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SYNTHESIS OF 6-OXO-6,9-DIHYDRO[1,2,5]OXADIAZOLO[3,4-h]-QUINOLINE-7-CARBOXYLIC ACID N-OXIDE

Yusuf M. Al-Hiari,a Monther A. Khanfar,b Ali M. Qaisi,a Mohammad Y. Abu Shuheil,c Mustafa M. El-Abadelah,*b and Roland Boesed

a Pharmaceutical Sciences Department, Faculty of Pharmacy, University of Jordan, 11942 Amman, Jordan
b Chemistry Department, Faculty of Science, University of Jordan, Amman, Jordan
c Hikma Pharmaceuticals, P. O. Box 182400, 11118 Amman, Jordan
d Institut fuer Anorganische Chemie, Universitaet Duisburg-Essen, Campus Essen Universitaetstrasse 3-5, D-45117 Essen, Germany
* E-mail: mustelab@ju.edu.jo

Abstract –Ethyl 9-cyclopropyl-4-fluoro-6-oxo-6,9-dihydro[1,2,5]oxadiazolo[3,4-h]quinoline-7-carboxylate N(3)-oxide (3) is synthesized via pyrolysis of the 7-azido-8-nitroquinoline-3-carboxylate precursor (6). Acid-catalyzed hydrolysis of the product (3) afforded the corresponding carboxylic acid (4), of which structures are confirmed by spectroscopic means including X-Ray measurements.

INTRODUCTION
The second generation "fluoroquinolones" (1), represented by ciprofloxacin (1a),1 have become one of the most promising groups of antibacterial drugs. On the other hand, benzofuroxans (2,1,3-benzoxadiazole 1-oxides (2)) exhibit a wide spectrum of biological activity (fungicidal, bactericidal, parasiticidal and insecticidal)2 for which several references were cited in the patent literature.3 As synthons, benzofuroxans have been extensively utilized in the "Beirut" reaction4 for the direct preparation of many quinoxaline-, benzimidazole- and phenazine N-oxides, some of which possess high level of antimicrobial potency. Time and again, these properties stimulated interest in the search for new substituted fluoroquinolones, benzofuroxans and their congeners.
In the present study, we describe the synthesis and properties of the benzofuroxan derivatives sharing the same structural feature of the second generation fluoroquinolones, namely, ethyl 6-oxo-6,9-dihydro [1,2,5]oxadiazolo[3,4-h]quinoline-7-carboxylate N-oxide (3) and the corresponding acid (4). We expect these hybrid compounds encompassing the structural features of both benzofuroxan (rings B, C) and fluoroquinolone (rings A, B) to exhibit interesting bio-properties such as antimicrobial activity.

RESULTS AND DISCUSSION

Benzofuroxans (2) are conveniently prepared by pyrolysis of the appropriate o-nitrophenyl azides.\(^{5,6}\) We envisage that the target compound (4) can be prepared utilizing the known ethyl 7-chloro-1-cyclopropyl-6-fluoro-8-nitro-4-oxo-1,4-dihydroquinoline-3-carboxylate (5) as starting material. The required ester (5) was prepared according to a literature procedure\(^ {7}\) with some modifications as outlined in Scheme 1.

Scheme 1.  
(i) SOCl\(_2\), benzene, reflux, 3-4 h.  
(ii) Et\(_2\)O\(\rightarrow\)CH\(\rightarrow\)CH\(_2\)Me\(_2\), benzene, NET\(_3\), reflux, 2 h.  
(iii) cyclopropylamine, CH\(_2\)Cl\(_2\), MeOH.  
(iv) DMF, K\(_2\)CO\(_3\), 85°C, 1.5 h.
The 7-chloroester (5) thus prepared was treated with sodium azide in DMSO at rt to afford ethyl 7-azido-1-cyclopropyl-6-fluoro-8-nitro-4-oxo-1,4-dihydroquinoline-3-carboxylate (6) in high yield. Thermolysis of the azide (6) in p-xylene under reflux furnished the benzofuroxan (3) in 72 % yield as the single, fairly pure product (Scheme 2). The structure of 3 is confirmed by X-Ray determination (*vide infra*).

We next carried out the hydrolysis of the ester (3) under acidic conditions. Thus, treatment of 3 with hydrochloric acid in ethanol at reflux furnished the acid (4) in good yield without affecting the furoxan moiety. (Scheme 2).

Benzofuroxans usually exhibit rapid equilibrium between two N-oxide forms,8 in this case 3A and 3B, a process which is believed to proceed via the elusive 7,8-dinitrosouquinolone intermediate (3C). Of the two forms, 3A predominates as evidenced by X-Ray data (*vide infra*) indicating that tautomer (3A) is more stable, at least in the solid state. This is possibly due to smaller steric crowdedness around the N(3)-oxide oxygen in 3A as compared to 3B (Scheme 3).

**Scheme 2.** (i) NaN₃, Me₂SO, rt. (ii) p-xylene, 120–150 °C (oil bath), 2 h. (iii) aq. HCl, EtOH, 65-70 °C, ~ 18 h.

**Scheme 3**

**X-RAY STRUCTURE**

X-Ray crystal structure determination was performed to confirm the structure of the benzofuroxan (3). A summary of data collection and refinement parameters is given in Table 1 and pertinent distances and angles are given in Table 2. The molecular structure of 3, based on crystallographic data, is displayed in Figure 2.9
Table 1. Summary of the crystal data and structure refinement parameters for 3.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Empirical formula</td>
<td>C_{15}H_{12}F_{N_{3}}O_{5}</td>
</tr>
<tr>
<td>Formula weight</td>
<td>333.28 Da</td>
</tr>
<tr>
<td>Temperature</td>
<td>203(2) K</td>
</tr>
<tr>
<td>Crystal size</td>
<td>0.28 × 0.08 × 0.06 mm</td>
</tr>
<tr>
<td>Crystal system</td>
<td>Triclinic</td>
</tr>
<tr>
<td>Space group</td>
<td>P 1</td>
</tr>
<tr>
<td>Unit cell dimensions</td>
<td>\begin{align*} a &amp; = 6.9065(11) \text{ Å} \quad \alpha = 81.494(3)^\circ \ b &amp; = 9.5664(16) \text{ Å} \quad \beta = 86.656(3)^\circ \ c &amp; = 11.0724(18) \text{ Å} \quad \gamma = 79.370(3)^\circ \end{align*}</td>
</tr>
<tr>
<td>Volume</td>
<td>710.7(2) Å(^3)</td>
</tr>
<tr>
<td>Z</td>
<td>2</td>
</tr>
<tr>
<td>Density (calculated)</td>
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<tr>
<td>Absorption coefficient</td>
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<tr>
<td>F(000)</td>
<td>344</td>
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<tr>
<td>Theta range for data collection</td>
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<tr>
<td>Completeness to theta = 28.34°</td>
<td>99.3 %</td>
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<tr>
<td>Index ranges</td>
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<tr>
<td>Reflections collected</td>
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<tr>
<td>Max. / min. transmission</td>
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<tr>
<td>R(_\text{merge}) before / after correction</td>
<td>0.055 / 0.021</td>
</tr>
<tr>
<td>Independent reflections</td>
<td>3522 [R(_\text{int}) = 0.0244]</td>
</tr>
<tr>
<td>Cell measurement reflections used</td>
<td>4058</td>
</tr>
<tr>
<td>Weighting details</td>
<td>\begin{align*} w &amp; = 1 / \left[ \sigma^2 (F_0)^2 + (0.0661 P)^2 + 0.1732 P \right] \ \text{where } P &amp; = \left[ (F_0)^2 + 2(F_c)^2 \right] / 3 \end{align*}</td>
</tr>
<tr>
<td>Data / restraints / parameters</td>
<td>2568 / 0 / 217</td>
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<tr>
<td>Goodness-of-fit on F(^2)</td>
<td>1.060</td>
</tr>
<tr>
<td>Final R indices ([I &gt; 2 \sigma (I)])</td>
<td>R(_1) = 0.0459, wR(_2) = 0.1180</td>
</tr>
<tr>
<td>R indices (all data)</td>
<td>R(_1) = 0.0667, wR(_2) = 0.1307</td>
</tr>
<tr>
<td>Largest difference peak and hole</td>
<td>0.327 and -0.281 e. Å(^{-3})</td>
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Table 2. Selected bond lengths [Å] and angles [°] for 3

<table>
<thead>
<tr>
<th>Bond Lengths</th>
<th>Angles</th>
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<tr>
<td>N(1)–O(1) 1.220(2)</td>
<td>N(2)–O(2)–N(1) 109.44(12)</td>
</tr>
<tr>
<td>N(1)–C(6) 1.330(2)</td>
<td>O(1)–N(1)–C(6) 135.78(18)</td>
</tr>
<tr>
<td>N(1)–O(2) 1.458(2)</td>
<td>C(6)–N(1)–O(2) 105.09(14)</td>
</tr>
<tr>
<td>N(2)–C(5) 1.318(2)</td>
<td>C(5)–N(2)–O(2) 105.92(14)</td>
</tr>
<tr>
<td>N(2)–O(2) 1.371(2)</td>
<td>N(2)–C(5)–C(6) 111.58(14)</td>
</tr>
<tr>
<td>C(6)–C(7) 1.414(2)</td>
<td>N(2)–C(5)–C(4) 129.80(15)</td>
</tr>
<tr>
<td>C(5)–C(6) 1.415(2)</td>
<td>N(1)–C(6)–C(7) 128.94(17)</td>
</tr>
<tr>
<td>C(4)–C(5) 1.456(2)</td>
<td>N(1)–C(6)–C(5) 107.96(15)</td>
</tr>
<tr>
<td>F(1)–C(7) 1.344(2)</td>
<td>F(1)–C(7)–C(6) 118.78(14)</td>
</tr>
</tbody>
</table>

Figure 2. ORTEP plot (50%) of the molecular structure of 3
The X-Ray structure shows close intramolecular N2⋯C10 contact (2.834 Å), a situation which makes placement of oxygen at N2 (structure 3B) sterically less convenient; other contacts involve O1⋯F1 (2.988 Å) and O3⋯O4 (2.82 Å). Besides, intermolecular hydrogen bonding exists between O1, O2 of one molecule and C(H)14, C(H)15, respectively, of another molecule. Such extended intermolecular hydrogen bridging causes the molecules to adopt a planar chain form (Figure 3).

**ANTIMICROBIAL ACTIVITY**

*In vitro* antibacterial screening of 3 and 4 was performed and compared with the reference ciprofloxacin (1a). The results showed that both compounds (3) and (4) exhibit moderate antibacterial activity against *E. coli* (ATCC 8739) and *S. aureus* (ATCC 6538p), representatives of Gram-negative and Gram-positive bacteria classes, respectively (Table 3). For both strains, the free acid (4) showed higher potency as compared to its ester (3). Compared to *E. coli*, the acid (4) showed higher activity against *S. aureus*, with $MIC \approx 10 \mu g / mL$. However, both compounds showed feeble antifungal activity against *Candida albicans* (ATCC 10231) and *Aspergillus niger* (ATCC 16404).

<table>
<thead>
<tr>
<th>Compound No.</th>
<th>S. aureus ATCC 6538p MIC (µg / mL)</th>
<th>E. coli ATCC 8739 MIC (µg / mL)</th>
<th>C. albicans ATCC 10231 MIC (µg / mL)</th>
<th>A. niger ATCC 16404 MIC (µg / mL)</th>
</tr>
</thead>
<tbody>
<tr>
<td>3</td>
<td>38</td>
<td>38</td>
<td>&gt; 156</td>
<td>&gt; 156</td>
</tr>
<tr>
<td>4</td>
<td>10</td>
<td>20</td>
<td>&gt; 156</td>
<td>&gt; 156</td>
</tr>
<tr>
<td>Ciprofloxacin</td>
<td>1.23</td>
<td>0.30</td>
<td>-</td>
<td>-</td>
</tr>
</tbody>
</table>
CONCLUSION
A furoxano-fluoroquinolone carboxylic acid (4) and its ester (3) have been prepared and their antimicrobial activity has been evaluated. Both compounds exhibited moderate antibacterial activity.

EXPERIMENTAL
2,4-Dichloro-5-fluoro-3-nitrobenzoic acid, ethyl 3-(dimethylamino)acrylate and cyclopropylamine were purchased from Acros. Melting points (uncorrected) were determined on a Gallenkamp electrothermal melting temperature apparatus. $^1$H- and $^{13}$C-NMR spectra were measured on a Bruker DPX-300 instrument with Me$_4$Si as internal reference. EIMS spectra were obtained using a Finnigan MAT TSQ-70 spectrometer at 70 eV; ion source temperature = 200 °C. High resolution MS-ESI data were obtained with Bruker Bio TOF III. IR spectra were recorded as KBr discs on a Nicolet Impact-400 FT-IR spectrophotometer. Microanalyses were performed at the Microanalytical Laboratory – Medicinal Chemistry division, Faculty of Pharmacy, Jordan University, Amman, Jordan.

Ethyl 7-azido-1-cyclopropyl-6-fluoro-8-nitro-4-oxo-1,4-dihydroquinoline-3-carboxylate (6)
Sodium azide (7.8 g, 120 mmol) was added to a solution of 5 (7.1 g, 20 mmol) in dimethylsulfoxide (100 mL). The resulting mixture became turbid within few minutes, and was further stirred at rt for 6 – 8 h. Thereafter, the reaction mixture was diluted with cold water (250 mL), and the precipitated solid product was collected, washed with cold water, and dried. Yield of 6 = 6.3 g (87 %), mp 183 – 184 °C (decomp). Anal. Caled for C$_{15}$H$_{12}$N$_5$O$_5$F (361.28): C, 49.87; H, 3.35; N, 19.38. Found: C, 49.78; H, 3.14; N, 19.02; IR (KBr): ν 3095, 3054, 3002, 2955, 2901, 2148, 1729, 1632, 1605, 1548, 1393, 1311, 1239, 1173, 1148, 1060 cm$^{-1}$; EI MS m / z (%): 361(8, M$^+$), 333[24, (M-N$_2$)$^+$], 288(12), 282(25), 275(13), 261(100), 252(40), 245(17), 232(45), 226(31), 205(49), 200(47), 172(35), 147(21), 133(20), 120(9); HRMS: calcd. for C$_{15}$H$_{12}$N$_5$O$_5$F: 361.08220, found 361.07920; HRMS: calcd. for C$_{15}$H$_{12}$N$_5$O$_5$F (M-N$_2$): 333.07606, found 333.07469; $^1$H NMR (300 MHz, CDCl$_3$): δ 1.01, 1.12 (2m, 4H, H$_2$-2’ / H$_2$-3’), 1.37 (t, $^3$J$_{H-F}$ = 7.1 Hz, 3H, C$_3$H$_2$-), 3.57 (m, 1H, H-1’), 4.37 (q, $J = 7.1$ Hz, 2H, -CH$_2$-Me), 8.33 (d, $^3$J$_{H-F}$ = 11.2 Hz, 1H, H-5), 8.57 (s, 1H, H-2); $^{13}$C NMR (75 MHz, CDCl$_3$): δ 11.0 (C-2’ / C-3’), 14.4 (CH$_3$CH$_2$-), 37.7 (C-1’), 61.5 (CH$_2$Me), 112.1 (C-3), 115.8 (d, $^2$J$_{C-F}$ = 21.2 Hz, C-5), 127.0 (d, $^3$J$_{C-F}$ = 5.8 Hz, C-4a), 128.5 (d, $^2$J$_{C-F}$ = 16.6 Hz, C-7), 131.0 (d, $^4$J$_{C-F}$ = 2.4 Hz, C-8a), 133.1 (d, $^3$J$_{C-F}$ = 1.6 Hz, C-8), 151.4 (C-2), 152.9 (d, $^1$J$_{C-F}$ = 250 Hz, C-6), 164.3 (CO$_2$Et), 170.9 (d, $^4$J$_{C-F}$ = 2 Hz, C-4).

Ethyl 9-Cyclopropyl-4-fluoro-6-oxo-6,9-dihydro[1,2,5]oxadiazolo[3,4-h]quinoline-7-carboxylate N(3)-oxide (3)
A stirred suspension of ethyl 7-azido-1-cyclopropyl-6-fluoro-8-nitro-4-oxo-1,4-dihydroquinoline-3-
carboxylate (6) (1.45 g, 4 mmol) in *p*-xylene (70 mL) was heated at 80 – 85 °C for 1 h. Thereafter, the reaction temperature was brought to 110 – 115 °C for 1 h, during which time a steady evolution of nitrogen gas took place. Finally, the temperature was raised slowly to 140 – 145 °C and maintained thereat for 4 h. The solvent was then removed *in vacuo*, the residual solid product was soaked in hexane, collected by suction filtration, and triturated with cold acetone (2 × 3 mL). Yield = 0.96 g (72 %); mp 192 – 193 °C (decomp). For analytical purposes, the title product was recrystallized from hot ethanol (or from acetone) to afford yellow needles. *Anal.* Calcd for C₁₅H₁₂N₃O₅F (333.27): C, 54.06; H, 3.63; N, 12.61. Found: C, 53.84; H, 3.58; N, 12.36; IR (KBr): ν 3097, 3063, 2994, 2925, 2846, 1729, 1635, 1608, 1587, 1542, 1461, 1426, 1384, 1365, 1342, 1316, 1270, 1236, 1195, 1171, 1128, 1045 cm⁻¹; EI MS m/z (%): 333 (19, M +), 288 (6), 275 (11), 261 (100), 252 (29), 245 (19), 232 (20), 226 (24), 200 (21), 172 (23), 146 (12), 133 (13), 125 (6), 120 (4); MS (TOF ES⁺): m/z 356 (M+Na)⁺; HRMS: calcd. for C₁₅H₁₂N₃O₅FNa 356.0659, found 356.0653; ¹H NMR (300 MHz, DMSO-ᵈ₆): δ 1.25 (t, J = 7.1 Hz, 3H, CH₃CH₂), 1.25 (m, 4H, H₂-2/2′, overlapped with / underneath the Me triplet), 4.04 (m, 1H, H-1′), 4.20 (q, J = 7.1 Hz, 2H, MeCH₂O), 7.47, 7.48 (2d, J₆H-F = 10.5, 10.4 Hz, 1H, H-5), 8.46 (s, 1H, H-8); ¹³C NMR (75 MHz, DMSO-ᵈ₆): δ 10.4 (C-2/2′, C-3/3′), 41.6 (C-1′), 106.7 (d, 2J_C-F = 18.6 Hz, C-5), 108.6 (d, 2J_C-F = 21.2 Hz, C-3a), 115.4 (C-7), 129.3 (d, 3J_C-F = 4.2 Hz, C-5a), 131.0 (d, 4J_C-F = 3.4 Hz, C-9a), 144.0 (d, 1J_C-F = 258 Hz, C-4), 148.0 (C-8), 149.3 (C-9b), 164.1 (CO₂Et), 170.3 (d, 4J_C-F = 2.0 Hz, C-6).

9-Cyclopropyl-4-fluoro-6-oxo-6,9-dihydro[1,2,5]oxadiazolo[3,4-h]quinoline-7-carboxylic acid N(3)-oxide (4)

A vigorously stirred suspension of 3 (1.0 g, 3 mmol) in 12 % aqueous HCl (50 mL) and ethanol (20 mL) was heated at 65-70 °C. Progress of the ester hydrolysis was monitored by TLC and the reaction completed within 16-18 h. Thereafter, the reaction mixture was cooled, the resulting heavy faint yellow precipitate was collected, washed with cold water (2 × 20 mL), dried, and recrystallized from acetone. Yield = 0.76 g (83 %), mp 239 – 240 °C (decomp). *Anal.* Calcd for C₁₃H₇N₃O₅F (305.22): C, 51.16; H, 2.64; N, 13.77. Found: C, 50.94; H, 2.61; N, 13.62; IR (KBr): ν 3435, 3228, 3120, 3081, 3007, 1723 (br), 1634, 1593, 1537, 1506, 1447, 1411, 1389, 1347, 1320, 1294, 1241, 1220, 1193, 1158, 1123, 1081, 1044, 1023 cm⁻¹; EI MS m/z (%) : 305 (17, M +), 289 (2), 261 (100), 245 (30), 232 (11), 200 (13), 172 (19), 146 (14), 133 (6), 125 (5), 120 (10); MS (TOF ES⁺): m/z 306 (M+H)⁺, HRMS calcd. for C₁₃H₈N₃O₅F 306.0526, found 306.0521; m/z 328 (M + Na)⁺, HRMS calcd. for C₁₃H₈N₃O₅FNa 328.0346, found 328.0340; ¹H NMR (300 MHz, DMSO-ᵈ₆): δ 1.29, 1.35 (2m, 4H, H₂-2/2′), 4.19 (m, 1H, H-1′), 7.65 (d, 3J_H-F = 10 Hz, 1H, H-5), 8.78 (s, 1H, H-8), 14.72 (br s, 1H, CO₂H); ¹³C NMR (75 MHz, DMSO-ᵈ₆): δ 10.4 (C-2/2′, C-3′).
(C-7), 127.6 (d, $^3J_{C-F} = 5.6$ Hz, C-5a), 132.6 (d, $^4J_{C-F} = 3.2$ Hz, C-9a), 145.1 (d, $^1J_{C-F} = 260$ Hz, C-4), 148.8 (d, $^3J_{C-F} = 1.5$ Hz, C-9b), 148.9 (C-8), 165.2 (CO$_2$H), 175.4 (d, $^4J_{C-F} = 2.4$ Hz, C-6).

**PHARMACOLOGICAL TESTS**

The minimal inhibitory concentrations (MICs) were determined by the conventional broth dilution method using the two-serial dilution technique. The standardization of bacterial test suspension was carried out according to McFarland standard method as described by the National Committee for Clinical Laboratories Standard (NCCLS, 1993). Stock solutions of the test compounds were prepared using DMSO. Serial dilutions were prepared to obtain test concentrations ranging from 156 $\mu$g/mL - 0.3 $\mu$g/mL. Each tube was then inoculated with 0.1 mL of the cultured bacteria (containing approximately 1 to 2 x 10$^8$ CFU/mL), mixed and incubated at 37°C for 24 h. Growth inhibition with concentrations at 156 $\mu$g/mL or lower were carried out in duplicates. All test tubes showing positive/negative growth were confirmed by the agar plate method. The results were recorded according to presence and absence of growth. The MICs were calculated as the average concentration of the test agent in the broth tubes showing consecutive positive and negative growth.

**COLLECTION OF X-RAY DIFFRACTION DATA AND STRUCTURE ANALYSIS OF 3**

Yellow needles were grown by allowing a clear solution of 3 in hot ethanol to stand at room temperature for 4 – 5 days. Crystal data collection was made with a Siemens SMART CCD diffractometer [Mo-K$\alpha$-radiation, graphite monochromator] operating in the omega scan mode (0.3°). The data were reduced with the Siemens-Bruker program suite XSCANS$^{10}$ and the structure was solved by the direct method using SHELXTL PLUS programs.$^{11}$ All non-hydrogen atoms were refined anisotropically by full-matrix, least-squares procedure based on $F^2$ using all unique data. Hydrogen atoms were placed in calculated positions and treated as riding groups, with the 1.2 fold (1.5 fold for methyl groups) isotropic displacement parameters of the equivalent Uij of the corresponding carbon atom.

**SUPPLEMENTARY MATERIAL**

Crystallographic data for the structural analysis of 3 have been deposited with the Cambridge Crystallographic Data Center under the depository No. CSD-295 564. Copies of information may be obtained free of charge from the Director, CCDC, 12 Union Road, Cambridge CB2 1EZ, UK (Fax: +44-1223-336033; e-mail: (deposit@ccdc.cam.ac.uk or http://www.ccdc.cam.ac.uk).

**ACKNOWLEDGEMENTS**

We wish to thank the Deanship of Scientific Research at the University of Jordan, Amman- Jordan for financial support.
REFERENCES AND NOTES


9. The numbering of the skeletal atoms in this ORTEP plot (and in Tables 1 and 2) is arbitrary, being different from the IUPAC numbering adopted in Scheme 2.

10. Bruker AXS SADABS program multiscan V2.03.

11. G. M. Sheldrick, SHELXTL PLUS System of computer programs for the determination of crystal structure from the X-Ray diffraction data, Vers.5.10.DOS/WIN 95/NT.