FACILE SYNTHESIS AND HERBICIDAL ACTIVITIES OF NEW ISOXAZOLE DERIVATIVES VIA 1,3-DIPOLAR CYCLOADDITION REACTION

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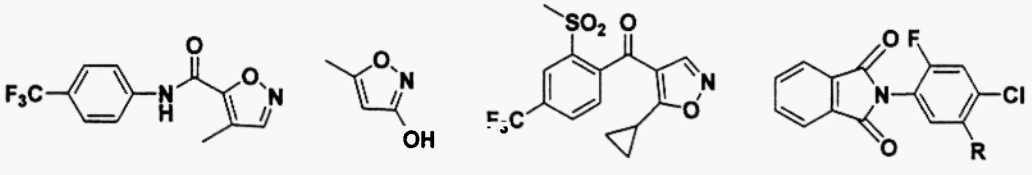
ABSTRACT: A series of cycloadducts *via* 1,3-dipolar cycloaddition reactions of generated nitrile oxides with *N*-(4-chloro-2-fluorophenyl)maleimides were described. The reaction of *N*-(4-chloro-2-fluorophenyl)maleimides with nitrile oxides gave 3,5-diaryl-3a,6a-dihydro-pyrrolo[3,4-d]isoxazole-4,6-diones through syn-addition pattern. The title compounds were characterized by ^{1}HNMR, IR, MS, and elemental analysis or HRMS. The single crystal structure of **6i** was determined by X-ray diffraction. The herbicidal activities of the title compounds were evaluated. Some of them exhibited certain herbicidal activities against barnyardgrass and rape.

Keywords: 1,3-dipolar cycloaddition; synthesis; isoxazole derivatives, nitrile oxide, crystal strucutre, herbicidal activity.

1. INTRODUCTION

Nitrogen linked heterocyclic compounds received considerable attention in recent times because of their medicinal and pesticidal importance. [1-3] Compounds containing isoxazole ring have been claimed to have beneficial medicinal and agricultural applications, [4-9] such as leflunomide, [10] hymexazol, [11] isoxaflutole [12](Figure **1**). Furthermore, it was also found that the *o*-phthalimide nucleus, which incorporates an isoxazole ring, exhibits a large number of biological activities, especially herbicidal activities [13]. Additionally, many biological compounds contain a fluoro moiety, which indicats that this moiety may be important for biological activity. [14]



leflunomide **hymexazol** **isoxaflutole** **N-arylphthalimide**

Fig. 1 Structures of biological activity compounds

Several procedures are available for the synthesis of isoxazole derivatives. 1,3-Dipolar

cycloaddition are the frequently-used method in synthesis of heterocycles. Reactions of nitrile oxides with alkenes leading to isoxazoles are a long-known and a well-studied area.[15]

In continuation of our research on the synthesis of biologically active heterocycles, [16-19] a series of new isoxazole derivatives (Figure 2) were synthesized and their herbicidal activities were tested.

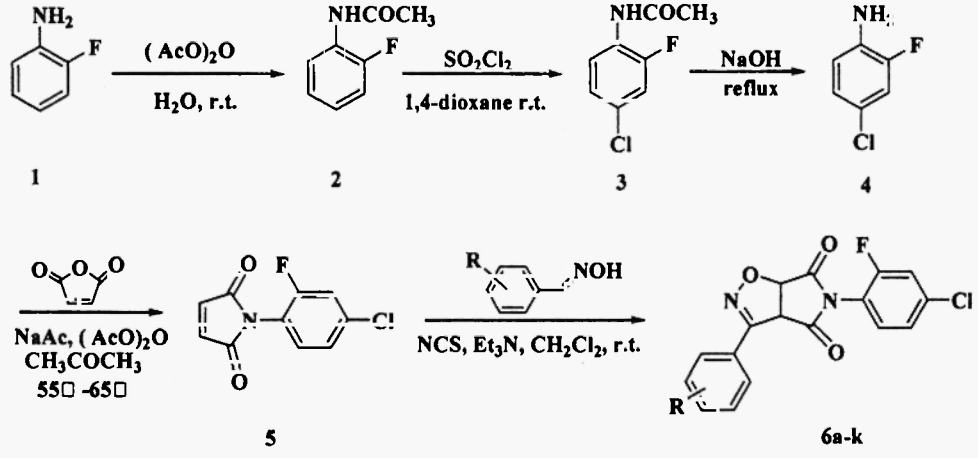
2. EXPREMENTAL

2.1 Instruments

Melting points were determined using an X-4 apparatus and were uncorrected. Infrared spectra were recorded on a Bruker Equinox55 spectrophotometer as potassium bromide tablets. ^{1}H NMR spectra were measured on a Bruker AC-P500 instrument (300MHz) or Varian instrument (400MHz) using tetramethylsilane as an internal standard and deuterochloroform as solvent. Mass spectra were recorded on a Thermo Finnigan LCQ Advantage LC/mass detector instrument. HRMS data was obtained on a FTICR-MS instrument (Ionspec 7.0T). Elemental analysis was performed on a Yanaco CHN Corder MF-3 automatic elemental analyser. Crystallographic data of the compound were collected on a Bruker SMART 1000 CCD diffractometer.

2.2 Synthesis of compounds

The route shown in Fig. 2 was used for the synthesis of the title compounds. The yields were not optimized. All the intermediates substituted benzaldehyde oximes were synthesized according to the reference.[20]



R=H, *o*-Cl, *p*-Cl, *o*-Br, *p*-Br, *o*-F, *p*-CH_3, *o*-OCH_3, *m*-OCH_3, *p*-OCH_3, 3,4-$(CH_3)_2$

Fig 2 Synthetic route of compounds **6a-k**

N-(4-chloro-2-fluorophenyl)maleimides (**5**).[21] Compound **5** was prepared with two step method. The first step: to a stirred solution of 2.25g (23mmol) of maleic anhydride in 20ml of acetone was added a solution of 3.34g (23mmol) of 4-chloro-2-fluoroaniline in 20ml of acetone dropwise over a period of 30 min and the stirring was continued for further 2 h. A solid was obtained in 77% yield. The second step: sodium acetate 2.94g (36mmol) and acetic anhydride 3.66g (36mmol) were added successively at room temperature to a stirred solution of a solid which obtained above procedure in acetone (80ml). Upon completion of addition

the reaction mixture was heated to 55-65□ under reflux for 5 h. The reaction mixture was cooled to room temperature and acetone was removed on a rotary evaporator to obtain a solid which washed with water, extracted with dichloromethane, dried over anhydrous sodium sulfate, and the solvent was removed under reduced pressure to give 7.13g of compound **5** (yield 88.4%).

2-fluoroacetaniline (**2**). To a stirred solution of 11.1g (100mmol) 2-fluoroaniline in 20 ml of water was added 12.3g (120mol) of acetic anhydride. Upon completion of addition the reaction mixture stirred at room temperature for 2 h was filtered to collect a solid. The solid was dried to give 12.1g of compound **2** (yield 78.9%).

4-chloro-2-fluoroacetaniline(**3**). To a stirred solution of 11g (72mmol) of 2-fluoroacetaniline in 20ml of 1,4-dioxane was slowly added dropwise 11.6g (86mmol) of sulfuryl chloride. Upon completion of addition the reaction mixture stirred at ambient temperature for 24 h. A solid was collected by filtration, washed with water, and dried to give 9.3g of compound **3** (yield 68.9%).

4-chloro-2-fluoroaniline (**4**).[22] To a stirred solution of 9.3g (53mmol) of compound 3 in 40 ml of ethanol was added dropwise a solution of 4.5g (110mmol) of sodium hydroxide in 15 ml of water. Upon completion of addition the reaction mixture was heated under reflux for 3 h. The reaction mixture was cooled to room temperature and extracted with diethyl ether. The diethyl ether solution was dried (Na_2SO_4) and concentrated under reduced pressure to a residual oil. The oil was distilled under reduced pressure to give 6.93g of compound **4** (b.p.83-85□/12mmHg, yield 95.6%).

5-(4-chloro-2-fluorophenyl)-3-phenyl-3a,6a-dihydro-pyrrolo[3,4-d]isoxazole-4,6-dione (**6a**).[23-24] General procedure: benzaldoxime (0.55g, 4.56mmol), *N*-(4-chloro-2-fluorophenyl)maleimide (0.85g, 3.8mmol) and *N*-chloro succinimide (0.53g, 4.0mmol) were taken in dry dichloromethane (15ml) and stirred magnetically at room temperature. To this well stirred reaction mixture was added dry triethylamine (0.58g, 5.7mmol) in dichloromethane (5ml) dropwise over a period of 30min and the stirring was continued for further 24h. Generation of nitrile oxide was indicated by the precipitation of triethylamine hydrochloride salt which was filtered off. The filtrate thus obtained was concentrated and the residue was purified by silica-gel column eluted with ethyl acetate/petroleum ether (volume ratio1:5) to obtain the white product **6a** in 39.6% yield. m. p 147-149□; ^{1}HNMR ($CDCl_3$, 300MHz), δ: 5.04 (d, J=9.6Hz, CHCO, 1H), 5.72 (d, J=9.6Hz, OCHCO, 1H), 7.17-7.26 (m, ArH, 3H), 7.47-7.49 (m, ArH, 3H) 8.01 (d, J=9.6Hz, ArH, 2H). MS (ESI), m/z: 343 $[M-H]^-$. FTMS (ESI) $[M-H+CH_3OH]^-$ ($C_{17}H_{10}ClFN_2O_3$): Calcd 375.0553, Found 375.0559. The other cycloadducts **6b-k** were obtained similarly following the above procedure.

5-(4-chloro-2-fluorophenyl)-3-(4-chlorophenyl)-3a,6a-dihydro-pyrrolo[3,4-d]isoxazole-4,6-di one (**6b**). The compound was obtained in 41.2% yield as a white crystal; m. p 154-155□; ^{1}HNMR ($CDCl_3$, 300MHz), δ: 5.53 (d, J=9.3Hz, CHCO, 1H), 5.85 (d, J=9.3Hz, OCHCO, 1H), 7.47-7.48 (m, ArH, 2H), 7.60 (d, J=8.7Hz, ArH, 2H), 7.69 (d, J=9.3Hz, ArH, 1H), 7.95 (d, J=8.4Hz, ArH, 2H). MS (ESI), m/z: 377 $[M-H]^-$. FTMS (ESI) $[M-H+CH_3OH]^-$ ($C_{17}H_9Cl_2FN_2O_3$): Calcd 409.0164, Found 409.0162.

5-(4-chloro-2-fluorophenyl)-3-(2-chlorophenyl)-3a,6a-dihydro-pyrrolo[3,4-d]isoxazole-4,6-di one (**6c**). The compound was obtained in 40.5% yield as a white crystal; m. p 157-159□; ^{1}HNMR ($CDCl_3$, 400MHz), δ: 5.39 (d, J=9.6Hz, CHCO, 1H), 5.74 (d, J=9.6Hz, OCHCO, 1H), 7.16-7.25 (m, ArH, 3H), 7.34-7.38 (m, ArH, 1H), 7.43-7.46 (m, ArH, 1H), 7.51 (d, J=7.6Hz, ArH, 2H). MS (ESI), m/z: 377 $[M-H]^-$. FTMS (ESI) $[M-H+CH_3OH]^-$ ($C_{17}H_9Cl_2FN_2O_3$): Calcd 409.0164, Found 409.0162.

3-(4-bromophenyl)-5-(4-chloro-2-fluorophenyl)-3a,6a-dihydro-pyrrolo[3,4-d]isoxazole-4,6-di one (**6d**). The compound was obtained in 42.3% yield as a white crystal; m. p 120-122□; ^{1}HNMR ($CDCl_3$, 400MHz), δ: 5.38 (d, J=9.6Hz, CHCO, 1H), 5.74 (d, J=9.6Hz, OCHCO, 1H), 7.15-7.25 (m, ArH, 2H), 7.32-7.42 (m, ArH, 4H), 7.69 (d, J=8.0Hz, ArH, 1H). MS (ESI),

m/z: 421 $[M-H]^-$. Elemental Anal. (%), Calcd: C, 48.20; H, 2.14; N 6.61; Found: C, 48.26, H, 2.28, N, 6.72.

3-(2-bromophenyl)-5-(4-chloro-2-fluorophenyl)-3a,6a-dihydro-pyrrolo[3,4-d]isoxazole-4,6-di one (**6e**). The compound was obtained in 35.7% yield as a white crystal; m. p 132-134□; ^{1}HNMR ($CDCl_3$, 300MHz), δ: 5.01 (d, J=9.9Hz, CHCO, 1H), 5.74 (d, J=9.9Hz, OCHCO, 1H), 7.20-7.28 (m, ArH, 3H), 7.63 (d, J=8.4Hz, ArH, 2H), 7.91 (d, J=8.4Hz, ArH, 2H). MS (ESI), m/z: 421 $[M-H]^-$. Elemental Anal. (%), Calcd: C, 48.20; H, 2.14; N 6.61; Found: C, 47.92, H, 2.26, N, 6.71.

5-(4-chloro-2-fluorophenyl)-3-(2-fluorophenyl)-3a,6a-dihydro-pyrrolo[3,4-d]isoxazole-4,6-di one (**6f**). The compound was obtained in 34.8% yield as a white crystal; m. p 147-149□; ^{1}HNMR ($CDCl_3$, 400MHz), δ: 5.24 (d, J=8.8Hz, CHCO, 1H), 5.70 (d, J=8.8Hz, OCHCO, 1H), 7.16-7.22 (m, ArH, 5H), 7.47-7.52 (m, ArH, 1H), 7.81-7.85 (m, ArH, 1H). MS (ESI), m/z: 361 $[M-H]^-$. FTMS (ESI) $[M-H+CH_3OH]^-$ ($C_{17}H_9ClF_2N_2O_3$): Calcd 393.0459, Found 393.0459.

5-(4-chloro-2-fluorophenyl)-3-(2-methoxyphenyl)-3a,6a-dihydro-pyrrolo[3,4-d]isoxazole-4,6-dione (**6g**). The compound was obtained in 45.2% yield as a white crystal; m. p 170-172□; ^{1}HNMR ($CDCl_3$, 300MHz), δ: 3.87 (s, OCH_3, 3H) 5.39 (d, J=8.7Hz, CHCO, 1H), 5.81 (d, J=8.7Hz, OCHCO, 1H), 7.03 (t,ArH, J=7.5Hz, 1H), 7.19 (d, J=8.4Hz, ArH, 1H), 7.41-7.51 (m, ArH, 4H), 7.69(d, J=9.6Hz, ArH ,1H). MS (ESI), m/z: 373 $[M-H]^-$. FTMS (ESI) $[M-H+CH_3OH]^-$ ($C_{18}H_{12}ClFN_2O_4$): Calcd 405.0659, Found 405.0656.

5-(4-chloro-2-fluorophenyl)-3-(4-methoxyphenyl)-3a,6a-dihydro-pyrrolo[3,4-d]isoxazole-4,6-dione (**6h**). The compound was obtained in 48.6% yield as a white crystal; m. p 193-195□; ^{1}HNMR ($CDCl_3$, 300MHz), δ: 3.82 (s, OCH_3, 3H) 5.46 (d, J=9.0Hz, CHCO, 1H), 5.76 (d, J=9.0 Hz, OCHCO, 1H), 7.07(d, J=8.7Hz, ArH, 2H) , 7.43-7.48 (m, ArH, 2H), 7.68-7.71 (m, ArH, 1H), 7.87 (d, J=8.7Hz, ArH ,2H). MS (ESI), m/z: 373 $[M-H]^-$. FTMS (ESI) $[M-H+CH_3OH]^-$ ($C_{18}H_{12}ClFN_2O_4$): Calcd 405.0659, Found 405.0659.

5-(4-chloro-2-fluorophenyl)-3-(3-methoxyphenyl)-3a,6a-dihydro-pyrrolo[3,4-d]isoxazole-4,6-dione (**6i**). The compound was obtained in 47.5% yield as a white crystal; m. p 140-141□; ^{1}HNMR ($CDCl_3$, 400MHz), δ: 3.85 (s, OCH_3, 3H) 5.01 (d, J=8.8Hz, CHCO, 1H), 5.71 (d, J=8.8 Hz, OCHCO, 1H), 7.02-7.05(m, ArH, 1H) , 7.16-7.25 (m, ArH, 3H), 7.35-7.39 (m, ArH, 1H), 7.55-7.59 (m, ArH ,2H) MS (ESI), m/z: 373 $[M-H]^-$ FTMS (ESI) $[M-H+CH_3OH]^-$ ($C_{18}H_{12}ClFN_2O_4$): Calcd 405.0659, Found 405.0657.

5-(4-chloro-2-fluorophenyl)-3-p-tolyl-3a,6a-dihydro-pyrrolo[3,4-d]isoxazole-4,6-dione (**6j**). The compound was obtained in 41.3% yield as a white crystal; m. p 79-81□; ^{1}HNMR ($CDCl_3$, 400MHz), δ: 2.39 (s, $ArCH_3$, 3H), 5.01 (d, J=9.6Hz, CHCO, 1H), 5.69 (d, J=9.6Hz, OCHCO, 1H), 7.23-7.25(m, ArH, 3H) , 7.26-7.28 (m, ArH, 2H), 7.89 (d, J=8.4Hz, ArH ,2H). MS (ESI), m/z: 357 $[M-H]^-$. Elemental Anal. (%), Calcd: C, 60.26; H, 3.37; N 7.81; Found: 59.98; H, 3.57; N, 7.56.

5-(4-chloro-2-fluorophenyl)-3-(3,4-dimethylphenyl)-3a,6a-dihydro-pyrrolo[3,4-d]isoxazole-4, 6-dione (**6k**). The compound was obtained in 35.2% yield as a white crystal; m. p 72-74□; ^{1}HNMR ($CDCl_3$, 300MHz), δ: 2.24 (s, $ArCH_3$, 6H), 4.93 (d, J=9.9Hz, CHCO, 1H), 5.61 (d, J=9.9Hz, OCHCO, 1H), 7.14-7.17(m, ArH, 4H) , 7.64-7.71 (m, ArH, 3H). MS (ESI), m/z: 371 $[M-H]^-$. FTMS (ESI) $[M-H+CH_3OH]^-$ ($C_{19}H_{14}ClFN_2O_3$): Calcd 403.0866, Found 403.0867.

2.3 The herbicidal activity tests

2.3.1 Inhibition of the Root-growth of Rape (*Brassica campestris*)

The compounds to be tested were made into an emulsion to aid dissolution. Rape seeds were soaked in distilled water for 4 hours before being placed on a filter paper in a 6-cm Petri plate, to which 2 ml of inhibitor solution had been added in advance. Usually, 15 seeds were used on each plate. The plate was placed in a dark room and allowed to germinate for 65 hours at 28 ± 1 °C. The lengths of 10 rape roots selected from each plate were measured and the means were calculated. The check test was carried out in distilled water only. The percentage of the inhibition was calculated.

2.3.2 Inhibition of the Seedling-growth of Barnyardgrass (*Echinochloa crusgalli*)

The compounds evaluated were made into an emulsion to aid dissolution. 10 barnyardgrass seeds were placed into a 50 ml cup covered with a layer of glass beads and a piece of filter paper at the bottom, to which 5 ml of inhibitor solution had been added in advance. The cup was placed in a bright room and allowed to germinate for 65 hours at 28 ± 1 °C. The heights of seedlings of above-ground plant parts from each cup were measured and the means were calculated. The check test was carried out in distilled water only. The percentage of the inhibition was calculated.

TABLE 1 Herbicidal Activity Data of Compounds (% inhibition)

Compound	R	Rape Root Test		Barnyardgrass Cup Test	
		10μg mL^{-1}	100μg mL^{-1}	10μg mL^{-1}	100μg mL^{-1}
6a	H	0	10.3	15.6	20.5
6b	*p*-Cl	0	23.8	0	1.5
6c	*o*-Cl	0	0	0	28.5
6d	*p*-Br	0	0	0	0
6e	*o*-Br	0	1.0	0	10.3
6f	*o*-F	0	7.8	6.5	24.0
6g	*o*-OCH_3	0	4.1	1.5	21.7
6h	*p*-OCH_3	0	0	10.6	37.1
6i	*m*-OCH_3	0	10.0	10.3	30.0
6j	*p*-CH_3	0	0	0	0
6k	3,4-$(CH_3)_2$	0	12.8	9.9	39.9

2.4 Crystal Structure determination

The crystal of compound **6i** with dimensions of 0.14mm × 0.10mm × 0.06mm was mounted on a Bruker SMART[25] 1000 CCD area-detector diffractometer with a graphite-monochromated MoKα radiation (λ = 0.71073Å) by using a phi and scan modes at 113(2) K in the range of $2.28° \leq \theta \leq 27.9°$. The crystal belongs to Monoclinic system with space group P2_1/c and crystal parameters of a= 15.940(3) Å, b = 10.991(2) Å, c = 9.4356(19) Å, α = 90°, β = 104.72(3)°, γ = 90 °. V= 1598.8(6) A^3 , Dc= 1.557 mg/m^3. The absorption coefficient μ=0.2800mm^{-1}, and Z=4. The structure was solved by direct methods with SHELXS-97 [26-27] and refined by the full-matrix least squares method on F^2 data using

SHELXL-97. The final full-matrix least squares refinement gave R = 0.042000 and wR =0.105000 ($w = 1/[\sigma^2(F_o^2) + (0.0562P)^2]$ where $P = (F_o^2 + 2F_c^2)/3$, S = 1.04, $(\Delta/\sigma)_{max}$ <0.001000, $\Delta\rho_{max}$ =0.300 e Å^3 and $\Delta\rho_{min}$ = -0.25 e Å^3.

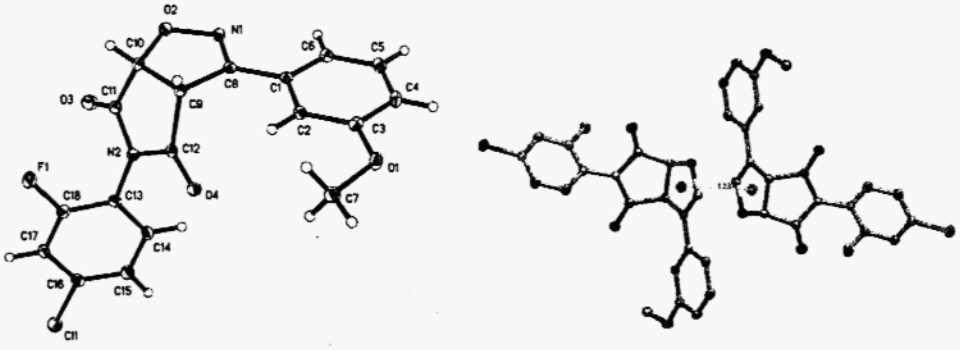
3. RESULTS AND DISCUSSION

3.1 Synthesis

Treatment of aldoximes with NCS in dichloromethane at room temperature gave the corresponding hydroximoyl chlorides which reacted with *N*-(4-chloro-2-fluorophenyl)maleimides as dipolarophiles in the presence of triethylamine to give the title compounds. In our experiments, we found that electron-donating groups or electron-withdrawing groups on the aromatic ring will affect the yields of the title compounds. The yield of the compounds with electron-donating groups was relatively higher than others while the presence of a electron-withdrawing group on the aromatic ring will not give product effectively. In the case of the strong electron-withdrawing NO_2, the target cycloadduct was not obtained at all.

3.2 Crystal Structure

The structure of compound **6i** was further confirmed by single crystal X-ray diffraction analysis (Fig.**3a**). In the crystal structure, we can see that isoxazoline ring and benzene ring are coplanar in the molecular structure, but the angel between the isoxazoline ring and maleimide ring is 108.97°. From the compound **6i** crystal structure, we also can see that two hydrogens of the bridge head are in the same side of the ring, thus it may be known, 1,3-dipolar cycloaddition reaction is according to carrying on the same surface addition(syn-addition pattern). There is intermolecular π-π interaction between neighbouring isoxazole rings (C8-C10, N1, O2), as shown in Fig. **3b**. The two rings parallel to each other with a dihedral angle of 0°. The centroid distance is 3.237 Å. All these structural parameters strongly indicate the existence of intermolecular π-π interaction.



(a) **(b)**

Fig 3 The crystal structure of compound **6i**

3.3 Herbicidal Activities

The herbicidal activities of the title compounds were determined with roots of rape, and the barnyardgrass cup test. The results are shown in Table **1**. Most of the compounds showed weak herbicidal activities. Compounds **6c, 6h,** and **6k** showed higher inhibition abilities of bamyardgrss root at 100μg/mL than those of rape tests.

Supplementary material

CCDC 686873 contains the supplementary crystallographic data for this paper. These data can be obtained free of charge at www.ccdc.cam.ac.uk/conts/ retrieving.html or from the Cambridge CrystallographicData Centre (CCDC), 12 Union Road, Cambridge CB2 1EZ, UK (Fax: +44-1223-336033; email: deposit@ccdc.cam.ac.uk or www: http://www.ccdc.cam.ac.uk).

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