 1, B).^{2b} Thus, it was reported that compounds **2** were obtained as a result of the reaction between imides **1** and TCNE in acetic acid,^{2a} wherein the carbon skeleton rearrangement of the incoming TCNE moiety is obvious from the structure of products **2** (Scheme 1, A). Also, the formation of pyrrole derivatives **4**, structurally similar to compounds **2**, was described for the reaction of 1,3-dicarbonyl compounds **3** with TCNE in benzene (Scheme 1, B).^{2b}



Scheme 1. Rearrangement during the reaction of methylene-active imides **1** (A) and 1,3-dicarbonyl compounds **3** (B) with TCNE.

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