# **Reactions of aromatic nitro compounds with acetophenone in the presence of alkali.** Crystal structure of **(E)-1,2-dibenzoyl-***o***-tolylaminoethylene**

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Methyl and methoxy substituted nitroarenes were reacted with acethophenone in the presence of sodium ethoxide in ethanol giving arylamino-conjugated diketo derivatives. A case of *ipso*-substitution of a methoxy group is discussed. The title compound  $C_{23}H_{19}NO_2$  ( $M_r = 341.4$ ) crystallizes in the monoclinic system, space group  $P2_1/n$  with a = 21.579(4), b = 8.526(2), c = 9.983(2) Å;  $\beta = 92.3(1)^\circ$ ; V = 1835.2(7) Å<sup>3</sup>, Z = 4,  $D_c = 1.24$  g cm<sup>-3</sup>,  $\mu$ (Cu-K $\alpha$ ) = 5.9 cm<sup>-1</sup>,  $\lambda = 1.5418$  Å, F(OOO) = 720. The molecule adopts the *E* configuration with the C=O carbonyl groups *trans* with respect to the ethylenic double bond.

#### Introduction

Substantial amounts of work has been done on the reactivity of aliphatic nitro-compounds with nucleophiles and, in particular, on the reactivity of  $\alpha$ -substituted aliphatic nitro-compounds (Bowmann, 1988). Most of the study has been on the mechanistic aspects of these reactions and on synthetic applications. A minor interest has been the study of aromatic nitro-compounds, mainly on the effect of the nitro group on the benzene ring rather than the reactivity and the transformation of the nitro group itself (Kornblum, 1983). Nitrobenzenes were converted into the corresponding azoxybenzenes by reaction with sodium ethoxide (Frey and Cameron 1927), and the reaction was kinetically investigated (Ogata and Mibae, 1961). o-Nitrostylbenes as well as o-nitrophenylethylenes (Hiremath and Hooper, 1978) were used to obtain isatogens via intramolecular cyclization. o-Nitroaniline derivatives, under reductive condition, undergo cyclization to form benzimidazoles (Harvey *et al.*, 1988; Stacy *et al.*, 1964) and benzimidazole-*N*-oxides (Berti *et al.*, 1979; Takahashi and Kano, 1963), depending on the reagent used. In the present paper we describe the reaction of some substituted nitrobenzenes with acetophenone in ethanol in the presence of sodium ethoxide. The structure of one of the products isolated, namely (E)-1,2-dibenzoyl-otolylaminoethylene (**3b**) is also reported. The reaction, already described by Scorrano (Paradisi *et al.*, 1981) under similar conditions was first studied using the *ortho*-nitrotoluene with the aim of obtaining cyclized compounds through the intermediate (**6**) of Scheme 2.

#### Experimental

#### Physical measurements

Melting points were determined on a micro hotstage melting point apparatus and are uncorrected. Solid state i.r. spectra were obtained using a Nicolet Fourier Transform Infrared 20-SX Spectrometer equipped with a Spectra Tech. Multiple Internal Reflectance, DRIFT, apparatus. <sup>1</sup>H nmr spectra were recorded on a Varian XL 100 using TMS as internal standard.

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Compounds (1a-e) and acethophenone were Aldrich while all solvents were Carlo Erba RP-ACS grade reagents.

# General procedure for the synthesis of compounds (3a-e)

To a solution of sodium (0.35 g, 0.02 mole) dissolved in 15 ml of absolute ethanol, the nitroderivative (1a-e) (0.02 mole) and acethophenone (2.0 g, 0.02 mole) were added at room temperature. The solution was boiled for 1 h, cooled (compounds (3a) and (3b) precipitated) and 100–200 ml of water were added. The mixture was extracted with diethyl ether, the organic layers washed with water until neutrality and dried over sodium sulphate. After taking the solution to dryness, the residue was passed through a silica column using petroleum ether/acetone 9:1 as an eluant. The yields for the yellow compounds (3a-e) are reported in Table 1.

## X-ray analysis

1,2-Dibenzoyl-o-tolylaminoethylene (3b) crystallizes in the monoclinic system as yellow prismatic crystals. A specimen of 0.19 × 0.19 × 0.43 mm was employed for data collection on a Siemens AED singlecrystal diffractometer on line to an IBM PS/2 M30 computer. Unit cell parameters were obtained by using a program (Belletti *et al.*, 1979) which repeatedly checks on the diffractometer the values of  $(\vartheta, \chi, \psi)$  angles of thirty reflections  $(20 \le \vartheta \le 30^\circ)$  to obtain the maximum of the peak when the angles are not moving more  $0.01^\circ$ . Crystallographic data are as follows: C<sub>23</sub>H<sub>19</sub>NO<sub>2</sub>,  $M_r = 341.4$ . Monoclinic, a = 21.579(4), b = 8.526(2), c = 9.983(2) Å;  $\beta = 92.3(1)^\circ$ ; V = 1835.2(7) Å<sup>3</sup>, Z = 4,  $D_c = 1.24 \text{ g cm}^{-3}$ ,  $\mu(\text{Cu}-K\alpha) = 5.9 \text{ cm}^{-1}$ ,  $\lambda = 1.5418 \text{ Å}$ , F(000) = 720. Space group  $P2_1/n(C_{2h}^5, \text{ No.} 14)$  from systematic absences.

The X-ray measurements were performed at T =293 K in the range  $(3 \le \vartheta \le 70^\circ)$  using Cu K $\alpha$  radiation. The diffraction angle for every reflection was determined on the basis of the orientation matrix and the outline of the diffraction peak was collected in the  $\vartheta - 2\vartheta$ step scanning mode using a scan width from  $(\vartheta - 0.60)^{\circ}$ to  $(\vartheta + 0.60 + \Delta \lambda / \lambda tg \vartheta)^\circ$ . The intensities  $I_{hkl}$  were determined by analyzing the reflection profile with the Lehmann and Larsen procedure (Lehmann and Larsen, 1974). One "standard" reflection, measured every 50 collected reflections to monitor crystal decomposition and instrumental linearity, showed no significant variation. 3830 symmetry independent reflections ( $-26 \le h$  $\leq 26, 0 \leq k \leq 10, 0 \leq l \leq 12$ ) were measured, of which 1850 (internal R merging factor 0.016) having  $I_{hkl}$ >  $2\sigma(I_{hkl})[\sigma(I_{hkl})]$  based on statistic counting] were used in the refinement. Corrections for Lorentz and polarization effects were applied, but none for absorption.

The structure was solved by direct methods by use of the SHELXS86 (Sheldrick, 1986) program and refined by SHELX76 (Sheldrick, 1976) with cycles of full-matrix anisotropic least-squares (hydrogen atoms isotropically) up to R = 0.043,  $R_w = 0.048$ ;  $\Sigma w (F_o - F_c)^2$  minimized with  $w = [\sigma^2(F_o) + 0.010F_o^2]^{-1}$ . All the hydrogen atoms were located in the  $\Delta F$  map. The final  $\Delta F$  map was featureless with no peaks >0.14 e Å<sup>-3</sup>; the maximum shift-to-error in the last cycle was 0.24. Atomic scattering factors from International Tables for X-ray Crystallography (1974).

All calculations were carried out on the GOULD 6040 POWERNODE computer of the Centro di Studio

		¥7.11		I.R. DRIFT			I.R. E			
Compd.	м.р. (°С)	(%)	Formula	C	H H	N	(NH)	(CO)	δ <sub>H</sub>	mass (m/e)
(3a)	126	31	C <sub>22</sub> H <sub>17</sub> NO <sub>2</sub>	80.38 (80.71)	5.54 (5.23)	4.02 (4.28)	3324s	1674	6.13(1H,=CH-);6.9-7.2(11H,m.arom.); 7.87-8.10(4H,m.arom.)	77(82);105(77);222 (100);327(12)
(3b)	105	15	$C_{23}H_{19}NO_2$	81.02 (80.92)	5.47 (5.61)	4.17 (4.10)	3332b	1678	2.43(3H,s,Me);6.17(1H,s,=CH-); 6.84-7.69(10H,m,arom.);7.87-8.10(4H,m,arom.)	77(100);91(20);105 (97);236(84);341(10)
(3c)	154	30	$C_{23}H_{19}NO_2$	80.78 (80.92)	5.87 (5.61)	4.02 (4.10)	3324s	1675	2.21(3H,s,Me);6.1(1H,s,=CH-);6.93(4H,d,arom) 7.58-7.62(6H,m,arom.);7.9-8.1(4H,m,arom.)	77(38);105(64);236 (100);341(13)
(3d)	120	35	$C_{23}H_{19}NO_3$	77.52 (77.29)	5.25 (5.36)	4.05 (3.92)	3324s	1670	3.70(3H,s.Me);6.08(1H.s.=CH); 6.83(4H,pseudo-q,p-MeO-C <sub>6</sub> H <sub>4</sub> );7.35-7.60 (6H,m,arom.);7.87-8.04(4H,m,arom.)	77(45);105(80);252 (70);357(15)
(3e)	139	8	C <sub>24</sub> H <sub>21</sub> NO <sub>3</sub>	77.05 (77.61)	5.98 (5.70)	3.89 (3.77)	3324s	1670	1.33(3H,t,CH <sub>2</sub> CH <sub>3</sub> );3.92(2H,q,CH <sub>2</sub> CH <sub>3</sub> ); 6.08(1H,s,=CH-);6.81(4H, <i>pseudo-q,p</i> -EtO-C <sub>6</sub> H <sub>4</sub> -); 7.18-7.60(6H,m,arom.);7.85-8.04(4H,m,arom.)	77(62);105(72);266 (57);371(16)

Table 1. Analytical and spectroscopic data of compounds (3a-e)

per la Strutturistica Diffrattometrica del C.N.R. of Parma. Bibliographic searches were carried out using the Cambridge Crystallographic Data Files through the Servizio Italiano di Diffusione Dati Crystallografici.

#### **Results and discussion**

#### Spectroscopic results

The reaction of nitrobenzenes (1a-c) with acetophenone (2) was carried out in ethanol in the presence of sodium ethoxide in 1:1:1 molar ratios under reflux for 1 h. In all cases the products (3) were isolated and identified. Analytical and spectroscopic data and the yields are reported in Table 1.

The reactions were not clean because many products were formed together with compounds (3), which generally were separated by chromatography on a silica gel column and, in some cases (3a) and (3b), by filtration of the cooled reaction mixture. All attempts to isolate other compounds in a pure solid state failed, because they give mixtures unresolvable by chromatography. The structure determination by X-ray analysis of compound (3b) (Fig. 1) and the consequent interpretation of its spectroscopic data allowed us to identify compounds (3a) and (3c-e) by comparing their spectroscopic data with those of compound (3b). For all the compounds (3a-e) the NH stretching frequencies fall at 3324  $\text{cm}^{-1}$ with the exception of (3b)  $(3332 \text{ cm}^{-1})$ . The carbonyl CO of (3a) (1674 cm<sup>-1</sup>) is red shifted in the  $\rho$ -methoxy derivative (3d) (1670  $\text{cm}^{-1}$ ) and blue shifted in the hindered (3b) (1678  $\text{cm}^{-1}$ , corresponding to that of the carbonyl group of the acetophenone reported in the Aldrich IR Library). The carbon-carbon aliphatic and aromatic double bond stretching modes, being conjugates, are located at lower wavenumbers and close to each other, making a distinction, among (3a-d), difficult. The particular experimental procedure (DRIFT) makes the small frequency differences reliable and allowed us to confirm, in the solid state, the presence of a chelate planar intra H-bonded structure (Conley, 1966; Kaberia et al., 1980). In compound (3b), the o-Me group introduces a distorsion in the six-membered ring, as proved by the above spectroscopic evidence. The <sup>1</sup>H nmr spectrum of (3b) shows a singlet at  $\delta 2.43$  for the methyl group, a singlet at  $\delta 6.17$  for the hydrogen linked to the carbon-carbon double bond, an aromatic multiplet at  $\delta$ 7.87–8.10 due to four aromatic hydrogens. The latter multiplet and the singlet at  $\delta 6.17$  are common to all the products (3a) and (3c-e). Since the multiplet is due to the four hydrogens in the *ortho* position to the carbonyl group, it can be concluded that the isolated compounds have the same structure.

It is worth noting that in the reaction of (1d) compound (3e) was isolated, though in very low yield, and its formation may be explained by a nucleophilic *ipso*substitution of the methoxy group of (3b) by an ethoxy group, a case which is well documented in the literature (Artakina, 1982; Illuminati, 1985; Terrier, 1982).

The reaction reported in Scheme 1, could be likely be interpreted as involving a nucleophilic attack of the anion (4) on the nitro group leading to the intermediate (6), whose formation could be explained by the sequences reported in Scheme 2.

Most of the reactions of the nitro-compounds car-



Scheme 1



ried out with nucleophiles concern the directing effects of the nitro group (Fever 1970; Kornblum, 1983), and only a few examples of nucleophilic addition directly concerning the nitro group by Grignard reagents have been reported (Bartoli et al., 1985); Mayer and Bilbroth, 1919). Thus, our conjecture on the formation of the intermediate (6) is plausible and the mechanism proposed in Scheme 2 may be considered reasonable even if no intermediate was identified. The mechanism proposed by Scorrano (Paradisi et al., 1981) hypothesizes the formation of a nitrosoarene as the first step of the reaction, but seems inconsistent with the observation that the yield of the final product is much lower when the reaction is carried out directly on the nitrosoarene. On this basis, we suggest that the acetophenone anion may directly attack the nitro group as indicated in Scheme 2.

# Molecular geometry

Final atomic coordinates and equivalent isotropic thermal parameters for nonhydrogen atoms are given in Table 2. Figure 1 shows a perspective view of the molecule together with the arbitrary numbering scheme used in the crystal analysis. Bond distances and angles, reported in Table 3 are in line with the hybridization expected for the atoms. In particular the C=C double bond [1.364(3) Å] is of special interest being longer than that found in other ethylene derivatives reported in the literature [1.321(3) Å in (Z)-9-(2,5-dimethyl-styryl)acridine, 1.328(3) Å in (E)-9-styrylacridine (Sgarabotto *et al.*, 1989); 1.314(4) Å in 1-anisyl-2,2-diphenylvinyl bromide, 1.320 Å in *p*-acetamido- $\alpha$ -bromostyrene (Kaftory *et al.*, 1985)] and also longer than

that found in similar ethylene substituted derivatives [1.31 and 1.32 Å in tetrabenzoylethylene, 1.327(6) Å in tetrabenzoylethylene carbon disulfide solvate, 1.344(2) Å in tetraacethyl ethylene (Cannon *et al.*, 1978] but comparable with the 1.365(4) Å found in 1,4-

C(25)

C(24)



C(27

Fig. 1. Perspective view of the compound (3b) showing the atomic numbering scheme.

diaryl-2-(arylamino)but-2-ene-1,4-diones (Paradisi *et al.*, 1981), a compound differing from that described here only by the absence of the C(27) methyl substituent at the aminic phenyl. The molecular conformations of these two compounds are exactly alike with the C=O carbonyl groups *trans* with respect to the ethylenic double bond (E configuration).

The two carbonyl groups are differently oriented with respect to the plane of the ethylenic bond: in fact O(4) is involved in an intramolecular hydrogen bond with the aminic nitrogen atom  $[N(2) \cdots O(4) 2.654(4),$ H(2)  $\cdots O(4) 1.89(3)$  Å; N(2)—H(2)  $\cdots O(4)$ 139.1(26)°] and, owing to this interaction, the C(4)=O(4) bond is forced to adopt a position coplanar with the ethylenic bond as shown by the value of the C(2)-C(3)-C(4)-O(4)  $[-0.4(5)^{\circ}]$  torsion angle reported in Table 3; on the contrary, the C(1)=O(1) carbonyl group which is not hindered in its rotational freedom around the C(1)-C(2) bond adopts a position consistent with the steric hindrance of the adjacent

**Table 2.** Fractional atomic coordinates  $(\times 10^4)$  and equivalent isotropic thermal parameters  $(\times 10^4 \text{ Å}^2)$  for non-H-atoms with esd's in parentheses

	U,			
	x	у	z	$U_{ m eq}$
0(1)	4024(1)	2060(2)	-4520(2)	678(4)
O(4)	5203(1)	1734(2)	10(2)	574(7)
N(2)	4161(1)	2795(3)	-1167(3)	509(8)
C(1)	4176(1)	2993(4)	-3643(3)	486(10)
C(2)	4438(1)	2401(3)	-2304(3)	455(9)
C(3)	4991(1)	1626(4)	-2313(3)	487(10)
C(4)	5365(1)	1305(3)	-1124(3)	480(10)
C(11)	4161(1)	4716(3)	-3867(3)	443(9)
C(12)	4425(2)	5763(4)	-2947(3)	557(12)
C(13)	4420(2)	7358(4)	-3202(4)	622(13)
C(14)	4148(2)	7910(4)	-4390(3)	611(11)
C(15)	3885(2)	6880(4)	-5308(4)	645(13)
C(16)	3895(2)	5286(4)	-5056(3)	574(11)
C(21)	3565(1)	3439(3)	-1026(3)	514(10)
C(22)	3070(2)	3003(5)	-1877(4)	686(13)
C(23)	2487(2)	3655(6)	-1685(5)	858(18)
C(24)	2403(2)	4683(6)	-660(5)	929(19)
C(25)	2893(2)	5087(5)	190(4)	804(16)
C(26)	3485(2)	4473(4)	38(3)	587(11)
C(27)	4010(2)	4883(5)	993(4)	748(15)
C(41)	5977(1)	527(4)	-1247(3)	488(9)
C(42)	6102(2)	-469(4)	-2313(3)	541(11)
C(43)	6681(2)	-1177(4)	-2385(4)	651(13)
C(44)	7136(2)	-880(5)	-1420(4)	752(15)
C(45)	7021(2)	120(5)	-369(4)	799(15)
C(46)	6442(2)	804(4)	-273(4)	663(14)

parentneses						
O(1)-C(1)	1.218(4)	C(15)-C(16)	1.382(5)			
O(4)-C(4)	1.253(4)	C(21) - C(22)	1.389(5)			
N(2) - C(2)	1.347(4)	C(21) - C(26)	1.396(4)			
N(2) - C(21)	1.411(3)	C(22)-C(23)	1.396(6)			
C(1)C(2)	1.517(4)	C(23) - C(24)	1.365(7)			
C(1)-C(11)	1.486(4)	C(24)-C(25)	1.373(6)			
C(2)-C(3)	1.364(3)	C(25)-C(26)	1.395(6)			
C(3)-C(4)	1.435(4)	C(26)-C(27)	1.493(6)			
C(4) - C(41)	1.487(3)	C(41) - C(42)	1.396(5)			
C(11) - C(12)	1.386(4)	C(41) - C(46)	1.389(5)			
C(11) - C(16)	1.386(4)	C(42)-C(43)	1.392(6)			
C(12) - C(13)	1.384(5)	C(43) - C(44)	1.371(6)			
C(13) - C(14)	1.384(5)	C(44) - C(45)	1.382(6)			
C(14)-C(15)	1.375(5)	C(45) - C(46)	1.386(6)			
C(2) - N(2) - C(21)	128.2(3)	N(2) - C(21) - C(22)	121.1(3)			
O(1) - C(1) - C(2)	119.8(3)	N(2) = C(21) = C(26)	117.6(3)			
O(1) - C(1) - C(11)	122.3(3)	C(22) - C(21) - C(26)	121.3(4)			
C(2) - C(1) - C(11)	117.8(3)	C(21) - C(22) - C(23)	119.0(4)			
N(2) - C(2) - C(1)	119.8(3)	C(22) - C(23) - C(24)	120.4(5)			
N(2) - C(2) - C(3)	123.0(3)	C(23) - C(24) - C(25)	120.2(5)			
C(1) - C(2) - C(3)	116.8(3)	C(24) - C(25) - C(26)	121.7(4)			
C(2) - C(3) - C(4)	123.4(3)	C(21) - C(26) - C(25)	117.5(3)			
O(4) - C(4) - C(3)	121.6(3)	C(21) = C(26) = C(27)	121.3(4)			
O(4) - C(4) - C(41)	119.1(3)	C(25) - C(26) - C(27)	121.2(4)			
C(3) - C(4) - C(41)	119.3(3)	C(4) - C(41) - C(42)	122.2(3)			
C(1)-C(11)-C(12)	122.1(3)	C(4) = C(41) = C(46)	119.0(3)			
C(1) - C(11) - C(16)	118.8(3)	C(42) - C(41) - C(46)	118.7(4)			
C(12) - C(11) - C(16)	119.1(3)	C(41) - C(42) - C(43)	120.3(3)			
C(11) - C(12) - C(13)	120.7(3)	C(42) - C(43) - C(44)	120.2(4)			
C(12) - C(13) - C(14)	119.5(4)	C(43) - C(44) - C(45)	120.1(4)			
C(13) - C(14) - C(15)	120.2(3)	C(44) - C(45) - C(46)	120.1(4)			
C(14) - C(15) - C(16)	120.2(4)	C(41) - C(46) - C(45)	120.5(4)			
C(11) - C(16) - C(15)	120.3(3)					
C(21) - N(2) - C(2) - C(1)	15.0(5)	C(2) - C(1) - C(11) - C(12)	5.7(5)			
C(21) = N(2) = C(2) = C(3)	-172.6(3)	N(2) - C(2) - C(3) - C(4)	-7.0(5)			
O(1) - C(1) - C(2) - N(2)	-122.6(4)	C(1) - C(2) - C(3) - C(4)	165.6(3)			
O(1) - C(1) - C(2) - C(3)	64.5(4)	C(2) = C(3) = C(4) = O(4)	-0.4(5)			
C(11)-C(1)-C(2)-N(2)	62.0(4)	C(2) = C(3) = C(4) = C(41)	-176.8(3)			
C(11) - C(1) - C(2) - C(3)	-110.9(4)	O(4) = C(4) = C(41) = C(46)	-24.1(5)			
$\frac{O(1) - C(1) - C(11) - C(16)}{2}$	8.0(3)	· · · · · · · · · · · · · · · · · · ·				

 Table 3. Bond distances (Å), angles (°) and selected torsion angles (°) with esd's in parentheses

phenyl ring  $[O(1)-C(1)-C(2)-C(3) \ 64.5(4)^{\circ}]$ . The stretching associated with the hydrogen bond is probably also the cause of the distortion from the theoretical trigonal planar configuration observed in the bond angles around the ethylenic carbon atoms, which nevertheless retain a high  $sp^2$  character, the sum of bond angles being 359.6 and 359.4° around C(2) and C(3) respectively; the corresponding value in 1,4-diaryl-2-(arylamino)but-2-ene-1,4-diones (Paradisi *et al.*, 1981) is 354.0°.

The benzene rings are planar within the experimental error: the angles they form with the C=C double bond plane [71.7(1), 40.9(1) and 35.1(1)° for the aromatic ring containing C(11), C(21), and C(41) atom, respectively] are comparable with those found in the corresponding 1,4-diaryl-2-(arylamino)but-2-ene-1,4diones (Paradisi *et al.*, 1981): the greater variation of 9.1° observed in the benzene containing C(21) is probably caused by the different steric hindrance of this ring by the presence of the substituent C(27) methyl group.

Packing is consistent with van der Waals interactions.

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Anisotropic thermal parameters, H atom coordinates and structure factor data have been deposited with the British Library, Boston Spa, Wetherby, West Yorkshire, U.K., as supplementary publication No. 63156 (15 pages).