Microwave-Assisted Three-Component Synthesis of Some Novel 1-Alkyl-1*H*-indole-2,3-dione 3-(*O*-Alkyloxime) Derivatives as Potential Chemotherapeutic Agents

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A convenient and efficient method for a one-pot conversion of *N*-alkylisatins to *N*-alkylisatin *O*-alkyloximes **7a**–**7n** as potential chemotherapeutic agents is described (*Scheme*) (isatin = 1*H*-indole-2,3-dione). In this method, the microwave-assisted three-component reaction of *N*-alkylisatins **8**, hydroxylamine hydrochloride, and diverse alkyl halides in the presence of K_2CO_3 and Bu_4NBr furnishes the corresponding *N*-alkylisatin *O*-alkyloximes under solvent-free condition in short times (2–10 min) and good to excellent yields (62–83%). The *O*-alkylation of *in situ* generated isatin oximes with alkyl halides was achieved regioselectively, and (*Z*)-*O*-alkyloximes were produced dominantly. PM3 Semi-empirical quantum-mechanic calculations were performed to rationalize the evidences, and the calculations indicated a lower heat of formation for the (*Z*)-*O*-alkyloximes.

Introduction. – Isatin (= 1*H*-indole-2,3-dione) is a versatile substrate in organic and medicinal chemistry, and it has been used extensively for the construction of various compounds exhibiting a wide spectrum of biological activities [1]. Among isatin derivatives, oxime and *O*-alkyloximes of isatin have attracted particular attention because of their broad range of applications as potential therapeutic agents and synthetic intermediates [1a–1c][2]. Isatin oximes and *O*-alkyloximes display diverse pharmacological activities such as antiviral [3], anticancer [4], anticonvulsant [5], anti-HIV [6], anti-inflammatory, as well as anti-asthmatic [7], anti-mycobacterium tuberculosis [8], and antidote activity [9]. Moreover, coumarin derivatives of isatin oximes are used in medicine as fluorescent agents [10]. The structures of several famous isatin oximes and *O*-alkyloximes that are used in medicinal chemistry, *i.e.*, of **1**–**6**, are shown in the *Figure*.

Two general methods have been established for the synthesis of isatin *O*-alkyloximes: first, the reaction of isatin with *O*-alkylhydroxylamines [3b][4d][11] and second, the reaction of isatin oxime with alkyl halides under various conditions [3a][12]. However, these methods are accompanied by several drawbacks such as the nonavailability of structurally diverse *O*-alkylhydroxylamines, the use of DMF and DMSO as solvent, tedious workup of the reaction mixture, long reaction times, and low yields. Moreover, conventional heating methods for the synthesis of *O*-alkyloximes

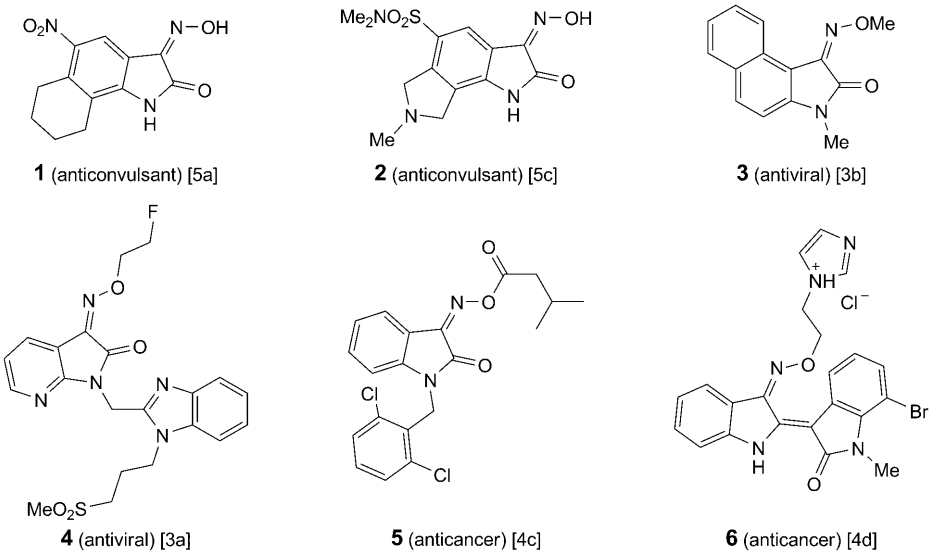


Figure. *Structures of several isatin oximes and* O*-alkyloximes exhibiting biological activities*

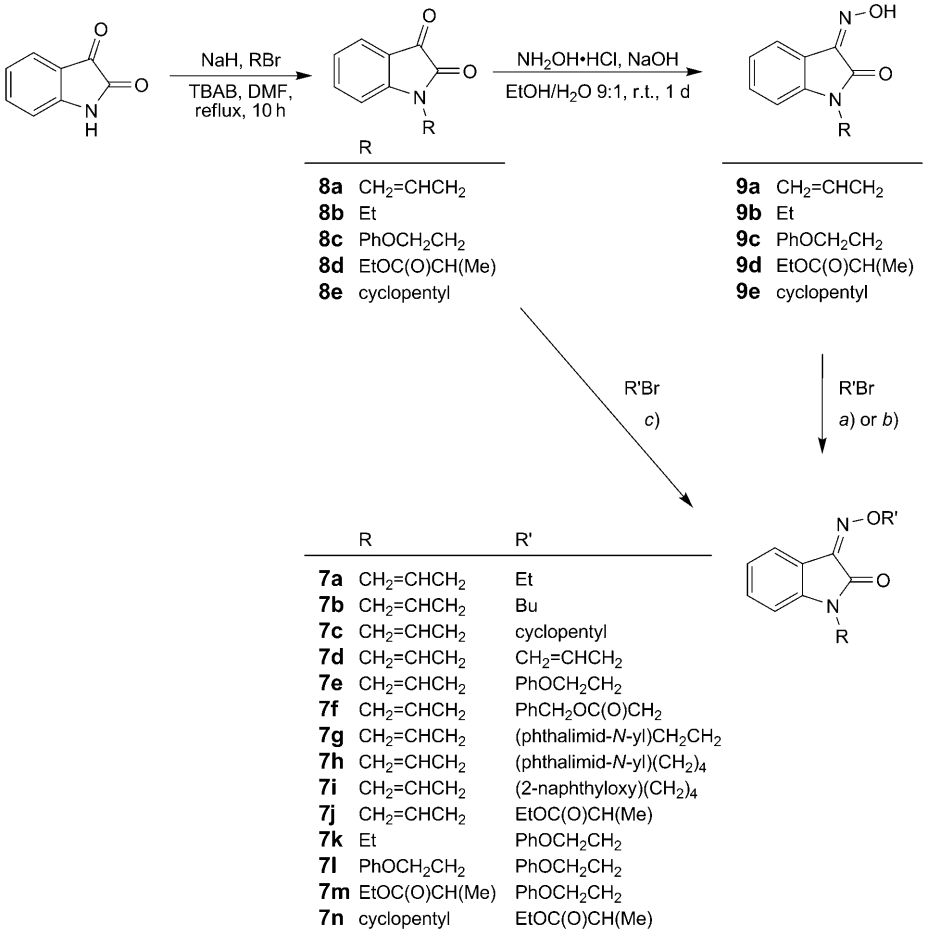
usually give a mixture of the two geometrical isomers whose separation is cumbersome and results in a decrease of the overall yield [13].

The multicomponent reactions (MCRs) are most important reactions in organic synthesis and medicinal chemistry [14]. The MCRs are one-pot processes combining three or more substrates simultaneously. Due to their ease of operation, atom economy, and quantitative yields, MCRs have found growing attention. Many important reactions such as the *Biginelli*, *Mannich*, *Passerini*, and *Ugi* reactions are well-known multicomponent reactions [14]. These reactions often involve condensation processes and ring closure. Indeed, there are only rare reports in which an S_N2 type reaction is a part of MCRs [15].

Microwave-assisted synthesis in organic reactions has been of growing interest as an efficient, economic, and clean procedure. Microwave irradiation often leads to a remarkable decrease in reaction time, increased yields, easier workup, compliance with environmentally benign chemistry, and may enhance the regio- and stereoselectivity of reactions [16].

Encouraged by the unique biological activities of isatin oximes and *O*-alkyloxime derivatives and also in continuation of our ongoing research in oxime chemistry [17] as well as multicomponent reactions [15], we report a convenient and simple method for the synthesis of some novel *N*-alkylated isatin *O*-alkyloximes **7a**–**7n** using microwave-assisted three-component coupling of *N*-alkylated isatin derivatives, hydroxylamine hydrochloride and alkyl halides in the presence of K_2CO_3 and tetrabutylammonium bromide (Bu_4NBr) under solvent-free condition (*Scheme*, *c*)). For comparison, we also examined the multi-step synthesis of the corresponding *N*-alkylated isatin *O*-alkyloximes using both conventional heating and microwave-assisted methods (*Scheme*, *a*) and *b*), resp.).

Scheme

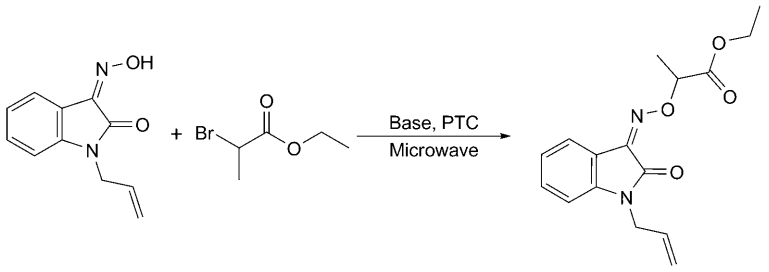


a) DMSO/H_2O 9:1, KOH, r.t., 3–5 h. *b*) K_2CO_3, MW (100 W), 0.08–10 min. *c*) K_2CO_3, Bu_4NBr, $NH_2OH \cdot HCl$, MW (300 W), 2–10 min.

Results and Discussion. – As shown in the *Scheme*, the alkylation of isatin was initially achieved with several alkyl halides to give *N*-alkylisatins **8a**–**8e** as the key intermediates. Several methods for *N*-alkylation of isatin have been published [3a], [12a][18], however, using NaH in refluxing DMF gave more satisfactory results [12a][18a]. Then, the obtained *N*-alkylisatins **8a**–**8e** were converted to the oxime derivatives **9a**–**9e** by stirring with $NH_2OH \cdot HCl$ in the presence of NaOH in EtOH at room temperature for a day (*Scheme*). Firstly, the synthesis of isatin *O*-alkyloximes **7a**–**7n** was achieved by conventional heating in solution (*Scheme*, *a*)). Compounds **9a**–**9e** reacted with diverse alkyl halides by stirring in 5M KOH/DMSO 1:9 at room temperature to afford **7a**–**7n** in moderate yields.

Since working with DMSO usually is not convenient, and often the extraction of products from the reaction media is followed by tedious workup procedures, we employed the microwave irradiation for the preparation of **7a**–**7n** from **9a**–**9e** under solvent-free condition (*Scheme*, *b*)). With the aim of optimizing the reaction conditions, we chose the reaction of *N*-allylisatin oxime **9a** with ethyl 2-bromopropanoate under microwave irradiation as a model reaction. The effect of several organic and inorganic bases, phase-transfer catalysts (PTCs), and irradiation power on the reaction time and yield of **7j** was examined. The results are summarized in *Table 1*. The best result was obtained in the presence of K_2CO_3 (*Entry 1*), and hence it was the base of choice for all reactions. Cs_2CO_3, NaH, and *t*-BuOK (*Entries 2*, *3*, and *7*) also furnished good yields of **7j**, while other bases gave only moderate yields.

Table 1. *Effect of Bases, PTCs, and Irradiation Power on the Conversion of* N-*Allylisatin Oxime* **9a** *into* O-*Alkyloxime Ether* **7j**



9a **7j**

Entry	Base	PTC	Irradiation power [Watt]	Time [s]	Yield [%][a])
1	K_2CO_3	–	100	5	93
2	Cs_2CO_3	–	100	5	82
3	NaH	–	100	5	75
4	CaO	–	100	10	60
5	Et_3N	–	100	10	64
6	MgO	–	100	15	70
7	*t*-BuOK	–	100	15	78
8	DABCO[b])	–	100	15	69
9	DMAP[c])	–	100	15	65
10	K_2CO_3	Bu_4NI	100	5	82
11	K_2CO_3	Bu_4NBr	100	5	93
12	K_2CO_3	Bu_4NCl	100	7	71
13	K_2CO_3	Bu_4NF	100	7	68
14	K_2CO_3	$Bu_4N(HSO_4)$	100	15	60
15	K_2CO_3	–	200	5	90
16	K_2CO_3	–	300	4	75
17	K_2CO_3	–	400	4	75
18	K_2CO_3	–	500	4	62

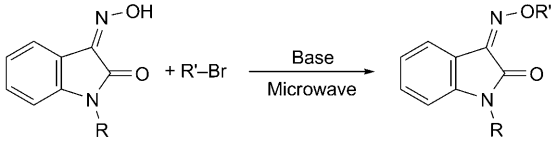
[a]) Yield of isolated product. [b]) 1,4-Diazabicyclo[2.2.2]octane. [c]) *N*,*N*-Dimethylpyridin-4-amine.

We also investigated the role of several tetrabutylammonium salts as phase-transfer catalysts (PTC) in the model reaction (*Table 1*). In the presence of PTCs (*Table 1*,

Entries 10–14), no distinguishable effect on the reaction was observed, and therefore, their addition is not necessary. We also studied the effect of microwave power on the progress of the model reaction (*Table 1*). The best results were achieved when a power of 100 or 200 Watt was used. Increment in radiation power did not enhance the yield of product.

To evaluate the general applicability of this method, the optimized conditions were applied to synthesize the *N*-alkylisatin *O*-alkyloximes **7a**–**7n** (*Scheme*). Comparing the results from the microwave-assisted with those of the solution procedure (*Table 2*) revealed that the microwave-assisted reactions (*Method b*)) proceeded more efficiently, and the desired isatin *O*-alkyloximes were obtained in good to excellent yields and after a short reaction time[1]).

Table 2. *Comparison by the Solution* (*Method a*)), *Microwave-Assisted* (*Method b*)), *and Microwave-Assisted Three-Component Procedure* (*Method c*)) *the Synthesis of Isatin* O-*Alkyloximes* **7a**–**7n**[1])



9 **7**

	Method a)		*Method b*)		*Method c*)	
	yield [%][a])	time [h]	yield [%][a])	time [min]	yield [%][a])	time [min]
7a	45	3	70	2	64	8
7b	58	3	82	3	75	5
7c	51	4	75	10	62	10
7d	53	3	78	0.5	68	4
7e	48	3	72	6	65	7
7f	59	4	90	1	77	6
7g	60	5	87	6	72	8
7h	60	4	80	5	74	7
7i	60	3	79	5	70	8
7j	65	5	93	0.08	83	3
7k	62	3	80	5	72	7
7l	58	3	78	6	69	8
7m	54	3	75	4	67	5
7n	51	5	70	0.5	65	2

[a]) Yield of isolated product.

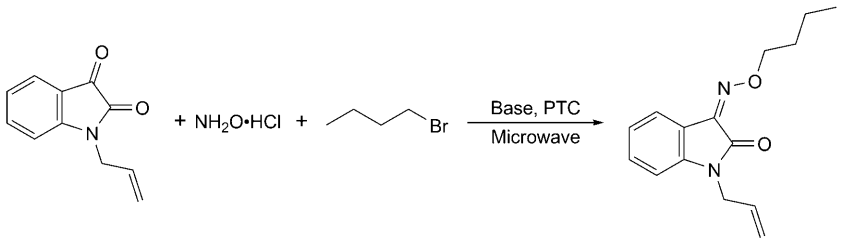
Although the microwave-assisted method is very useful for the preparation of the *O*-alkyloximes **7a**–**7n** from **9a**–**9e**, the problem usually associated with the oxime synthesis still remains since accessing pure oximes **9** usually requires the reiteration of recrystallization which is cumbersome, time-wasting, and eventually leads to a decrease in the overall yield. Hence, the synchronous oximation of *N*-alkylisatins and *O*-alkylation of *in situ* generated *N*-alkylisatin oximes with alkyl halides would be a

[1]) For R and R′, see *Scheme*.

suitable and efficient strategy. As there is no report that has exemplified this plan, we employed this novel method for the synthesis of isatin *O*-alkyloximes as illustrated in the *Scheme*, *Method c*). So, the synthesis of isatin oxime ethers **7a**–**7n** was achieved *via* a three-component reaction of **8a**–**8e**, $NH_2OH \cdot HCl$, and alkyl halides in the presence of K_2CO_3 and Bu_4NBr under microwave-assisted solvent-free conditions.

The first step for the three-component one-pot synthetic approach involved optimization of the reaction conditions. The choice of the base is of great importance for the *in situ* generation of isatin oximes and, therefore, the effect of various organic and inorganic bases, as well as of different PTCs and irradiation powers on the reaction of **8a**, $NH_2OH \cdot HCl$, and BuBr in the presence of Bu_4NBr was studied (*Table 3*). According to the results, K_2CO_3 (*Table 3*, *Entry 1*) was the most appropriate base. Moderate yields of *O*-alkyloxime **7b** were obtained in the presence of Cs_2CO_3 and *t*-BuOK (*Entries 2* and *7*).

Table 3. *The Effect of Bases, PTCs, and Irradiation Power on the Three-Component One-Pot Reaction of* N-*Allylisatin* **8a** *into* O-*Alkyloxime* **7b**



8a **7b**

Entry	Base	PTC	Irradiation power [Watt]	Time [min]	Yield [%][a])
1	K_2CO_3	Bu_4NBr	300	5	75
2	Cs_2CO_3	Bu_4NBr	300	5	65
3	NaH	Bu_4NBr	300	5	40
4	CaO	Bu_4NBr	300	10	37
5	Et_3N	Bu_4NBr	300	15	32
6	MgO	Bu_4NBr	300	10	25
7	*t*-BuOK	Bu_4NBr	300	10	67
8	DABCO[b])	Bu_4NBr	300	17	22
9	DMAP[c])	Bu_4NBr	300	17	27
10	K_2CO_3	–	300	40	n.r.[d])
11	K_2CO_3	Bu_4NI	300	8	60
12	K_2CO_3	Bu_4NCl	300	8	60
13	K_2CO_3	Bu_4NF	300	10	45
14	K_2CO_3	$Bu_4N(HSO_4)$	300	10	43
15	K_2CO_3	Bu_4NBr	100	25	35
16	K_2CO_3	Bu_4NBr	200	20	42
17	K_2CO_3	Bu_4NBr	400	4	56
17	K_2CO_3	Bu_4NBr	500	4	40

[a]) Yield of isolated product. [b]) 1,4-Diazabicyclo[2.2.2]octane. [c]) *N*,*N*-Dimethylpyridin-4-amine. [d]) No reaction.

In the absence of PTC, no reaction of the three components was achieved, even after prolonging the reaction time or enhancing the microwave power, and Bu_4NBr (*Table 3*, *Entry 1*) proved to be most efficient, while Bu_4NI and Bu_4NCl gave moderate yields of **7b**. Lower yields of **7b** were obtained when Bu_4NF and $Bu_4N(HSO_4)$ were used as PTC. The optimal result was attained when 50 mol-% of Bu_4NBr were used. Two roles of Bu_4NBr can be considered: *i*) the molten Bu_4NBr (m.p. 102–103°) creates a homogeneous reaction media, and it is assumed to behave as an ionic liquid; *ii*) Bu_4NBr absorbs the microwave irradiation and increases the temperature of the reaction media.

We also evaluated the effect of microwave power in the range 100–500 W on the progress of the model reaction (*Table 3*). The best result was obtained when irradiation at 300 W was used. Low yields of **7b** were obtained at 100 and 200 W; moreover, irradiation power >300 W had no distinguishable effect on the progress of the reaction.

The generality and versatility of the three-component one-pot method was demonstrated by its extension to various structurally diverse alkyl halides, including primary, secondary, and allylic as well as alicyclic halides (*Scheme*, *Table 2*). While *Methods a*) and *b*) required total times of >1 d for accessing **7a**–**7n**, only a few minutes (2–10 min) were necessary with *Method c*).

MCRs can use domino reactions with inseparable intermediates or sequential reactions with separable intermediates [19]. In our synthetic methodology, the first step involved the reaction of *N*-alkylisatin with NH_2OH to afford *N*-alkylisatin oxime as main intermediate. The oxime anion then attacks the alkyl halides to provide *N*-alkylisatin *O*-alkyloximes. The *in situ* generation of the *N*-alkylisatin oximes was established by TLC comparison of *in situ* generated *N*-alkylisatin oximes with the authentic samples **9a**–**9e**. Moreover, the intermediate oxime was also isolated at an early stage of the reaction and characterized by ^{1}H- and ^{13}C-NMR and IR spectroscopy.

Oximes are known to be ambident nucleophiles [20]. The alkylation of an oxime can be achieved either at the O-atom to afford *O*-alkyloximes or at the N-atom to generate nitrones [3a][12a][21]. It is known that various factors can influence the site of alkylation of oximes [21][22]. However, with the presented method, mainly *O*-alkyloximes were obtained, and no nitrones were detected, not even in trace amounts.

All compounds were fully characterized, and their structures were confirmed by elemental analysis, ^{1}H- and ^{13}C-NMR, and IR spectroscopy. Compounds **7a**–**7n** were expected to be formed as two geometrical isomers ((*E*)- or (*Z*)-isomer); however, (*Z*)-isomers were obtained predominantly as their structure was identified in the ^{1}H- and ^{13}C-NMR spectra by comparing the corresponding chemical-shift values. Indeed, the amide C(2)=O of isatin can deshield the nearby atoms in the (*Z*)-isomer. The minor (*E*)-isomer was detected in trace amounts ($<5\%$). To rationalize the dominant formation of (*Z*)-isomers rather than (*E*)-isomers, PM3 semi-empirical quantum-mechanic calculations were performed with MOPAC in CS Chem 3D Ultra 8 (*Cambridge Soft*, 2004) or Hyperchem (*Hypercube Inc.*, Version 7). The results are summarized in *Table 4*. The calculated ΔE for all *O*-alkyloximes **7a**–**7n** has a negative value. There is conformity of the experimental observations and calculated data (*Table 4*) establishing the higher stability of the (*Z*)-isomers and hence predominant formation of (*Z*)-products. The biological studies of **7a**–**7n** are currently under investigation and will be reported in due course.

Table 4. *Heat of Formation of Isatin* O-*Alkyloximes* **7a**–**7n** *Calculated with PM3*

	E_E[a]) [kcal/mol]	E_Z[b]) [kcal/mol]	ΔE[c]) [kcal/mol]
7a	28.36148	22.31933	– 6.04215
7b	17.06473	11.68065	– 5.38408
7c	16.22828	15.71943	– 0.50885
7d	53.27466	48.32347	– 4.95119
7e	26.57922	22.60886	– 3.97036
7f	– 9.74525	– 13.60926	– 3.86401
7g	4.70148	– 9.03000	– 13.73148
7h	– 17.36600	– 23.24679	– 5.88079
7i	30.70157	27.85983	– 2.84174
7j	– 50.60039	– 60.19243	– 9.59204
7k	5.54907	– 2.15372	– 7.70279
7l	8.43917	– 2.70314	– 11.14231
7m	– 75.33985	– 83.29056	– 7.95071
7n	– 84.01162	– 87.44306	– 3.43144

[a]) Heat of formation of the (*E*)-isomer. [b]) Heat of formation of the (*Z*)-isomer. [c]) $\Delta E = E_Z - E_E$.

In conclusion, a facile and highly efficient synthetic methodology is described for a three-component one-pot synthesis of novel *N*-alkylisatin *O*-alkyloximes **7a**–**7n** by *O*-alkylation of *in situ* generated *N*-alkylisatin oximes with alkyl halides under microwave-assisted solvent-free conditions. The selectivity of the reaction, conformity with environmentally benign chemistry, simple experimental procedure, mild reaction conditions and relatively high product yield are the advantages of this method. Finally, to rationalize the experimental evidences, the quantum-mechanical studies confirmed the considerable preference for (*Z*)-*O*-alkyloxime formation.

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Experimental Part

1. *General*. All chemicals were obtained from *Fluka* or *Merck*. Solvents were purified and dried by standard procedures, and stored over 3-Å molecular sieves. Microwave oven: *MB-245* domestic microwave oven from *Butan Industrial Co*. TLC: *SILG/UV 254* silica-gel plates. Column chromatography (CC): silica gel *60* (SiO_2; 0.063–0.200 mm, 70–230 mesh; *ASTM*). Melting points (M.p.): *Büchi-510* apparatus; in open capillaries; uncorrected. IR Spectra: *Shimadzu-FT-IR-8300* spectrophotometer; in cm^{-1}. ^1H- and ^{13}C-NMR Spectra: *Bruker-Avance-DPX-250* spectrometer; at 250 and 62.5 MHz, resp., δ in ppm, *J* in Hz. Elemental analyses (CHNS): *Perkin-Elmer-240-B* micro-analyzer.

2. N-*Alkylisatins* **8a**–**8e***: General Procedure.* A mixture of isatin (1.47 g, 0.01 mol), appropriate alkyl halide (0.012 mol), NaH (60% in paraffin wax, 0.4 g, 0.01 mol), and a cat. amounts of Bu_4NBr (0.1 g) was dissolved in dry DMF (30 ml) and then, heated to reflux for 10 h (TLC control). The solvent was evaporated, the residue dissolved in $CHCl_3$ (150 ml), and the soln. washed with H_2O (2×100 ml), dried (10 g of Na_2SO_4), and concentrated to afford the crude product which was purified by CC (SiO_2).

3. N-*Alkylisatin Oximes* **9a**–**9e***: General Procedure.* A mixture of the appropriate *N*-alkylisatin **8a**–**8e** (0.01 mol), $NH_2OH \cdot HCl$ (1.03 g, 0.015 mol), NaOH (0.6 g, 0.015 mol), and H_2O (minimum amount to dissolve $NH_2OH \cdot HCl$ and NaOH) was dissolved in EtOH (20 ml), and then the soln. was stirred for 24 h at r.t. Afterwards, the mixture was poured into 20 g of ice/10 g of H_2O. The oxime precipitate formed

immediately, which was filtered, washed with cold H_2O, and dried. Recrystallization from hot MeOH/H_2O afforded pure oximes **9a**–**9e** which were used for the next step.

4. *General Procedure for the Synthesis of* N-*Alkylisatin* O-*Alkyloximes* **7a**–**7n**, *Method* a). An appropriate alkyl halide (0.013 mol) was added portionwise to a soln. of the appropriate oxime **9a**–**9e** (0.01 mol), KOH (0.56 g, 0.01 mol), and H_2O (2 ml) in DMSO (20 ml). The mixture was stirred for 3–5 h at r.t. (TLC control). Then, the crude product was dissolved in $CHCl_3$ (150 ml), the soln. washed with H_2O (3×200 ml), dried (10 g of Na_2SO_4), and concentrated to afford the crude product which was purified by CC (SiO_2, AcOEt/hexane 1:5).

Method b). To the appropriate *N*-alkylisatin oxime **9a**–**9e** (5 mmol) in a mortar was added K_2CO_3 (0.69 g, 5 mmol), and the mixture was crushed vigorously to give a homogeneous mass. The mixture was transferred to a test tube, and then an alkyl halide (6 mmol) was added and mixed with a tiny spatula. The mixture was then irradiated in the microwave oven at 100 W for several time intervals (5 s). The irradiation was continued until TLC monitoring indicated no further improvement in the reaction (*Table 2*). The crude product was suspended in $CHCl_3$ (100 ml) and washed with H_2O (2×100 ml), the org. layer dried (10 g of Na_2SO_4) and concentrated, and the crude product purified by CC (SiO_2, AcOEt/hexane 1:5).

Method c). To the appropriate *N*-alkylisatin **8a**–**8e** (5 mmol) in a mortar was added a mixture of K_2CO_3 (0.69 g, 5 mmol), $NH_2OH \cdot HCl$ (0.68g, 10 mmol), and Bu_4NBr (2.5 mmol). The mixture was crushed vigorously to give a homogeneous mass, which was transferred to a test tube, and then an alkyl halide (6 mmol) was added and mixed with a tiny spatula. The mixture was then irradiated in the microwave oven at 300 W for several time intervals (1 min). The irradiation was continued until TLC monitoring indicated no further improvement in the reaction (*Table 2*). The crude product was suspended in $CHCl_3$ (100 ml) and washed with H_2O (2×100 ml), the org. layer dried (10 g of Na_2SO_4) and concentrated, and the crude product purified by CC (SiO_2, AcOEt/hexane 1:5).

*1-(Prop-2-en-1-yl)-1*H-*indole-2,3-dione* (**8a**). Purified by CC (SiO_2, AcOEt/hexane 1:8). Yield 2.70 g (72%). Red crystals. R_f (AcOEt/hexane 1:8) 0.65. M.p. 172–173°. IR (KBr): 3100*m*, 2964*m*, 2828*m*, 1715*s*, 1689*s*, 1489*m*. ^{1}H-NMR ($CDCl_3$): 8.63–6.87 (*m*, 4 arom. H); 5.76–5.92 (*m*, =CH); 5.35 (*dd*, $J = 1.8, 5.4$, $=CH_2$); 4.37 (*dd*, $J = 1.3, 4.0$, CH_2N). ^{13}C-NMR ($CDCl_3$): 42.00; 110.88; 116.88; 118.63; 123.79; 125.37; 130.31; 138.02; 138.12; 160.31; 180.81. Anal. calc. for $C_{11}H_9NO_2$ (187.19): C 70.58, H 4.85, N 7.48; found: C 70.45, H 4.70, N 7.35.

*1-Ethyl-1*H-*indole-2,3-dione* (**8b**). Purified by CC (SiO_2, AcOEt/hexane 1:5). Yield 2.40 g (68%). Yellow-orange crystals. R_f (AcOEt/hexane 1:5) 0.72. M.p. 162–163°. IR (KBr): 3050*m*, 2945*m*, 2855*m*, 1720*s*, 1689*s*, 1453*m*. ^{1}H-NMR ($CDCl_3$): 7.60–6.47 (*m*, 4 arom. H); 3.52 (*q*, $J = 5.0$, CH_2N); 1.01 (*t*, $J = 5.0$, Me). ^{13}C-NMR ($CDCl_3$): 12.46; 34.89; 110.11; 117.47; 123.59; 125.29; 138.44; 150.59; 157.80; 183.68. Anal. calc. for $C_{10}H_9NO_2$ (175.18): C 68.56, H 5.18, N 8.00; found: C 68.65, H 5.23, N 7.91.

*1-(2-Phenoxyethyl)-1*H-*indole-2,3-dione* (**8c**). Purified by CC (SiO_2, AcOEt/hexane 1:3). Yield 2.90 g (54%). Yellow crystals. R_f (AcOEt/hexane 1:3) 0.58. M.p. >250° (dec.). IR (KBr): 3096*m*, 2925*m*, 2834*m*, 1735*s*, 1689*s*, 1459*m*. ^{1}H-NMR ($CDCl_3$): 8.11–6.68 (complex, 9 arom. H); 4.11 (*t*, $J = 5.0$, CH_2O); 3.96 (*t*, $J = 5.0$, CH_2N). ^{13}C-NMR ($CDCl_3$): 40.12; 69.09; 111.23; 114.30; 114.53; 117.50; 121.38; 123.78; 125.18; 129.56; 138.33; 151.39; 158.48; 183.18. Anal. calc. for $C_{16}H_{13}NO_3$ (267.28): C 71.90, H 4.90, N 5.24; found: C 71.83, H 4.75, N 5.10.

*Ethyl 2,3-Dihydro-α-methyl-2,3-dioxo-1*H-*indole-1-acetate* (**8d**). Purified by CC (SiO_2, AcOEt/hexane 1:8). Yield 3.50 g (71%). Yellow crystals. R_f (AcOEt/hexane 1:8) 0.69. M.p. 222–223°. IR (KBr): 3096*m*, 2822*m*, 2848*m*, 1753*s*, 1732*s*, 1669*s*, 1382*s*, 1062*m*. ^{1}H-NMR ($CDCl_3$): 7.76–6.53 (*m*, 4 arom. H); 4.84 (*q*, $J = 7.3$, MeC*H*N); 3.93 (*q*, $J = 4.5$, MeCH_2O); 1.40 (*d*, $J = 7.3$, *Me*CHN); 0.90 (*t*, $J = 4.5$, *Me*CH_2O). ^{13}C-NMR ($CDCl_3$): 13.99; 14.19; 49.18; 62.04; 111.42; 117.77; 123.81; 125.44; 138.25; 149.44; 157.63; 169.30; 182.66. Anal. calc. for $C_{13}H_{13}NO_4$ (247.25): C 63.15, H 5.30, N 5.67; found: C 63.10, H 5.22, N 5.56.

*1-Cyclopentyl-1*H-*indole-2,3-dione* (**8e**). Purified by CC (SiO_2, AcOEt/hexane 1:8). Yield 3.30 g (76%). Yellow crystals. R_f (AcOEt/hexane 1:8) 0.69. M.p. 207–208°. IR (KBr): 3047*m*, 2954*m*, 1720*s*, 1665*s*, 1467*m*. ^{1}H-NMR ($CDCl_3$): 7.85–6.61 (*m*, 4 arom. H); 4.71–4.58 (*m*, NCH); 2.07–1.92 (*m*, CH_2CHCH_2); 1.72–1.67 (*m*, CHCH$_2$$CH_2$$CH_2$). ^{13}C-NMR ($CDCl_3$): 24.90; 27.73; 52.99; 111.35; 117.91;

123.30; 125.43; 138.03; 150.41; 158.06; 183.76. Anal. calc. for $C_{13}H_{13}NO_2$ (215.25): C 72.54, H 6.09, N 6.51; found: C 72.40, H 5.95, N 6.41.

*1-(Prop-2-en-1-yl)-1*H-*indole-2,3-dione 3-Oxime* (**9a**). Recrystallized from MeOH/H_2O. Yield 1.63 g (80%). Yellow crystals. R_f (AcOEt/hexane 2:1) 0.62. M.p. 211–212°. IR (KBr): 3216 (br.), 3050*m*, 2953*m*, 1715*s*, 1604*s*. ^{1}H-NMR ((D$_6$)DMSO): 10.26 (*s*, OH, exchangeable with D_2O); 9.08–6.90 (*m*, 4 arom. H); 5.89–5.70 (*m*, =CH); 5.14 (*dd*, *J* = 1.3, 8.3, =CH_2); 4.01 (*dd*, *J* = 1.4, 7.5, CH_2N). ^{13}C-NMR ((D$_6$)DMSO): 41.18; 109.08; 115.56; 116.69; 122.33; 125.96; 130.87; 131.97; 142.05; 143.10; 163.31. Anal. calc. for $C_{11}H_{10}N_2O_2$ (202.21): C 65.34, H 4.98, N 13.85; found: C 65.30, H 4.91, N 13.83.

*1-Ethyl-1*H-*indole-2,3-dione 3-Oxime* (**9b**). Recrystallized from MeOH/H_2O. Yield: 1.45 g (76%). Yellow crystals. R_f (AcOEt/hexane 2:1) 0.62. M.p. 175–176°. IR (KBr): 3234 (br.), 3091*m*, 2988*m*, 2818*m*, 1726*s*, 1653*s*. ^{1}H-NMR ($CDCl_3$): 10.39 (*s*, OH, exchangeable with D_2O); 8.40–6.77 (*m*, 4 arom. H); 3.81 (*q*, *J* = 7.0, CH_2N); 1.25 (*t*, *J* = 7.0, Me). ^{13}C-NMR ($CDCl_3$): 12.74; 34.70; 108.45; 115.79; 123.04; 128.15; 132.00; 143.08; 144.30; 164.14. Anal. calc. for $C_{10}H_{10}N_2O_2$ (190.2): C 63.15, H 5.30, N 14.73; found: C 63.10, H 5.24, N 14.65.

*1-(2-Phenoxyethyl)-1*H-*indole-2,3-dione 3-Oxime* (**9c**). Recrystallized from MeOH/H_2O. Yield 2.10 g (74%). Yellow crystals. R_f (AcOEt/hexane 2:1) 0.66. M.p. 163–164°. IR (KBr): 3205 (br.), 3009*m*, 2812*m*, 1700*s*, 1646*s*. ^{1}H-NMR ((D$_6$)DMSO): 13.44 (*s*, OH, exchangeable with D_2O); 8.51–6.54 (*m*, 9 arom. H); 4.17 (*t*, *J* = 5.0, CH_2O); 4.08 (*t*, *J* = 5.0, CH_2N). ^{13}C-NMR ((D$_6$)DMSO): 39.11; 64.87; 109.63; 114.31; 115.14; 120.77; 122.51; 126.71; 129.42; 131.85; 143.20; 157.98; 163.17; 164.31. Anal. calc. for $C_{16}H_{14}N_2O_3$ (282.29): C 68.07, H 5.00, N 9.92; found: C 68.01, H 4.98, N 9.89.

*Ethyl 2,3-Dihydro-3-(hydroxyimino)-α-methyl-2-oxo-1*H-*indole-1-acetate* (**9d**). Recrystallized from MeOH/H_2O. Yield 2.12 g (80%). Yellow crystals. R_f (AcOEt/hexane 2:1) 0.66. M.p. >250° (dec.). IR (KBr): 3215 (br.), 2956*m*, 2854*m*, 1710*s*, 1680*s*, 1378*s*. ^{1}H-NMR ((D$_6$)DMSO): 10.83 (*s*, OH, exchangeable with D_2O); 7.81–6.34 (*m*, 4 arom. H); 4.80 (*q*, *J* = 7.0, MeC*H*N); 4.56 (*q*, *J* = 6.3, MeCH_2O); 1.19 (*d*, *J* = 7.0, *Me*CHN); 1.05 (*t*, *J* = 6.3, *Me*CH_2O). ^{13}C-NMR ((D$_6$)DMSO): 15.50; 18.96; 39.25; 50.45; 110.90; 113.63; 115.58; 121.69; 125.70; 130.62; 142.36; 149.94; 163.83. Anal. calc. for $C_{13}H_{14}N_2O_4$ (262.26): C 59.54, H 5.38, N 10.68; found: C 59.50, H 5.30, N 10.62.

*1-Cyclopentyl-1*H-*indole-2,3-dione 3-Oxime* (**9e**). Recrystallized from MeOH/H_2O. Yield 1.83 g (79%). Yellow crystals. R_f (AcOEt/hexane 2:1) 0.68. M.p. >250° (dec.). IR (KBr): 3208 (br.), 3100*m*, 2865*m*, 1715*s*, 1674*s*. ^{1}H-NMR ((D$_6$)DMSO): 10.26 (*s*, OH, exchangeable with D_2O); 8.38–6.93 (*m*, 4 arom. H); 4.62–4.22 (*m*, CHN); 1.43–1.25 (complex, 4 CH_2). ^{13}C-NMR ((D$_6$)DMSO): 20.15; 20.28; 42.50; 108.99; 117.32; 120.99; 121.38; 125.95; 137.22; 145.61; 167.28. Anal. calc. for $C_{13}H_{14}N_2O_2$ (230.26): C 67.81, H 6.13, N 12.17; found: C 67.79, H 6.10, N 12.10.

*(3Z)-1-(Prop-2-en-1-yl)-1*H-*indole-2,3-dione 3-(*O-*Ethyloxime)* (**7a**). Purified by CC (SiO_2, AcOEt/hexane 1:5). Yield 0.73 g (64%). Orange oil. R_f (AcOEt/hexane 1:5) 0.66. IR (film): 3049*m*, 2875*m*, 1706*s*, 1670*m*, 1420*m*. ^{1}H-NMR ($CDCl_3$): 8.56–6.69 (*m*, 4 arom. H); 5.89–5.70 (*m*, =CH); 5.16 (*dd*, *J* = 1.3, 10.1, =CH_2); 4.49 (*q*, *J* = 3.5, CH_2ON); 4.27 (*dd*, *J* = 1.5, 4.25, CH_2N); 1.44 (*t*, *J* = 3.5, Me). ^{13}C-NMR ($CDCl_3$): 14.69; 42.08; 72.94; 108.50; 117.76; 122.95; 124.67; 130.93; 132.17; 140.10; 143.48; 159.95; 163.25. Anal. calc. for $C_{13}H_{14}N_2O_2$ (230.26): C 67.81, H 6.13, N 12.17; found: C 67.76, H 6.09, N 12.12.

*(3Z)-1-(Prop-2-en-1-yl)-1*H-*indole-2,3-dione 3-(*O-*Butyloxime)* (**7b**). Purified by CC (SiO_2, AcOEt/hexane 1:5). Yield 0.96 g (75%). Brown oil. R_f (AcOEt/hexane 1:5) 0.88. IR (film): 3006*m*, 2931*m*, 2790*m*, 1708*s*, 1699*m*, 1490*m*. ^{1}H-NMR ($CDCl_3$): 8.64–6.73 (*m*, 4 arom. H); 5.79–5.72 (*m*, =CH); 5.18 (*dd*, *J* = 1.4, 10.3, =CH_2); 4.72 (*t*, *J* = 7.5, CH_2ON); 4.38 (*dd*, *J* = 1.5, 4.3, CH_2N); 1.93–1.81 (*m*, CH_2CH_2ON); 1.42–1.33 (*m*, MeCH_2); 0.89 (*t*, *J* = 7.3, Me). ^{13}C-NMR ($CDCl_3$): 13.64; 19.82; 30.51; 41.91; 62.70; 108.50; 117.71; 118.09; 120.74; 122.52; 123.03; 124.79; 131.13; 140.10; 160.10. Anal. calc. for $C_{15}H_{18}N_2O_2$ (258.32): C 69.74, H 7.02, N 10.84; found: C 69.69, H 6.97, N 10.80.

*(3Z)-1-(Prop-2-en-1-yl)-1*H-*indole-2,3-dione 3-(*O-*Cyclopentyloxime)* (**7c**). Purified by CC (SiO_2, AcOEt/hexane 1:5). Yield 0.52 g (62%). Brown oil. R_f (AcOEt/hexane 1:5) 0.77. IR (film): 3100*m*, 2941*m*, 1704*s*, 1663*m*. ^{1}H-NMR ($CDCl_3$): 8.70–6.72 (*m*, 4 arom. H); 5.79–5.72 (*m*, =CH); 5.20 (*dd*, *J* = 1.2, 10.5, =CH_2); 4.34 (*dd*, *J* = 1.3, 4.5, CH_2N); 3.50–3.24 (*m*, CHON); 2.08–1.83 (*m*, 4 CH_2). ^{13}C-NMR ($CDCl_3$): 25.93; 32.23; 42.28; 70.58; 108.40; 117.71; 118.43; 122.45; 123.00; 124.71; 131.04; 131.27; 139.91; 160.10. Anal. calc. for $C_{16}H_{18}N_2O_2$ (270.33): C 71.09, H 6.71, N 10.36; found: C 71.02, H 6.67, N 10.30.

*(3Z)-1-(Prop-2-en-1-yl)-1*H*-indole-2,3-dione 3-[*O*-(Prop-2-en-1-yl)oxime]* (**7d**). Purified by CC (SiO_2, AcOEt/hexane 1:5). Yield 0.82 g (68%). Orange crystals. R_f (AcOEt/hexane 1:5) 0.58. M.p. 28–29°. IR (KBr): 3003*m*, 2896*m*, 1703*s*, 1665*m*, 1436*m*. ^{1}H-NMR ($CDCl_3$): 7.90–6.71 (*m*, 4 arom. H); 6.25–5.95 (*m*, CH_2=*CH*CH_2N); 5.82–5.66 (*m*, CH_2=*CH*CH_2O); 5.37 (*dd*, *J* = 1.5, 10.2, C*H*$_2$=CHCH$_2$O); 5.26 (*dd*, *J* = 1.3, 10.5, C*H*$_2$=CHCH$_2$N); 4.92 (*dd*, *J* = 1.3, 4.75, CH_2=CHC*H*$_2$O); 4.30 (*dd*, *J* = 1.75, 5.12, CH_2=CHC*H*$_2$N). ^{13}C-NMR ($CDCl_3$): 42.14; 78.00; 109.29; 115.72; 117.76; 119.03; 122.94; 127.92; 131.06; 132.40; 132.77; 143.61; 143.77; 163.16. Anal. calc. for $C_{14}H_{14}N_2O_2$ (242.27): C 69.41, H 5.82, N 11.56; found: C 69.37, H 5.79, N 11.52.

*(3Z)-1-(Prop-2-en-1-yl)-1*H*-indole-2,3-dione 3-[*O*-(2-Phenoxyethyl)oxime]* (**7e**). Purified by CC (SiO_2, AcOEt/hexane 1:5). Yield 1.04 g (65%). Yellow-orange crystals. R_f (AcOEt/hexane 1:5) 0.64. M.p. 49–50°. IR (KBr): 3023*m*, 2836*m*, 1703*s*, 1656*m*, 1324*s*. ^{1}H-NMR ($CDCl_3$): 8.24–6.70 (*m*, 9 arom. H); 5.81–5.66 (*m*, =CH); 5.26 (*dd*, *J* = 1.5, 10.3, =CH_2); 4.76–4.68 (*m*, PhOC*H*$_2$C*H*$_2$); 4.30 (*dd*, *J* = 1.4, 5.3, CH_2N). ^{13}C-NMR ($CDCl_3$): 42.19; 66.28; 75.41; 109.31; 114.53; 117.83; 120.98; 121.11; 123.06; 128.23; 129.43; 131.03; 131.33; 132.55; 143.72; 158.59; 163.14. Anal. calc. for $C_{19}H_{18}N_2O_3$ (322.36): C 70.79, H 5.63, N 8.69; found: C 70.70, H 5.59, N 8.65.

*2-({[(3Z)-1,2-Dihydro-2-oxo-1-(prop-2-en-1-yl)-3*H*-indol-3-ylidene]amino}oxy)acetic Acid Benzyl Ester* (**7f**). Purified by CC (SiO_2, AcOEt/hexane 1:5). Yield 1.34 g (77%). Yellow crystals. R_f (AcOEt/hexane 1:5) 0.65. M.p. 105–106°. IR (KBr): 3045*m*, 2868*m*, 1733*s*, 1702*s*, 1656*m*, 1324*s*, 1034*m*. ^{1}H-NMR ($CDCl_3$): 8.77–6.98 (*m*, 9 arom. H); 5.88–5.72 (*m*, =CH); 5.62 (*s*, PhC*H*$_2$O); 5.20 (*dd*, *J* = 1.3, 10.0, =CH_2); 4.36 (*dd*, *J* = 1.5, 4.8, CH_2N); 4.03 (*s*, CH_2ON). ^{13}C-NMR ($CDCl_3$): 41.96; 66.59; 72.87; 110.37; 115.22; 117.54; 123.32; 128.70; 128.87; 132.03; 133.79; 135.99; 140.84; 144.19; 160.00; 162.23; 165.85; 169.97. Anal. calc. for $C_{20}H_{18}N_2O_4$ (350.37): C 68.56, H 5.18, N 8.00; found: C 68.50, H 5.13, N 7.98.

*(3Z)-1-(Prop-2-en-1-yl)-1*H*-indole-2,3-dione 3-{*O*-[2-(1,3-Dioxo-1,3-dihydro-2*H*-isoindol-2-yl)ethyl]oxime}* (**7g**). Purified by CC (SiO_2, AcOEt/hexane 1:5). Yield 1.35 g (72%). Yellow-orange crystals. R_f (AcOEt/hexane 1:5) 0.44. M.p. 129–130°. IR (KBr): 3027*m*, 2812*m*, 1703*s*, 1679*m*, 1598*m*, 1024*m*. ^{1}H-NMR ($CDCl_3$): 8.27–6.64 (complex, 8 arom. H); 5.73–5.49 (*m*, CH_2=C*H*CH_2N); 5.13 (*dd*, *J* = 1.25, 8.12, C*H*$_2$=CHCH$_2$N); 4.59 (*t*, *J* = 5.0, =NOCH$_2$C*H*$_2$N); 4.16 (*dd*, *J* = 1.8, 5.7, CH_2=CHC*H*$_2$N); 4.02 (*t*, *J* = 5.0, =NOC*H*$_2$CH$_2$N). ^{13}C-NMR ($CDCl_3$): 37.28; 42.04; 73.09; 108.51; 115.45; 117.40; 123.30; 124.81; 128.15; 130.85; 131.45; 133.95; 140.16; 140.96; 144.33; 162.84; 167.61. Anal. calc. for $C_{21}H_{17}N_3O_4$ (375.38): C 67.19, H 4.56, N 11.19; found: C 67.16, H 4.53, N 11.16.

*(3Z)-1-(Prop-2-en-1-yl)-1*H*-indole-2,3-dione 3-{*O*-[4-(1,3-Dioxo-1,3-dihydro-2*H*-isoindol-2-yl)butyl]oxime}* (**7h**). Purified by CC (SiO_2, AcOEt/hexane 1:5). Yield 1.48 g (74%). Yellow-orange crystals. R_f (AcOEt/hexane 1:5) 0.52. M.p. 97–98°. IR (KBr): 3025*m*, 2843*m*, 1702*s*, 1686*m*, 1601*m*, 1024*m*. ^{1}H-NMR ($CDCl_3$): 8.55–6.71 (*m*, 8 arom. H); 5.83–5.66 (*m*, CH_2=C*H*CH_2N); 5.26 (*dd*, *J* = 1.0, 8.7, C*H*$_2$=CHCH$_2$); 4.75 (*t*, *J* = 7.0, =NOCH$_2$CH$_2$CH$_2$C*H*$_2$N); 4.39 (*dd*, *J* = 1.7, 5.3, CH_2=CHC*H*$_2$N); 3.66 (*t*, *J* = 7.0, =NOC*H*$_2$CH$_2$CH$_2$CH$_2$N); 2.00–1.89 (*m*, CH_2); 1.79–1.69 (*m*, CH_2). ^{13}C-NMR ($CDCl_3$): 25.59; 25.69; 37.29; 42.29; 61.90; 108.55; 109.26; 115.75; 117.76; 123.17; 124.82; 127.79; 131.09; 131.40; 132.02; 133.80; 140.17; 160.03; 168.26. Anal. calc. for $C_{23}H_{21}N_3O_4$ (403.43): C 68.47, H 5.25, N 10.42; found: C 68.45, H 5.21, N 10.40.

*(3Z)-1-(Prop-2-en-1-yl)-1*H*-indole-2,3-dione 3-{*O*-[4-(Naphthalen-2-yloxy)butyl]oxime}* (**7i**). Purified by CC (SiO_2, AcOEt/hexane 1:5). Yield 1.40 g (70%). Yellow-orange oil. R_f (AcOEt/hexane 1:5) 0.55. IR (film): 3021*m*, 2941*m*, 1707*s*, 1601*m*, 1224*m*. ^{1}H-NMR ($CDCl_3$): 8.64–6.69 (complex, 11 arom. H); 5.82–5.61 (*m*, CH_2=C*H*CH_2N); 5.16 (*dd*, *J* = 1.3, 8.9, CH_2=CHCH_2N); 4.82 (*t*, *J* = 7.0, =NOCH$_2$-CH$_2$CH$_2$C*H*$_2$OAr); 4.28 (*dd*, *J* = 1.3, 3.7, CH_2=CHC*H*$_2$N); 4.01 (*t*, *J* = 7.0, =NOC*H*$_2$CH$_2$CH$_2$CH$_2$OAr); 2.16–2.10 (*m*, CH_2); 1.91–1.85 (*m*, CH_2). ^{13}C-NMR ($CDCl_3$): 25.27; 26.32; 42.32; 62.36; 67.12; 106.61; 108.59; 117.80; 119.00; 120.84; 123.11; 123.53; 124.89; 126.23; 126.29; 127.62; 128.95; 129.33; 131.02; 131.19; 131.45; 134.55; 140.20; 159.85; 160.13. Anal. calc. for $C_{25}H_{24}N_2O_3$ (400.47): C 74.98, H 6.04, N 7.00; found: C 74.92, H 6.01, N 6.96.

*2-({[(3Z)-1,2-Dihydro-2-oxo-1-(prop-2-en-1-yl)-3*H*-indol-3-ylidene]amino}oxy)propanoic Acid Ethyl Ester* (**7j**). Purified by CC (SiO_2, AcOEt/hexane 1:5). Yield 1.25 g (83%). Yellow crystals. R_f (AcOEt/hexane 1:5) 0.48. M.p. 61–62°. IR (KBr): 3021*m*, 2973*m*, 2935*m*, 1716*s*, 1702*s*, 1610*m*, 1475*m*, 1354*s*. ^{1}H-NMR ($CDCl_3$): 8.33–6.76 (*m*, 4 arom. H); 5.86–5.71 (*m*, CH_2=C*H*CH_2N); 5.22 (*dd*, *J* = 1.7, 8.2, C*H*$_2$=CHCH$_2$N); 4.36–4.14 (*m*, CH_2=CHC*H*$_2$N, MeC*H*ON, MeC*H*$_2$O); 1.67 (*d*, *J* = 6.7, *Me-*

CHON); 1.18 (*t*, *J* = 7.2, *Me*CH_2O). ^{13}C-NMR ($CDCl_3$): 14.01; 14.77; 42.40; 62.20; 67.50; 108.72; 115.50; 117.92; 123.26; 125.23; 131.05; 131.82; 132.20; 143.81; 162.50; 171.20. Anal. calc. for $C_{16}H_{18}N_2O_4$ (302.33): C 63.56, H 6.00, N 9.27; found: C 63.52, H 5.94, N 9.21.

*(3Z)-1-Ethyl-1*H*-indole-2,3-dione 3-[*O*-(2-Phenoxyethyl)oxime]* (**7k**). Purified by CC (SiO_2, AcOEt/hexane 1:5). Yield 1.11 g (72%). Yellow oil. R_f (AcOEt/hexane 1:5) 0.60. IR (film): 3089*m*, 2973*m*, 2835*m*, 1701*s*, 1625*m*. ^{1}H-NMR ($CDCl_3$): 8.01–6.81 (*m*, 9 arom. H); 4.26 (*t*, *J* = 4.0, PhOC*H*$_2$CH$_2$); 3.96 (*t*, *J* = 4.0, PhOCH$_2$C*H*$_2$); 3.72 (*q*, *J* = 7.0, MeC*H*$_2$N); 1.17 (*t*, *J* = 7.0, *Me*CH_2N). ^{13}C-NMR ($CDCl_3$): 14.77; 42.40; 60.41; 68.06; 108.50; 113.52; 113.70; 120.08; 123.03; 124.79; 127.36; 128.50; 131.13; 134.10; 157.57; 160.10. Anal. calc. for $C_{18}H_{18}N_2O_3$ (310.35): C 69.66, H 5.85, N 9.03; found: C 69.61, H 5.82, N 9.00.

*(3Z)-1-(2-Phenoxyethyl)-1*H*-indole-2,3-dione 3-[*O*-(2-Phenoxyethyl)oxime]* (**7l**). Purified by CC (SiO_2, AcOEt/hexane 1:5). Yield 1.38 g (69%). Yellow oil. R_f (AcOEt/hexane 1:5) 0.56. IR (film): 3050*m*, 2920*m*, 2835*m*, 1702*s*, 1675*m*. ^{1}H-NMR ($CDCl_3$): 8.21–6.72 (complex, 14 arom. H); 4.74 (*t*, *J* = 4.5, PhOC*H*$_2$CH$_2$O); 4.28 (*t*, *J* = 4.5, PhOC*H*$_2$CH$_2$N); 4.14 (*t*, *J* = 4.5, PhOCH$_2$C*H*$_2$O); 4.06 (*t*, *J* = 4.5, PhOCH$_2$C*H*$_2$N). ^{13}C-NMR ($CDCl_3$): 39.81; 65.67; 66.27; 75.42; 109.51; 114.35; 114.72; 121.11; 121.20; 123.08; 124.56; 124.89; 128.15; 129.49; 129.63; 131.47; 132.50; 143.81; 144.62; 162.99. Anal. calc. for $C_{24}H_{22}N_2O_4$ (402.44): C 71.63, H 5.51, N 6.96; found: C 71.59, H 5.48, N 6.92.

*Ethyl (3Z)-2,3-Dihydro-α-methyl-2-oxo-3-[(2-phenoxyethoxy)imino]-1*H*-indole-1-acetate* (**7m**). Purified by CC (SiO_2, AcOEt/hexane 1:5). Yield 1.28 g (67%). Brown oil. R_f (AcOEt/hexane 1:5) 0.49. IR (film): 3050*m*, 2973*m*, 2835*m*, 1740*s*, 1705*s*, 1671*m*, 1322*s*. ^{1}H-NMR ($CDCl_3$): 8.79–6.74 (*m*, 9 arom. H); 4.42 (*q*, *J* = 4.0, MeC*H*N); 4.17 (*q*, *J* = 7.0, MeC*H*$_2$O); 3.96 (*t*, *J* = 4.0, PhOC*H*$_2$CH$_2$); 3.85 (*t*, *J* = 4.0, PhOCH$_2$C*H*$_2$); 1.41 (*d*, *J* = 4.0, *Me*CHN); 1.20 (*t*, *J* = 7.0, *Me*CH_2O). ^{13}C-NMR ($CDCl_3$): 14.12; 16.79; 51.27; 61.40; 63.42; 69.11; 111.40; 114.56; 114.65; 115.97; 117.66; 121.10; 121.23; 121.32; 129.54; 132.13; 133.03; 134.28; 158.62. Anal. calc. for $C_{21}H_{22}N_2O_5$ (382.41): C 65.96, H 5.80, N 7.33; found: C 65.92, H 5.79, N 7.30.

*2-({[(3Z)-1-Cyclopentyl-1,2-dihydro-2-oxo-3*H*-indol-3-ylidene]amino}oxy)propanoic Acid Ethyl Ester* (**7n**). Purified by CC (SiO_2, AcOEt/hexane 1:5). Yield 1.07 g (65%). Yellow oil. R_f (AcOEt/hexane 1:5) 0.51. IR (film): 3050*m*, 2944*m*, 2841*m*, 1726*s*, 1700*s*, 1664*m*, 1326*s*. ^{1}H-NMR ($CDCl_3$): 8.04–6.86 (*m*, 4 arom. H); 5.08 (*q*, *J* = 7.0, MeC*H*ON); 4.61 (*q*, *J* = 7.0, MeC*H*$_2$O); 4.20–4.11 (*m*, CHN); 2.11–2.09 (*m*, *CH*$_2$CH*CH*$_2$); 1.76–1.58 (*m*, CHCH$_2$*CH*$_2$*CH*$_2$); 1.39 (*d*, *J* = 7.0, *Me*CHON); 1.23 (*t*, *J* = 7.0, *Me*CH_2O). ^{13}C-NMR ($CDCl_3$): 13.90; 14.12; 24.32; 30.91; 43.88; 61.24; 80.01; 110.03; 114.69; 121.38; 122.56; 128.81; 129.55; 132.48; 134.28; 158.62. Anal. calc. for $C_{18}H_{22}N_2O_4$ (330.38): C 65.44, H 6.71, N 8.48; found: C 65.40, H 6.69, N, 8.45.

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