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The Reactions of Isatin and Its Derivatives with Aromatic and Heterocyclic *o*-Diamines

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Studies on the reaction of isatin and its derivatives with aromatic and heterocyclic *o*-diamines are reviewed, and the mechanism of these reactions as well as the methods for the identification of their products are examined.
The bibliography includes 57 references.

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I. INTRODUCTION

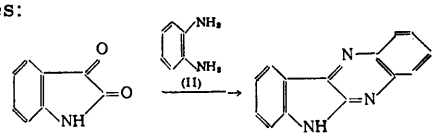
The reactions of isatin and its derivatives with aromatic and heterocyclic *o*-diamines, discovered in 1886 by Hinsberg[1], constitute an important method for the synthesis of condensed heteroaromatic systems, particularly purine analogues, as well as heteropolymers. The products of the reactions of isatins with aromatic heterocyclic *o*-diamines are of considerable interest as biologically active substances[2-15] (antimetabolites[2] and bactericidal[3-6], antifungal[7], antispasmodic, etc. agents), catalysts in colour photography[16], dyes for wool[17], and polydentate ligands capable of forming metal chelates stabilised by intramolecular hydrogen bonds[18-21], and are of interest as selective extractants[22].

It was believed for many years that the condensation of isatins with *o*-diamines always leads to derivatives of indolo[2,3-*b*]quinoxaline, but the latest data[15,20,21,23] have shown that the reaction does not proceed unambiguously and not only indolo[2,3-*b*]quinoxaline derivatives but also spiro-compounds are formed depending on the reaction conditions and the structures of the initial compounds.

Since the condensation of isatins with *o*-diamines has been attracting increasing attention in recent years, it is useful to survey the results of research on this reaction taking into account the latest data, with due attention to spectroscopic methods for the identification of the compounds formed.

II. THE REACTIONS OF ISATINS AND ITS DERIVATIVES WITH AROMATIC *o*-DIAMINES

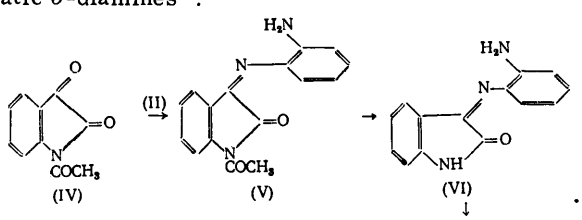
Schunk and Marchlewski[24-28] were two of the first investigators who studied the reactions of isatin (I) and its derivatives with aromatic *o*-diamines (II) in connection with the investigation of the properties of isatin. They obtained indolo[2,3-*b* quinoxaline] (III) and some of its derivatives:



(I) (III)

Subsequently the reactants employed in this reaction were isatin[1,15,17,21,23,24,27-35], *N*-substituted[6,23,28,30], alkyl- and alkoxy-[12,25,31,36-38], halogeno-[17,24,25,34,36,37], nitro-[17,34,36,37], naphtho-[37], and hydro-isatins[39,40] and isatin-carboxylic acids[8], on the one hand, and *o*-phenylenediamine[6,8,12,24-26,28,30,31,36,38-42], *o*-aminoalkylanilines and their salts[27,28,34,37,43-47], mono- and di-alkyl (alkoxy)-[1,17,21,26,28,33], mono- and di-halogeno[17], halogenoalkyl-[29], halogenonitro-[37], and nitro-*o*-phenylenediamines[31,37], *o*-diaminobenzoic acids[28], and *o*-naphthylenediamines[23], on the other. The reaction is usually carried out in aqueous or anhydrous acetic acid[23-25,33] and ethanol[15,23,48], more rarely in aqueous ethanol[38,48], benzene[32,33], and polyphosphoric acid[49,50], or without solvent[1].

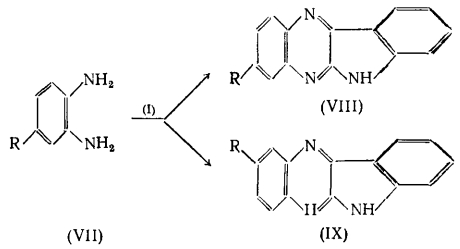
When 1-acetylisatin (IV) was condensed with *o*-phenylenediamine, Schunk and Marchlewski[38] obtained the acetylhydrazone (V), which was converted by hydrolysis into the hydrazone (VI), the latter being cyclised to 6*H*-indolo[2,3-*b*]quinoxaline (III). Similar hydrazones were obtained also by the reaction of 1-acetylisatin with other aromatic *o*-diamines[36]:



(IV) (V) (VI) (III)

When asymmetric *o*-diamines (VII) interact with isatin and its derivatives, there is a possibility of the formation of the structural isomers (VIII) and (IX), but, as a rule, one of the isomers is the dominant product[17], both isomers being obtained in approximately equal amounts only in the

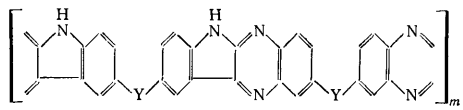
reactions of isatin with 4-ethoxy-*o*-phenylenediamine[28] and of 5-bromo-7-nitroisatin with 4-methoxy-*o*-phenylenediamine[17].



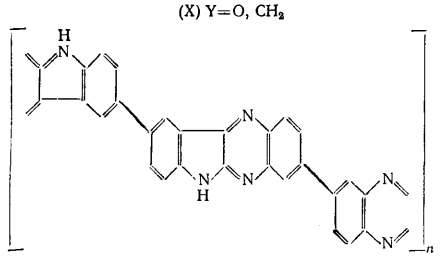
(VII) (VIII) (IX)

It is noteworthy that none of the structures of the indolo[2,3-*b*]quinoxalines obtained by the reactions of isatins with asymmetric *o*-diamines have been demonstrated.

The use as reactants in this process of 5,5′-oxybisisatinyl[49,50] and 5,5′-bisisatinyl[51], on the one hand, and oxybis-(3,4-diaminobenzene)[50], 4,4′-methylenebis-(*o*-phenylenediamine)[49], and 3,3-diaminobenzidine, on the other, made it possible to obtain the heteropolymers (X) and (XI). The condensation was carried out in polyphosphoric acid:

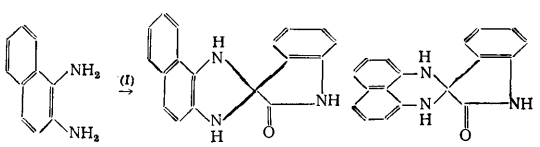


(X) Y=O, CH_2



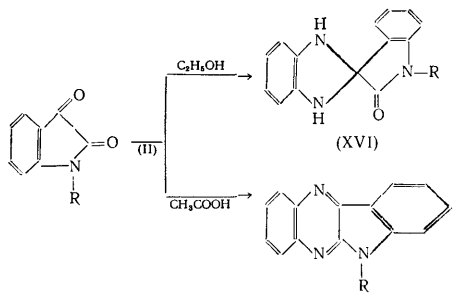
(XI)

Until 1958, it was believed that the only products of the reactions of isatins with *o*-diamines are derivatives of indolo[2,3-*b*]quinoxaline. However, Henseke and Lemke[48] established that the condensation of 1,2-naphthylenediamine (XII) with isatin proceeds with liberation of one water molecule. They suggested the spiro-structure (XIII) for the product of this reaction[48] by analogy with compound (XIV) obtained previously by Sachs[52] from 1,8-naphthylenediamine and isatin:



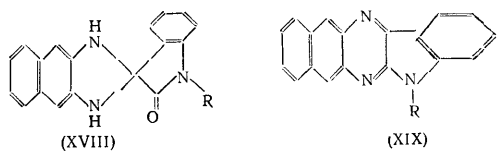
(XII) (XIII) (XIV)

Popp showed later[15,23] that, depending on the reactants and the reaction conditions (solvent), either linear products, or spiro-compounds, or both simultaneously may be formed. For example, the reaction of 1-methylisatin (XV) with *o*-phenylenediamine (II) in ethanol leads to the formation of 1′-methylspiro[benzimidazoline-2,3′-indolin]-2′-one (XVI), while in dilute acetic acid the product is 6-methylindolo[2,3-*b*]quinoxaline (XVII):



(XV) (XVI) (XVII)

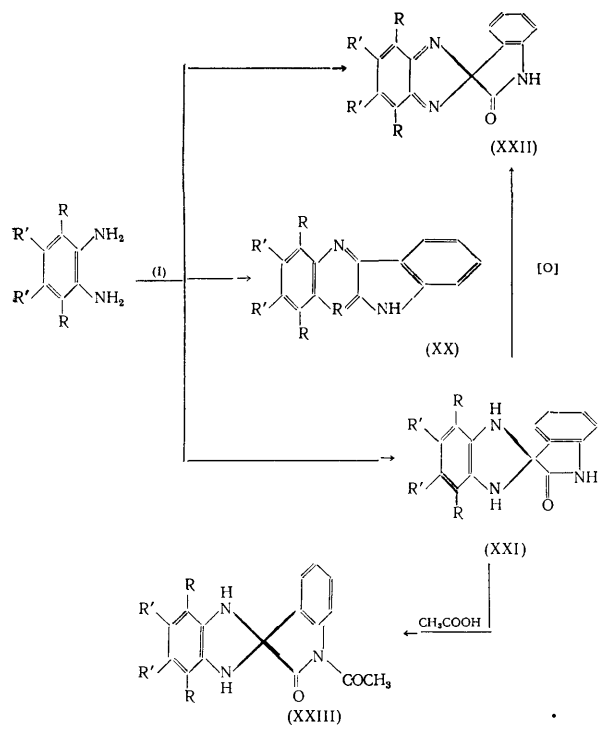
The reaction of 1-methylisatin with 2,3-diaminonaphthalene proceeds similarly. In contrast to its reaction with 1-methylisatin, 2,3-diaminonaphthalene reacts with 1-acetylisatin in ethanol to form a mixture of the spiro-compound (XVIII) and 8,9-benzo-6-acetylindolo[2,3-*b*]-quinoxaline (XIX), while the interaction in acetic acid of 9,10-diaminophenanthrene with 1-acetylisatin yields, in contrast to the reaction of the latter with *o*-phenylenediamine, only the spiro-compound:



(XVIII) (XIX)

The study of the reactions of 3,6- and 4,5-di-n-butoxy-*o*-phenylenediamines with isatin showed[21,33] that derivatives of spiro[2*H*-benzimidazole-2,3′-indolin]-2′-one (XXII) are formed together with derivatives of indolo-[2,3-*b*]quinoxaline (XX) and spiro[benzimidazoline-2,3′-indolin]-2′-one (XXI). When 3,6-di-n-butoxy-*o*-phenylenediamine interacts with isatin in 50% acetic acid, the product is a mixture consisting of indolo[2,3-*b*]quinoxaline (XX, R = n-BuO) and the spiro-compounds (XXI) and (XXII) with R = n-BuO and R′ = H, while in benzene in the presence of acetic acid only the spiro-compound (XXI, R = n-BuO, R′ = H) is formed. The mixture of products formed as a result of this reaction can be readily resolved owing to the different solubilities of these compounds in aqueous alcoholic solutions of alkalis and in benzene. The spiro-compound (XXI, R = n-BuO) is oxidised to the spiro-compound (XXII, R = n-BuO, R′ = H) by silver (I) oxide and is

acetylated by 80% acetic acid to the acetyl derivative (XXIII, R = n-Bu, R = H):

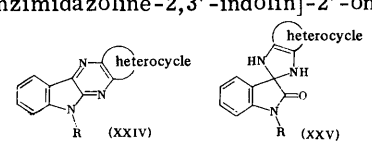


In contrast to 3,6-di-n-butoxy-*o*-phenylenediamine, the interaction of 4,5-di-n-butoxy-*o*-phenylenediamine with isatin in 50% acetic acid results in the formation of only the indolo[2,3-*b*]quinoxaline (XX, R = H, R′ = n-BuO) and the spiro-compound (XXII, R = H, R′ = n-BuO), while the reaction of these compounds in benzene in the presence of acetic acid results in the formation of only the indolo-[2,3-*b*]quinoxaline (XX, R = H, R′ = n-BuO).

III. THE REACTIONS OF ISATINS WITH HETEROCYCLIC *o*-DIAMINES

The heterocyclic *o*-diamines employed in this reaction were 1,2-diamino-5-n-propyl-4-phenylimidazole[32], 2,3- and 3,4-diaminopyridines[23], 4,5-diaminopyrimidine[15,23] and its derivatives[23,33], and 4,5-diaminobenzothiazole[53], the products consisting of a series of heterocyclic systems.

In contrast to the reactions with aromatic *o*-diamines, the reactions of isatins with heterocyclic *o*-diamines yielded only analogues of indolo[2,3-*b*]quinoxaline (XXIV) and spiro[benzimidazoline-2,3′-indolin]-2′-one (XXV):

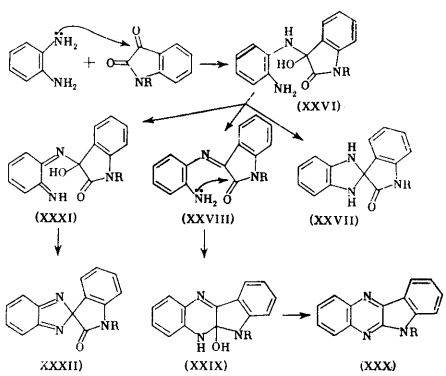


The structures of the initial compounds and the reaction conditions have a marked influence on the mode of reaction. Thus the interaction of 1,2-diamino-5-n-propyl-4-phenyl-imidazole[32], 4,5-diaminobenzothiazole[53], and certain derivatives of 4,5-diaminopyrimidine[33] with isatin yielded only linear products (XXIV). When 4,5-diamino-1,3-dimethyluracil was allowed to interact with isatin in acetic acid[23], the spiro-product (XXV) was obtained, while the interaction with 1-acetylisatin gave the linear product (XXIV).

The reaction of 3,4-diaminopyridine with isatin in ethanol leads to the formation of the linear product (XXIV), while 2,3-diaminopyridine and 4,5-diaminopyrimidine yielded the spiro-compound (XXV) under the same conditions[15]; the reactions of 2,3-and 3,4-diaminopyridines and 4,5-diaminopyrimidine with 1-acetylisatin also give rise to the spiro-compound (XXV).[15]

IV. THE MECHANISM OF THE REACTIONS OF ISATINS WITH *o*-DIAMINES

The studies examined above suggest the following mechanism for the interaction of *o*-diamines with isatins. The first stage involves the formation of the hydroxy-compound (XXVI), which loses a water molecule and is converted either into spiro[benzimidazoline-2,3′-indolin]-2′-one (XXVII) or into a Schiff base (XXVIII):



It is noteworthy that the formation of the Schiff base (XXVIII) was observed by Schunk and Marchlewski[36,38] in the reaction of 1-acetylisatin with aromatic *o*-diamines.

The Schiff base (XXVIII) cyclises to the hydroxy-compound (XXIX), which liberates a water molecule and is converted into the indolo[2,3-*b*]quinoxaline (XXX).

The formation of spiro[-2*H*-benzimidazole-2,3′-indolin]-2′-one (XXXII) can be regarded as oxidation, during the reaction, of the hydroxy-compound (XXVI) to compound (XXXI), which cyclises to the spiro-compound (XXXII) with liberation of a water molecule. This mechanism of the formation of the spiro-compound (XXXII) may be put forward in view of the fact that the latter is not formed during the reaction from the spiro-compound (XXII).[33]

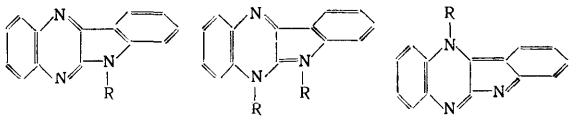
V. METHODS FOR THE IDENTIFICATION OF THE REACTION PRODUCTS

Since the reactions of isatins with *o*-diamines do not proceed unambiguously, with the possibility of the formation of not only the isomeric indolo-[2,3-*b*]quinoxalines (VIII) and (IX) but also of the spiro-compounds (XXI) and (XXII), depending on the reaction conditions and the initial compounds, the problem arises of the identification of the reaction products.

Ultraviolet Spectra

The ultraviolet spectra of indolo[2,3-*b*]quinoxaline derivatives have been investigated by a number of workers[33,35,42,54-56], but systematic studies have been made only by Badger and Nelson[56], who investigated the ultraviolet spectra of 6*H*-indolo[2,3-*b*]quinoxaline and its *N*-methyl derivatives in neutral, acid, and alkaline media.

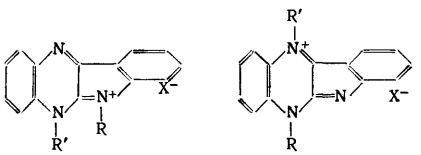
In theory, indolo[2,3-*b*]quinoxaline can exist in the form of three tautomeric forms (XXXIII)–(XXXV). By comparing its ultraviolet spectra with those of the *N*-methyl derivatives, it was shown[56] that indolo[2,3-*b*]quinoxaline exists preferentially in the 6*H*-form (XXXIII, R = H), since its ultraviolet spectrum is very similar to that of 6-methylindolo[2,3-*b*]quinoxaline and differs markedly from the spectrum of 5-methylindolo[2,3-*b*]quinoxaline in the range 400–520 nm.



(XXXIII) (XXXIV) (XXXV)

The spectrum of indolo[2,3-*b*]quinoxaline in sodium hydroxide solution is therefore similar to that of 5-methylindolo[2,3-*b*]quinoxaline, so that an appreciable proportion of the negative charge in the anion of indolo[2,3-*b*]quinoxaline is localised at the nitrogen atom in the 5-position.

The study of the spectra of the quaternary salts obtained by the protonation and methylation of indolo[2,3-*b*]quinoxaline showed that 6-methylindolo[2,3-*b*]quinoxaline is methylated and protonated in the 5-position with formation of the salt (XXXVI, R = R′ = CH_3; R = CH_3, R′ = H). 6*H*-Indolo[2,3-*b*]quinoxaline is also protonated in the 5-position, but this salt is capable of tautomerism and the solution probably contains both forms—(XXXVI) and (XXXVII) with R = R′ = H:



(XXXVI) (XXXVII)

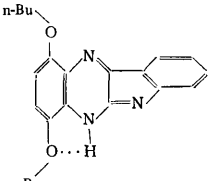
Methylation of 6*H*-indolo[2,3-*b*]quinoxaline by methyl toluene-*p*-sulphonate and methyl iodide as well as its protonation by toluene-*p*-sulphonic acid and HI lead to 5-methyl-11*H*-indolo[2,3-*b*]quinoxalinium iodide or toluene-*p*-sulphonate (XXV, R = CH_3, R′ = H, X = $CH_3C_6H_4SO_3^-$ or I^-). It is noteworthy that methylation of 6*H*-indolo[2,3-*b*]quinoxaline by methyl iodide or dimethyl sulphate in the presence of alkali as well as by diazomethane always yields a mixture of 5- and 6-methylindolo[2,3-*b*]quinoxalines. The formation of 11-methylindolo[2,3-*b*]quinoxaline was never observed under these conditions[56].

5-Methylindolo[2,3-*b*]quinoxaline is methylated in the 11-position with formation of 5,11-dimethylindolo[2,3-*b*]quinoxalinium toluene-*p*-sulphonate (XXXVII, R = R′ = CH_3, X = $CH_3C_6H_4SO_3^-$). Its ultraviolet spectrum resembles that of the protonated 5-methylindolo[2,3-*b*]quinoxaline, so that the latter is probably also protonated in the 11-position (XXXVI, R = CH_3, R′ = H, X = $CH_3C_6H_4SO_3^-$). Thus ultraviolet spectra can be used successfully for the identification of *N*-alkylindolo[2,3-*b*]quinoxalines.

Infrared Spectra

Owing to their low solubility in the corresponding solvents, the infrared spectra of the majority of the known indolo[2,3-*b*]quinoxalines and spiro[-2*H*-benzimidazole-2,3′-indolin]-2′-ones have not been investigated. The infrared spectroscopic data obtained by Ivashchenko and coworkers[21,33] are of interest, since they make it possible to interpret the structures of certain indolo[2,3-*b*]quinoxalines and demonstrate the possibility of using infrared spectroscopy for the identification of the products of the reactions of isatins with *o*-diamines.

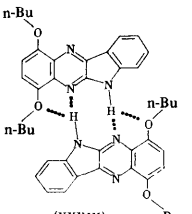
The infrared spectra of dilute solutions of indolo[2,3-*b*]-quinoxalines (XX) in chloroform have absorption bands characteristic of the stretching vibrations of the unassociated N–H bond (3425 and 3432 cm^{-1}). This shows that compound (XX) exists in the 6*H*-form, since otherwise the infrared spectrum of indolo[2,3-*b*]quinoxalines (XX, R = n-BuO, R′ = H) would be expected to exhibit a marked bathochromic shift of ν(NH) relative to the spectrum of indolo[2,3-*b*]quinoxaline (XX, R = H, R′ = n-BuO) owing to the formation of an intramolecular hydrogen bond, as in formula (XXXVIII):



(XXXVIII)

The existence of the 6*H*-forms of compounds (XX) is also confirmed by their ultraviolet spectra[33] which do not have absorption bands in the long-wavelength region.

The infrared spectra of solid specimens of indolo-[2,3-*b*]quinoxalines (XX) differ markedly from one another in the presence of different substituents in the region of the stretching vibrations of the N–H bon[21]. On the other hand, the spectrum of compound (XX) with R = H and R′ = n-BuO has a broad absorption band in the range 3000–3300 cm^{-1} with the fine structure characteristic of polyassociated compounds; the spectrum of compound (XX, R = n-BuO, R′ = H) has a single broadened band in this region at 3230 cm^{-1}), which apparently corresponds to the dimer. It is noteworthy that there is a possibility of two three-centre hydrogen bonds in the dimer of compound (XX, R = n-BuO, R′ = H), as in formula (XXXIX):



(XXXIX)

Since the formation of a "purely" three-centre hydrogen bond is extremely rare, it would be of undoubted interest to carry out X-ray diffraction studies on this compound.

The infrared spectrum of the spiro-compound (XXI, R = n-BuO, R′ = H) has an absorption band at 1668 cm^{-1} (amide-I) and two bands in the region of the stretching vibrations of the N–H bond. The band intensities led to the following assignments[33]: the 3454 cm^{-1} band (ϵ = 147)

to the stretching vibrations of the N–H bonds of the indoline ring and the 3370 cm^{-1} band (ϵ = 428) to the stretching vibrations of the N–H bonds of the benzimidazoline ring. When this compound is acylated, the 3454 cm^{-1} band disappears, while that at 3370 cm^{-1} remains unchanged, which indicates the acylation of the spiro-compound (XXI, R = n-BuO, R′ = H) at the NH group of the indoline ring with formation of compound (XXIII, R = n-BuO, R′ = H).

The infrared spectra of the spiro-compounds (XXII) [33] contain bands due to the stretching vibrations of the N–H bond of the indoline ring at 3390 cm^{-1} and the amide-I bands at 1732 and 1721 cm^{-1}.

Mass Spectrometry

The mass spectra of indolo[2,3-*b*]quinoxaline and spiro-[-2*H*-benzimidazole-2,3′indolin]-2′-one have been studied in greatest detail in three investigations[21,33,57], where they were used successfully to identify the products of the reactions of isatins with aromatic heterocyclic *o*-diamines.

The mass spectra of the isatin–*o*-diamine reaction products contained intense peaks due to the molecular ions. The main decomposition pathways of indolo[2,3-*b*]quinoxaline, spiro[2*H*-benzimidazole-2,3′-indolin]-2′-one and spiro[benzimidazoline-2,3′-indolin]-2′-one in dissociative ionisation have been examined in detail[21,33,57].

In conclusion, we must emphasise that the reactions of isatins with aromatic and heterocylic *o*-diamines do not proceed unambiguously, so that they can lead to derivatives of indolo[2,3-*b*]quinoxaline, spiro-compounds, or both simultaneously. The study of the ultraviolet, infrared, and mass-spectra of derivatives of indolo[2,3-*b*]-quinoxaline, its analogues, and spiro[2*H*-benzimidazole-2,3′-indolin]-2′-one showed that these methods can be used successfully in many instances for the identification of the compounds formed in the condensation reactions of isatins with *o*-diamines.

Many studies carried out without taking into account the latest data and the evidence for the structures of the compounds obtained must be treated critically, since the structures of indolo[2,3-*b*]quinoxaline derivatives have been traditionally attributed without proof (and without demonstrating their individuality) to the products of the reactions of isatins with *o*-diamines.

The reactions have not been studied systematically and analysis of the data discussed in this review does not reveal systematic features in the formation of derivatives of indolo[2,3-*b*]quinoxaline and spiro-compounds as a function of the reaction conditions and the structures of the initial reactants. The study of these reactions is therefore of undoubted interest from both practical and theoretical points of view.

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