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## **Research Paper :**

# Catalytic transfer hydrogenation studies on Arylhydrazones

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#### ABSTRACT

The catalytic transfer hydrogenation studies on several arylhydrazones, prepared by coupling aryldiazonium salts with diethylmalonate and acetyl acetone have been carried out. It has been found that even under a variety of reaction conditions the reduction of diethyl mesoxalate-2-o-nitrophenylhydrazone or pentane-2,3,4- trioxo-3-o-nitrophenylhydrazone in anhydrous formic acid as a solvent and as a hydrogren donor with 10 per cent pd-C as a catalyst produces the N-formyl derivative of the free amino compound.

**KEY WORDS**: Catalytic Transfer hydrogenation studies, N- formyl derivative

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The synthetically and industrially important reduction of nitro compounds has been effected in many ways<sup>1-</sup> <sup>3</sup> (Entwistley *et al.*, 1977, Coutts and Edwards, 1966 and Rylander, 1970). The reduction of nitrogroup attached to the aromatic nucleus of complex arylhydrazones (I) has been accomplished by various research groups<sup>4-6</sup> (Cadagon *et al.*, 1970, Campbell and Rees, 1969 and Rissi *et al.*, 1968). During the present course of investigation to design new snythetic strategies to different benzofused heterocyclic compounds<sup>7</sup> (Selvi and Perumal, 2002), an attempt was made to reduce the hydrazones (I) by catalytic transfer hydrogenation. But contrary to the expectations, the N-formyl derivative of free amino compound<sup>8</sup> (Prasad and Singh, 2004) was furnished. Its broad scope as well as the easy access to N-formyl



derivatives should make this methodology widely applicable in organic sythesis.

Catalytic transfer reductions are a considerable technical improvement over rather. messy traditional reduction with metal and acid and appear to be selective than regular catalytic hydrogenation<sup>9</sup> (Braude *et al.*, 1954).

It is the reduction of an organic compound by use of a hydrogen donor in the presence of a catalyst. The reaction can be generalised as follows:

$$DH_{x} + nA \xrightarrow{Catalyst} nAH_{x} + D$$
  
Solvent

The donor compound DHx can be any organic compound whose oxidation potential is sufficiently low so that hydrogen transfer can occur under mild conditions.

The early pioneering work by Braude<sup>9</sup> was largely ingonred because of poor yields and long reacion times, but the situation has changed considerably following the appearance of a stimulating review and the introduction of greater catalyst loadings and different hydrogen donors.

The reduction of diethylmesoxalate-2-onitrophenylhydrazone (II) in anhydrous formic acid as a solvent and as a hydrogen donor with 10 per cent Pd-C produced the N- formylderivative of the free amino compound (III). By using ethanol as a solvent and sodium formate as a hydrogen donor, the free amino compound (IV) was obtained as a red oil.

The compounds (III) and (IV) were the same