Pergamon

Tetrahedron, Vol. 53, No. 14, pp. 5305-5324, 1997

0040-4020/97 $17.00 + 0.00

PII: S0040-4020(97)00194-4

The Reaction of Phthalidylidene Dichloride with Primary Amines. Synthesis and X-Ray Molecular Structure of N-Substituted Phthalisoimides

Antonio Guirado*, Andrés Zapata

Departamento de Química Orgánica, Facultad de Química, Universidad de Murcia, Campus de Espinardo, E-30071, Murcia, Apartado 4021, Spain.

M. Carmen Ramírez de Arellano

Departamento de Química Inorgánica, Facultad de Química, Universidad de Murcia, Campus de Espinardo, E-30071, Murcia, Apartado 4021, Spain.

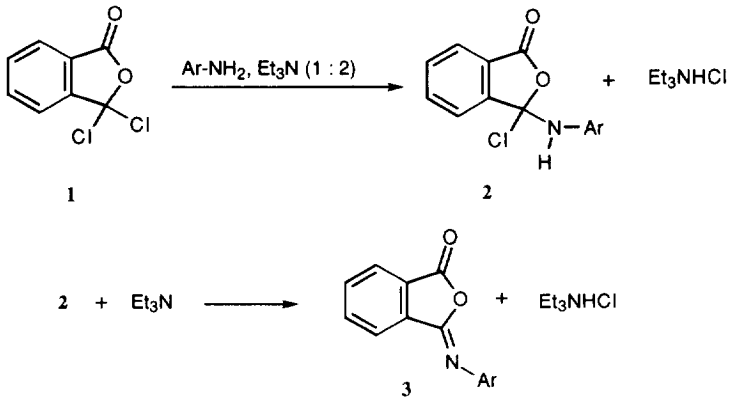
Abstract : An efficient method for the synthesis of N-substituted phthalisoimides, by reaction of phthalidylidene dichloride with primary amines, is described. The reactions with arylamines, arylenediamines and alkylenediamines lead to the corresponding phthalisoimides or bisphthalisoimides in nearly quantitative yields. However, the reactions with alkylamines are not useful because of the relatively high nucleophilicity of alkylamines. Certain particular behaviours of arylamines, associated with the presence of specific ortho-substituents have been found. The reactions of arylamines bearing an o-hydroxymethyl group provide a convenient method for preparing 2-benzoxazinylbenzoic acids. The X-ray crystallographic structures of N-(2-methoxyphenyl)phthalisoimide **3a** and 2-(4H-3,1-benzoxazin-2-yl)benzoic acid **15a** have been determined. © 1997 Elsevier Science Ltd.

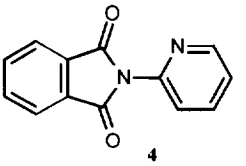
N-Substituted phthalisoimides have normally been prepared by dehydration of phthalamic acids.[1] Some less usual reactions yielding phthalisoimides have also been reported.[2-4] The dehydrative cyclization reactions of phthalamic acids can be promoted by a wide variety of reagents. However, the formation of the isomers phthalimides is in many cases a severely competitive process. In spite of the variety of reagents causing efficient dehydration, ethyl chloroformate, trifluoroacetic anhydride and N,N'-dicyclohexylcarbodiimide are only the most frequently reported agents that give phthalisoimides.[1,5-7] A comparative study of these three reagents and acetic anhydride has been reported, showing N,N'-dicyclohexylcarbodiimide as the most general in yielding phthalisoimides.[6] Regarding the competition between phthalisoimide against phthalimide formation, a common reaction pathway involving intramolecular interaction of the amide function with suitably disposed activated acyl groups has been evidenced.[6, 8-10] Each dehydrative reagent plays a substantial role in the activation type of the acyl group, whereas the relative nucleophilicity of nitrogen and oxygen atoms

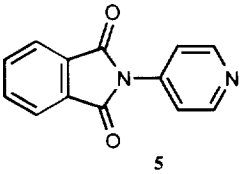
of the amide group is another crucial factor in determining the reaction result. The facile thermal or catalyzed transformation of phthalisoimides into phthalimides[1,10-12] is also an important factor. Therefore, it is possible that in spite of a kinetically favoured formation of phthalisoimide, the phthalimide may be found as the final product, especially if the reaction is carried out at a relatively high temperature or if any nucleophilic catalyst is present in the reaction medium. To summarize, the synthesis of phthalisoimides is more difficult than the synthesis of phthalimides since an efficient phthalisoimide preparation is dependent on many factors in conjunction, whose effective control is often difficult in practice. This provides an explanation for erratic results that are observed even when a reagent recognized as appropriate for preparing phthalisoimides is used.[13] Given the above, the development of improved methods for synthesizing phthalisoimides in order to suppress ambiguity in the firstly formed products and avoid their isomerization is thus a subject of considerable interest.

Phthalidylidene dichloride **1** is an inexpensive and easily available reagent[14] bearing an active centre which suggested an attractive entry to 3-substituted phthalides. Reported reactions include the reaction with phosphines giving trans-diphthalyl as the main product.[15] Several phthalide derivatives were electrochemically prepared.[16] The efficient synthesis of 3-indenylidenephthalides[17] and the reaction with 4,5-dimercapto-1,3-dithiole-2-thione dianion giving the corresponding spiro tetrathiapentalene-2-thione in excellent yield[18] were recently reported. As was preliminarily communicated the reaction of phthalidylidene dichloride with arylamines provided an efficient and easy method for the synthesis of N-arylphthalisoimides.[19] In this paper we describe full details of the previous communication as well as additional new results derived from exploring the scope and limitations of the reported reaction.

When phthalidylidene dichloride **1** was treated with equimolecular amounts of arylamines and two equivalents of triethylamine, an almost instantaneous formation of a precipitate of triethylamine hydrochloride was observed upon addition of the first drops of reagent which suggested a remarkably fast reaction. The products formed were isolated and identified as the corresponding N-arylphthalisoimides **3** by i.r., m.s., n.m.r. spectroscopy and microanalyses. Direct comparisons with some authentic specimens as well as isomerizations to N-arylphthalimides were also used for characterization. The results for a number of experiments are shown in Scheme 1. Yields were nearly quantitative. Geometrical characteristics of this class of compound were determined by X-ray crystallography of product **3b**. The molecular structure found is illustrated in Figure 1. The phthalisoimide ring system is planar (mean deviation 0.024 Å). The mutual orientation of the rings is given by the torsion angle C14-C9-N-C8: 63.7°. Selected intramolecular distances (crystallographic numbering of atoms) and selected bond angles are given in Table 1.




4


5

Entry	Ar
3a	C_6H_5
3b	2-$CH_3OC_6H_4$
3c	4-$CH_3OC_6H_4$
3d	2-BrC_6H_4
3e	2-IC_6H_4
3f	4-$NO_2C_6H_4$
3g	4-$CH_3C_6H_4$
3h	4-BrC_6H_4
3i	2-$CH_3C_6H_4$
3j	2,4-$Cl_2C_6H_3$
3k	4-ClC_6H_4
3l	2-$C_{10}H_7$
3m	1-$C_{10}H_7$
3n	2-$(CH_3)_3CC_6H_4$

Scheme 1

With few exceptions the reaction of phthalidylidene dichloride with primary aromatic amines lead unambiguously to the corresponding phthalisoimides **3**. The availability of starting materials, the mildness of the reaction conditions as well as the efficiency are relevant advantages of this synthetic method. The case of the easy isolation of products is also an important advantage; highly pure phthalisoimides can be obtained by removing the solvent under reduced pressure and by washing the solid residue with cold water.[20] This is especially significant in order to avoid isomerization processes, since for phthalisoimides in an impure state, they present special proclivity to isomerize due to the variety of agents that are effective catalysts for this process.[8] Evidently, any impurity present might catalyze the isomerization. On attempting the conversion of N-phenylphthalisoimide **3a** into N-phenylphthalimide, a notable reluctance of this product towards

isomerization was observed. This process has been reported as occurring completely in 1 h in boiling benzene[21] (bp 80 °C). However, our sample could not be transformed under similar conditions and was recovered unchanged after boiling for 24 h. Similar results were obtained with acetonitrile (bp 82 °C) and toluene (bp 111 °C) when the solution was also refluxed for 24 h. The transformation finally occurred in DMF (bp 152 °C) with N-phenylphthalimide being quantitatively formed after heating in refluxing for 1 h. These facts can only be explained on the basis of the absence of impurities in the prepared phthalisoimide and solvents.[13]

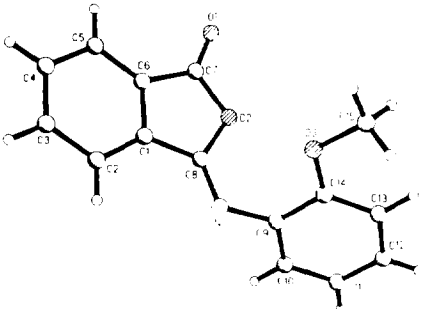


Figure 1. Molecular structure of **3b**, *showing the crystallographic numbering system used*

Table 1. Selected Bond Lengths and Bond Angles in Crystal Structure of **3b**

lengths (Å)

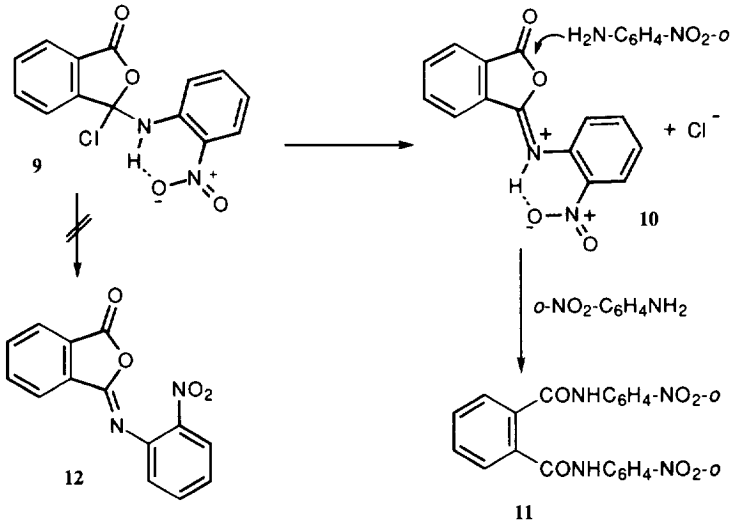
N-C(8)	1.253(2)	N-C(9)	1.425(2)
O(1)-C(7)	1.197(2)	O(2)-C(7)	1.395(2)
O(2)-C(8)	1.405(2)	O(3)-C(14)	1.367(2)
O(3)-C(15)	1.431(2)	C(1)-C(6)	1.384(2)
C(6)-C(7)	1.473(2)	C(1)-C(8)	1.469(2)

angles (°)

C(8)-N-C(9)	122.42(11)	C(7)-O(2)-C(8)	109.36(9)
C(14)-O(3)-C(15)	116.88(10)	C(2)-C(1)-C(8)	130.76(12)
O(3)-C(14)-C(9)	116.19(11)	C(6)-C(1)-C(8)	107.84(11)
C(14)-C(9)-N	120.81(12)	C(1)-C(6)-C(7)	107.79(11)
C(5)-C(6)-C(7)	130.21(12)	O(1)-C(7)-O(2)	120.70(12)
O(1)-C(7)-C(6)	131.72(13)	O(2)-C(7)-C(6)	107.57(11)
N-C(8)-O(2)	124.76(11)	N-C(8)-C(1)	127.82(11)
O(2)-C(8)-C(1)	107.42(10)	C(10)-C(9)-N	119.30(12)

The reactions with 2- and 4-aminopyridines were also carried out. In this case N-pyridylphthalimides **4** and **5** were almost quantitatively formed. This result is attributable to autocatalyzed isomerization of the firstly formed N-pyridylphthalisoimides.

It is to be noted that the reaction of phthalidylidene dichloride with 4-nitroaniline yielded N-(4-nitrophenyl)phthalisoimide **3f**. However, the reaction with 2-nitroaniline gave N,N'-di(2-nitrophenyl)phthalamide **11** (74%) instead of the expected N-(2-nitrophenyl)phthalisoimide **12**. In view of these contrary results the reaction with 2-terbutylaniline was carried out. The expected N-(2-terbutylphenyl)phthalisoimide **3n** was obtained in nearly quantitative yield. Consequently, it is to be presumed that electron-withdrawing properties and steric hindrance of substituents play no important roles in causing the observed anomalous behaviour. It is also to be considered, as will be discussed later, that the attack of amines on phthalisoimides to yield phthalamides requires stronger nucleophiles than arylamines. Hence, the participation of an activated intermediate with enhanced electrophilic reactivity should be postulated. This, therefore points clearly towards attributing the particular behaviour of 2-nitroaniline to an intramolecular hydrogen bonding effect. Because of hydrogen bonding (Scheme 2), a relatively disfavoured elimination of hydrogen chloride from the intermediate **9** would result. Alternatively, the generation of the phthalisoimidium salt **10** and its reaction with 2-nitroaniline would participate in the formation of the phthalamide **11**. This pathway is also well supported by the fact that reactions of phthalisoimidium salts with amines leading to phthalamides have been described.[21]



Scheme 2

The reactions of phthalidylidene dichloride with 2-aminobenzyl alcohols provided non-phthalisoimide products. The product derived of 2-aminobenzyl alcohol was first considered to be the expected isoimide **13a**. However, it is an isomer which was unequivocally identified by X-ray crystallography as the benzoxazine

derivative **15a**. The molecular structure found is illustrated in Figure 2. Selected intramolecular distances (crystallographic numbering of atoms) and selected bond angles are given in Table 2. The adjacent molecules (Figure 3) interact through one hydrogen bond. Intermolecular bond distances: O2···N = 2.644 (2) Å; H2···N = 1.835 (2) Å; O2-H2-N angle = 161.20 (2)°.

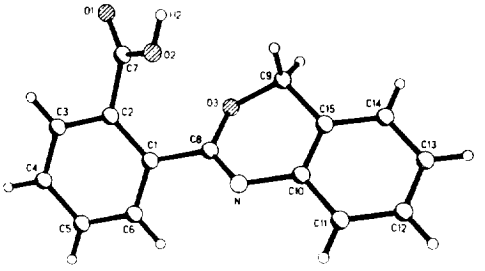


Figure 2. Molecular structure of **15a**, *showing the crystallographic numbering system used*

Table 2.Selected Bond Lengths and Bond Angles in Crystal Structure of **15a**

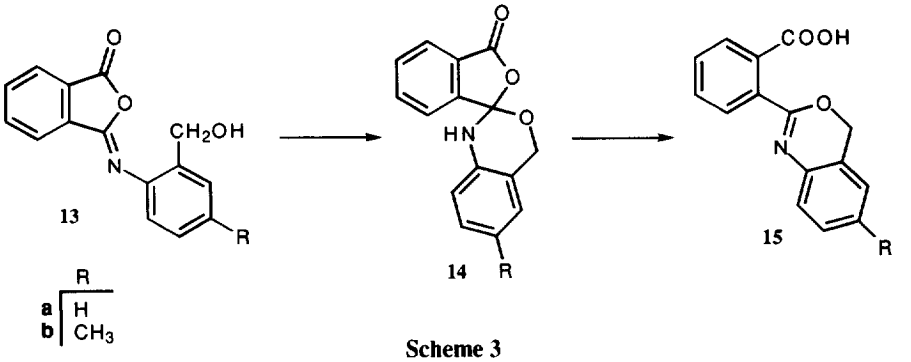
lengths (Å)

O(1)-C(7)	1.210(2)	O(2)-C(7)	1.322(2)
O(3)-C(8)	1.348(2)	O(3)-C(9)	1.453(2)
N-C(8)	1.282(2)	N-C(10)	1.419(2)
C(1)-C(8)	1.480(2)	C(9)-C(15)	1.496(2)
C(2)-C(7)	1.497(2)		

angles (°)

C(8)-O(3)-C(9)	116.54(10)	C(8)-N-C(10)	117.83(11)
C(10)-C(15)-C(9)	117.15(13)	C(6)-C(1)-C(8)	119.50(13)
C(3)-C(2)-C(7)	118.24(13)	C(1)-C(2)-C(7)	121.96(13)
O(1)-C(7)-O(2)	125.29(14)	O(1)-C(7)-C(2)	123.05(13)
O(2)-C(7)-C(2)	111.58(12)	N-C(8)-O(3)	125.57(13)
N-C(8)-C(1)	122.55(12)	O(3)-C(8)-C(1)	111.69(11)
O(3)-C(9)-C(15)	111.19(12)	C(14)-C(15)-C(9)	122.95(13)
C(15)-C(10)-N	120.52(12)	C(11)-C(10)-N	119.36(12)

The formation of products **15** can be rationalized by prior formation of phthalisoimides **13** as is shown in Scheme 3. Addition is a common feature of the reactivity of the imino group instead of nucleophilic substitution. Hence, the intermediates **13** would undergo an intramolecular addition-elimination process, where the plausible intermediates **14** would be involved, to afford the final products **15**. The preparation of these products has not been previously reported. Considering the previously known syntheses of benzoxazines[22] this specific reaction has significant interest as a preparative method.



Scheme 3

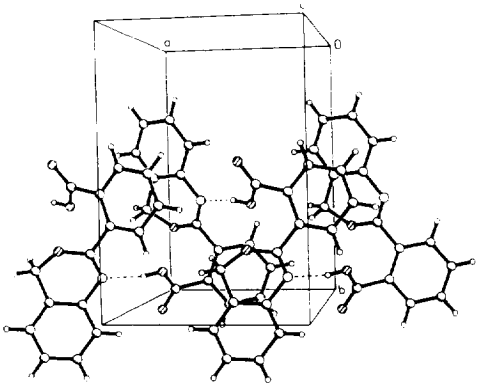
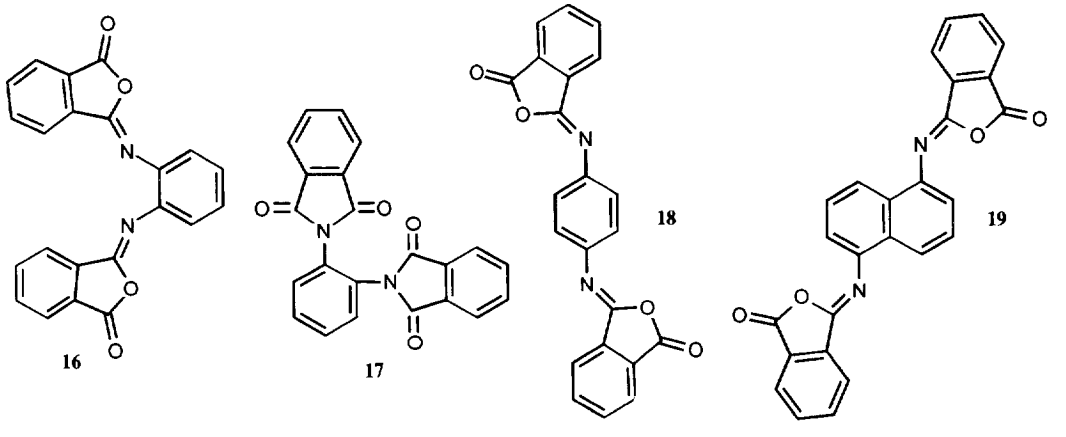
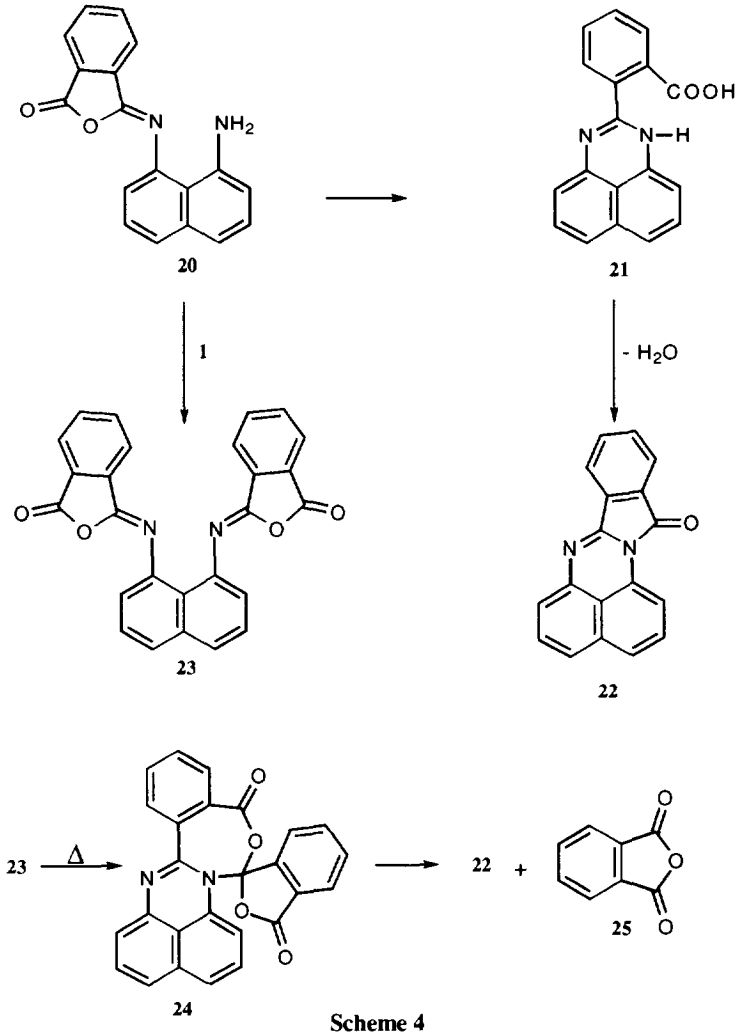


*Figure 3. A perspective of the crystal packing of **15a**, showing the intermolecular hydrogen interactions*

The reactions with some aromatic diamines were also carried out; 1,2- and 1,4-phenylenediamines as well as 1,5-naphthalenediamine gave the corresponding bisphthalisoimides **16**, **18**, and **19** in almost quantitative yield. However, 1,8-naphthalenediamine gave the expected bisphthalisoimide **23** as the



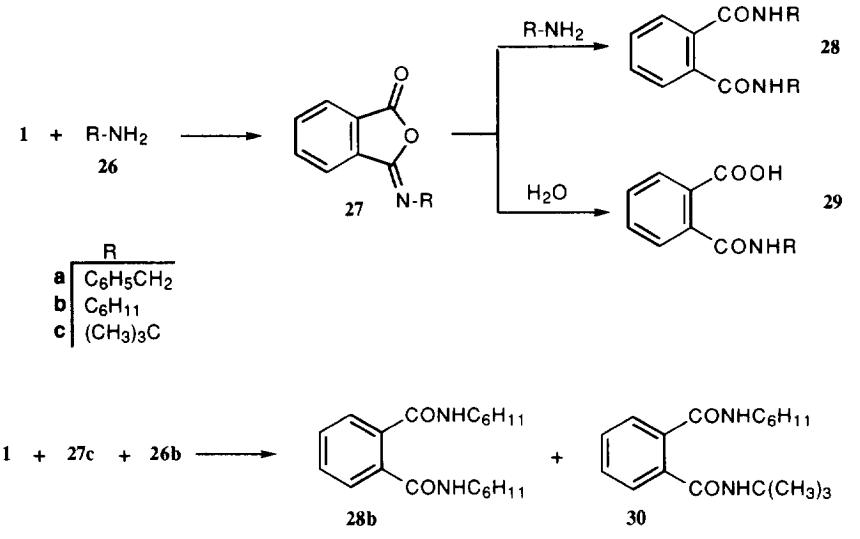
major product accompanied by a product which was identified as phthaloperinone **22**. The product **23** could not be obtained by the amic acid dehydration method since the reaction of phthalic acid anhydride with 1,8-diaminonaphthalene leads to 2-(2-perimidyl)benzoic acid **21** which dehydrates to **22**. The structure of product **22** was confirmed by direct comparison with an authentic sample prepared by the above mentioned reactions.[23] Because of the extreme insolubility of product **23** in the usual spectroscopic solvents its structure was confirmed by X-ray crystallography.[24] The formation of products **22** and **23** is rationalized in Scheme 4, involving the partially condensed species **20** as a common intermediate.



Scheme 4

Product **23** melts with a remarkably rapid change from yellow to red. To get an insight into the nature of the observed process the heating of an appropriate amount of sample was carried out. The quantitative

transformation of **23** into a mixture of phthaloperinone **22** and phthalic anhydride **25** in a 1:1 ratio was observed. The formation of these products (Scheme 4) can be rationalized via a free reagent intramolecular process involving the generation of the spiro intermediate **24**. It seems reasonable to assume that thermal decomposition of **24** would liberate products **22** and **25** directly. In contrast to the observed behaviour of bisphthalisoimide **23**, the heating of the bisphthalisoimide **16** gave only the corresponding bisphthalimide **17**.

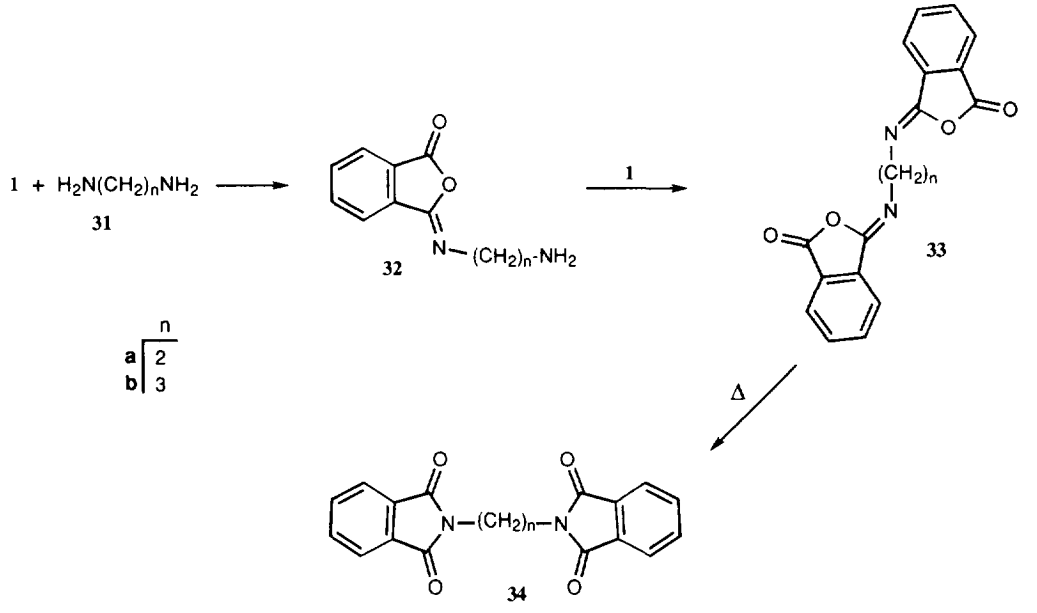


Scheme 5

Results of the reactions with alkylamines were remarkably different to those obtained with arylamines. When phthalidylidene dichloride reacted with alkylamines **26** two major products were observed which were isolated and characterized by comparisons with authentic specimens as the corresponding N,N'-dialkylphthalamides **28** and N-alkylphthalamic acids **29** . The reasons for this remarkably different behaviour were clarified as follows: The fresh crude products of the reaction of phthalidylidene dichloride with terbutylamine, were checked before being washed with water. In this case N-terbutylphthalisomide **27c** and N,N'-diterbutylphthalamide **28c** were unequivocally identified as the reaction products by direct comparison with authentic samples. When this mixture was worked up in the usual manner the transformation of **27c** to **29c** was observed. On the other hand, the formation of phthalamides **28** must necessarily be attributed to the reaction of alkylamines with phthalisoimides. These results also indicate that such reactions as well as those of alkylamines **26** with phthalidylidene dichloride **1** would be competitive processes with comparable rates. This was confirmed when a mixture of equimolecular amounts of phthalidylidene dichloride **1** and N-

terbutylphthalisoimide **27c** (Scheme 5) was treated with a deficient amount of cyclohexylamine **26b**. The formation of N,N'-dicyclohexylphthalamide **28b** and N-cyclohexyl-N'-terbutylphthalamide **30** in a ratio of 5 : 4, was determined. Therefore, it was firmly shown that in comparison with the reactions of arylamines, the stronger nucleophilic character of alkylamines and the facile hydrolysis of N-alkylphthalisoimides are the causes of the lack of synthetic usefulness.

In spite of the above adverse results, the preparation of the alkylidene bisphthalisoimides **33** by reaction with alkylenediamines **31** was attempted successfully. The diamines **31** (Scheme 6) gave the corresponding bisphthalisoimides **33** in nearly quantitative yield. This apparently discordant result can be explained if the relative nucleophilicity of monoamines **26** and that of diamines **31** is considered. Nucleophilicity of amines is roughly correlated to basicity which is remarkably lower in alkylidenediamines than in alkylamines,[25] i.e. comparing pKa values, ethylenediamine is closer to aniline than to ethylamine. Because of the -I effect, the introduction of a second amino group decreases the base strength of diamines **31**. It is reasonable to assume a similar effect of the nitrogen atom of the imino group lowering the basicity of the amino group of the intermediate **32**. Thus, the attack of diamines **31** to the final products **33** as well as the similar reactions of intermediates **32** would be considerably prevented. Therefore, useful reactions just as the reactions with arylamines occurred. Bisphthalisoimides **33** were thermally converted into bispthalimides **34** in nearly quantitative yield.



Scheme 6

EXPERIMENTAL

General. NMR spectra were determined on Bruker AC-200 or Varian AC-300 Unity instruments with tetramethylsilane as internal reference. Electron-impact mass spectra were obtained on a Hewlett-Packard 5995 spectrometer with direct insertion probe and an ionizing voltage of 70 eV. IR spectra (nujol emulsion) were recorded on a Nicolet-5DX spectrophotometer. Microanalyses were performed on Perkin-Elmer 240 or Carlo Erba EA-1108 analyzers. All melting points were determined on a Kofler hot-plate melting point apparatus and are uncorrected. N-Arylphthalisoimides[6] **3a**, **3c**, **3e**; N-pyridylphthalimides[26] **4**, **5**; Phthaloperinone[27] **22**; N-alkylphthalisoimides[21] **27**; N,N'-dialkylphthalamides[21,28] **28**, **30**; and N-alkyl-phthalamic acids[21,29] **29**, were prepared by conventional procedures and were used as standard reference materials.

X-Ray Crystallographic Analysis of 3b

Crystal Data. $C_{15}H_{11}NO_3$; Fwt.. 253.25; monoclinic; $P2_1/c$; a = 8.0302(10) Å, b = 20.256(2) Å, c = 7.4859(10) Å, β = 104.708(8)°; V = 1177.7(2) Å^3; Z = 4, D_{calc} 1.428 g/cm^{-3}; crystal size 0.76 x 0.56 x 0.18 mm; F_{000} 528; μ(MoKa) 0.101 mm^{-1}.

Data Collection. A yellow prism of **3b** was mounted in inert oil on a glass fibre and transferred to the diffractometer (Siemens P4 with LT2 low-temperature attachment). Unit cell parameters were determined from a least-squares fit of 69 accurately centered reflections ($8.8 < 2\theta < 24.9$). A total of 4375 intensity data were collected at 173(2) K with graphite monochromated Mo-Kα radiation (λ = 0.71073 Å) to $2\theta_{max} = 50°$. Merging equivalents gave 2068 unique data (R_{int}= 0.023), which were used for calculations.

Structure Solution and Refinement. The structure was solved by direct methods and refined anisotropically on F^2 (program SHELXTL).[30] Non hydrogen atoms were refined anisotropically. Hydrogen atoms were included using a riding model or as rigid methyl group. The final R(F) was 0.0302, for 173 parameters and 1687 observed reflections [$I > 2\sigma(I)$] and wR(F^2) was 0.0784 for all data. The weighting scheme was $w^{-1} = \sigma^2(F^2) + (aP)^2 + bP$, where $3P = (2F_c^2 + F_o^2)$ and a and b are constants adjusted by the program. Maximum Δ/σ = 0.0001, maximum $\Delta\rho$ = 0.17 eÅ^3.

X-Ray Crystallographic Analysis of 15a

Crystal Data. $C_{15}H_{11}NO_3$; Fwt.. 253.25; orthorhombic; $P2_12_12_1$; a = 7.775(2) Å, b = 11.406(2) Å, c = 13.288(2) Å; V = 1178.4(4) Å^3; Z = 4, D_{calc} 1.427 g/cm^{-3}; crystal size 0.52 x 0.34 x 0.26 mm; F_{000} 528; μ(MoKα) 0.101 mm^{-1}.

Data Collection. A colorless prism of **15a** was mounted on a glass fibre and transferred to the

diffractometer (Siemens P4). Unit cell parameters were determined from a least-squares fit of 72 accurately centered reflections ($13.3 < 2\theta < 24.9$). A total of 6043 intensity data were collected at room temperature with graphite monochromated Mo-Kα radiation ($\lambda = 0.71073$ Å) to $2\theta_{max} = 50^{o}$. Merging equivalents gave 2079 unique data ($R_{int} = 0.023$), which were used for calculations.

Structure Solution and Refinement. The structure was solved by direct methods and refined anisotropically on F^2 (program SHELXTL).[30] Non hydrogen atoms were refined anisotropically. Hydrogen atoms were included using a riding model or as a rigid hydroxide group. Possible hydrogen bond O2-H2···N(x-0.5, 0.5-y, -z); O···N 2.644(2) Å, H···N1.835(2) Å, O-H···N 161.2(3)°.The final R(F) was 0.0300, for 173 parameters and 1881 obseved reflections [$I > 2\sigma(I)$] and $wR(F^2)$ was 0.0615 for all data. The weighting scheme was $w^{-1} = \sigma^2(F^2) + (aP)^2 + bP$, where $3P = (2F_c^2 + F_o^2)$ and a and b are constants adjusted by the program. Maximum $\Delta/\sigma = 0.0001$, maximum $\Delta\rho = 0.12$ eÅ^3.

Typical preparative procedure

A solution of the appropriate amine (10 mmol) or diamine (5 mmol) and triethylamine (20 mmol) in acetonitrile (20 mL) was added dropwise within 10 min to a vigorously stirred cooled solution (-10 °C) of phthalidylidene dichloride (10 mmol) in acetonitrile (20 mL). The temperature was maintained between -10 °C and -5 °C. A precipitate was formed after a few drops of mixture were added. The suspension was allowed to warm up to room temperature with continued stirring for 5 min. The solvent was removed under reduced pressure and the solid crude product was shaken twice with ice-cold water and collected and dried by vacuum filtration. High purity crude products[20] were isolated in nearly quantitative yield, were crystallized from the appropriate solvent and were identified by i.r., m.s., high field n.m.r. spectroscopy[31] and microanalyses.

The reaction with 1,8-diaminonaphthalene gave a mixture of products **22** and **23** . The solid was washed with warm chloroform. Product **23** (65%) remained as an insoluble bright yellow crystalline material. Product **22** (32%) was isolated by removing chloroform and was crystallized from acetonitrile.

The reactions with aminopyridines, 2-nitroaniline and aminobenzylalcohols were also carried out following the general procedure described.

N-Phenylphthalisoimide (**3a**)

(88%), pale yellow needles (pet ether) mp 121-122 °C. (Lit[32], mp 120-122 °C). (Found: C, 75.41; H, 4.05; N, 6.29. $C_{14}H_9NO_2$ requires: C, 75.33; H, 4.06; N, 6.27); ^{1}H n.m.r. δ ($CDCl_3$, 200 MHz): 7.20 - 7.26 (m, 1H), 7.37 - 7.44 (m, 4H), 7.72 (td, 1H, Jt = 7.4, Jd = 1.3), 7.82 (td, 1H, Jt = 7.4, Jd = 1.2), 7.95 (d, 1H, J = 7.3), 8.07 (d, 1H, J = 7.5); ^{13}C n.m.r. δ ($CDCl_3$, 50.3 MHz): 123.57, 124.41, 125.21, 126.30, 127.62, 128.82, 133.02, 135.32, 136.98, 143.75, 146.89, 164.81; m.s., m/z (%): 223 (M^+, 49), 179 (78), 104 (29), 76 (100), 51 (36), 50

(60); i.r.: 1789, 1707, 1591, 1489, 1219, 934, 779, 766, 694 cm^{-1}.

N-(2-Methoxyphenyl)phthalisoimide (**3b**)

(86%), yellow needles (aq acetone) mp 123-125 °C. (Lit[1], mp 121 °C). (Found: C, 71.06; H, 4.35; N, 5.49. $C_{15}H_{11}NO_3$ requires: C, 71.14; H, 4.38; N, 5.53); ^1H n.m.r. δ ($CDCl_3$, 300 MHz): 3.86 (s, 3H), 6.95 - 7.01 (m, 2H), 7.16 - 7.25 (m, 2H), 7.75 (td, 1H, Jt = 7.5, Jd = 0.9), 7.84 (td, 1H, Jt = 7.5, Jd = 1.2), 7.96 (d, 1H, J = 7.5), 8.16 (d, 1H, J = 7.5); ^{13}C n.m.r. δ ($CDCl_3$, 75.4 MHz): 55.76, 111.53, 120.59, 123.24, 123.87, 125.18, 126.68, 128.05, 133.08, 133.64, 135.28, 136.55, 148.24, 151.41, 164.69; m.s., m/z (%): 253 (M^+, 14), 180 (16), 179 (16), 130 (19), 104 (53), 76 (100), 51 (48), 50 (70); i.r.: 1800, 1715, 1495, 1244, 1213, 924, 765, 702 cm^{-1}.

N-(4-Methoxyphenyl)phthalisoimide (**3c**)

(85%), yellow needles (aq acetone) mp 135-136 °C. (Lit[1], mp 134 °C). (Found: C, 71.26; H, 4.37; N, 5.57. $C_{15}H_{11}NO_3$ requires: C, 71.14; H, 4.38; N, 5.53); ^1H n.m.r. δ ($CDCl_3$, 200 MHz): 3.82 (s, 3H), 6.91 (d, 2H, J = 8.9), 7.54 (d, 2H, J = 8.9), 7.68 (td, 1H, Jt = 7.4, Jd = 1.0), 7.79 (td, 1H, Jt = 7.4, Jd = 1.2), 7.93 (d, 1H, J = 7.5), 8.03 (d, 1H, J = 7.4); ^{13}C n.m.r. δ ($CDCl_3$, 75.4 MHz): 55.43, 114.13, 123.35, 125.18, 127.29, 127.46, 132.57, 135.25, 136.58, 137.54, 145.16, 158.54, 165.21; m.s., m/z (%): 253 (M^+, 25), 238 (20), 130 (47), 106 (52), 104 (36), 76 (100), 63 (41), 51 (36, 50 (78); i.r.: 1801, 1694, 1505, 1275, 1250, 1213, 922, 829, 773, 696 cm^{-1}.

N-(2-Bromophenyl)phthalisoimide (**3d**)

(82%), pale yellow needles (aq acetone) mp 135-137 °C. (Found: C, 55.76; H, 2.65; N, 4.64. $C_{14}H_8BrNO_2$ requires: C, 55.66; H, 2.67; N, 4.64); ^1H n.m.r. δ ($CDCl_3$, 200 MHz): 7.06 (td, 1H, Jt = 7.2, Jd = 1.9), 7.24 - 7.37 (m, 2H), 7.62 (d, 1H, J = 7.9), 7.74 - 7.91 (m, 2H), 7.98 (d, 1H, J = 7.5), 8.16 (d, 1H, J = 7.3); ^{13}C n.m.r. δ ($CDCl_3$, 50.3 MHz): 117.34, 123.16, 124.01, 125.41, 126.63, 127.78, 128.00, 132.88, 133.58, 135.55, 136.22, 143.40, 148.63, 164.25; m.s., m/z (%): 301 (M^+, 2), 222 (51), 166 (12), 104 (25), 90 (19), 76 (100), 75 (34), 50 (91); i.r.: 1824, 1800, 1705, 1276, 1222, 916, 761, 697 cm^{-1}.

N-(2-Iodophenyl)phthalisoimide (**3e**)

(87%), pale yellow needles (acetonitrile) mp 146-148 °C. (Found: C, 47.99; H, 2.32; N, 4.00. $C_{14}H_8INO_2$ requires: C, 48.16; H, 2.31; N, 4.01); ^1H n.m.r. δ ($CDCl_3$, 200 MHz): 6.91 (td, 1H, Jt = 7.0, Jd = 1.9), 7.26 - 7.41 (m, 2H), 7.74 - 7.90 (m, 3H), 7.97 (d, 1H, J = 7.6 Hz), 8.16 (d, 1H, J = 7.3); ^{13}C n.m.r. δ ($CDCl_3$, 75.4 MHz): 94.17, 122.59, 124.11, 125.53, 127.17, 127.93, 128.85, 133.63, 135.69, 136.47, 139.11, 146.05, 148.23, 164.46; m.s., m/z (%): 349 (M^+, 3), 222 (39), 166 (17), 104 (21), 76 (100), 64 (27), 63 (30), 50 (88); i.r.: 1794, 1707, 1267, 1221, 1103, 932, 774, 762, 700 cm^{-1}

N-(4-Nitrophenyl)phthalisoimide (**3f**)

(83%), pale yellow prisms (acetonitrile) mp 172-173 °C. (Lit[1], mp 172 °C). (Found: C, 62.72; H, 2.99; N, 10.41. $C_{14}H_8N_2O_4$ requires: C, 62.69; H, 3.01; N, 10.44); ^{1}H n.m.r. δ (CD_2Cl_2, 200 MHz): 7.40 (d, 2H, J = 8.9), 7.81 - 7.96 (m, 2H), 8.02 (d, 1H, J = 7.8), 8.11 (d, 1H, J = 7.3), 8.26 (d, 2H, J = 8.9); ^{13}C n.m.r. δ (CD_2Cl_2, 50.3 MHz): 124.16, 124.39, 124.99 , 126.02, 128.27, 134.52, 136.28, 136.53, 145.66, 150.04, 150.96, 164.37; m.s., m/z (%): 268 (M^+, 12), 166 (13), 130 (18), 104 (52), 76 (100), 63 (30), 50 (64); i.r.: 1842, 1721, 1589, 1526, 1514, 1346, 1223, 1103, 918, 862, 843, 708 cm^{-1}.

N-(4-Methylphenyl)phthalisoimide (**3g**)

(79%), pale yellow needles (aq acetone) mp 123-125 °C. (Lit[1], mp 123 °C). (Found: C, 76.02; H, 4.66; N, 5.88. $C_{15}H_{11}NO_2$ requires: C, 75.94; H, 4.67; N, 5.90); ^{1}H n.m.r. δ ($CDCl_3$, 200 MHz): 2.38 (s, 3H), 7.21 (d, 2H, J = 8.3), 7.40 (d, 2H, J = 8.3), 7.72 (td, 1H, Jt = 7.3, Jd = 1.1), 7.83 (td, 1H, Jt = 7.3, Jd = 1.0), 7.96 (d, 1H, J = 7.5), 8.07 (d, 1H, J = 7.5); ^{13}C n.m.r. δ ($CDCl_3$, 50.3 MHz): 21.06, 123.47, 124.93, 125.17, 127.53, 129.47, 132.78, 135.24, 136.52, 137.23, 141.04, 146.24, 164.99; m.s., m/z (%): 237 (M^+, 47), 193 (55), 192 (28), 165 (15), 104 (39), 91 (19), 90 (18), 89 (18), 76 (100), 50 (57); i.r.: 1795, 1709, 1508, 1275, 1215, 1098, 920, 812, 774, 698 cm^{-1}.

N-(4-Bromophenyl)phthalisoimide (**3h**)

(87%), pale yellow needles (aq acetone) mp 165-167 °C. (Lit[33], mp 166-167 °C). (Found: C, 55.74; H, 2.66; N, 4.61. $C_{14}H_8BrNO_2$ requires: C, 55.66; H, 2.67; N, 4.64); ^{1}H n.m.r. δ ($CDCl_3$, 200 MHz): 7.31 (d, 2H, J = 8.7), 7.50 (d, 2H, J = 8.7), 7.76 (td, 1H, Jt = 7.3, Jd = 1.0), 7.85 (td, 1H, Jt = 7.3, Jd = 0.9), 7.97 (d, 1H, J = 7.4), 8.07 (d, 1H, J = 7.3); ^{13}C n.m.r. δ ($CDCl_3$, 50.3 MHz): 119.95, 123.69, 125.41, 126.33, 127.61, 131.97, 133.26, 135.49, 136.89, 142.76, 147.48, 164.55; m.s., m/z (%): 301 (M^+, 3), 178 (13), 104 (31), 76 (100), 63 (22), 50 (72); i.r.: 1817, 1701, 1487, 1277, 1215, 1096, 1073, 917, 722, 700 cm^{-1}.

N-(2-Methylphenyl)phthalisoimide (**3i**)

(86%), pale yellow needles (aq acetone) mp 136-137 °C. (Lit[1], mp 136 °C). (Found: C, 76.11; H, 4.69; N, 5.94. $C_{15}H_{11}NO_2$ requires: C, 75.94; H, 4.67; N, 5.90); ^{1}H n.m.r. δ ($CDCl_3$, 200 MHz): 2.32 (s, 3H), 7.05 - 7.16 (m, 1H), 7.20 - 7.25 (m, 3H), 7.73 (t, 1H, J = 7.4), 7.83 (t, 1H, J = 7.3), 7.95 (d, 1H, J = 7.6), 8.09 (d, 1H, J = 7.4); ^{13}C n.m.r. δ ($CDCl_3$, 50.3 MHz): 18.03, 121.97, 123.53, 125.18, 125.70, 126.14, 127.88, 130.32, 131.55, 133.05, 135.29, 136.66, 142.92, 146.61, 164.77; m.s., m/z (%): 237 (M^+, 29), 219 (50), 193 (41), 165 (17), 104 (39), 90 (24), 89 (28), 76 (100), 51 (37), 50 (64); i.r.: 1791, 1709, 1489, 1273, 1227, 926, 772, 756, 739, 698 cm^{-1}.

N-(2,4-Dichlorophenyl)phthalisoimide (**3j**)

(87%), pale yellow needles (aq acetone) mp 118-120 °C. (Found: C, 57.72; H, 2.40; N, 4.83. $C_{14}H_7Cl_2NO_2$ requires: C, 57.56; H, 2.42; N, 4.79); ^{1}H n.m.r. δ ($CDCl_3$, 200 MHz): 7.24 (br s, 2H), 7.44 (s, 1H), 7.76 - 7.92 (m, 2H), 7.99 (d, 1H, J = 7.4), 8.13 (d, 1H, J = 7.2); ^{13}C n.m.r. δ ($CDCl_3$, 50.3 MHz): 124.07, 124.24, 125.51, 127.35, 127.94, 128.44, 129.62, 131.24, 133.73, 135.62, 136.08, 140.63, 149.30, 163.98; m.s., m/z (%): 291 (M^+, 4), 256 (32), 124 (18), 104 (25), 76 (100), 75 (29), 50 (71); i.r.: 1826, 1712, 1378, 1258, 1226, 1103, 920, 867, 699 cm^{-1}.

N-(4-Chlorophenyl)phthalisoimide (**3k**)

(84%), pale yellow needles (aq acetone) mp 159-160 °C. (Lit[1], mp 160 °C). (Found: C, 64.98; H, 3.11; N, 5.40. $C_{14}H_8ClNO_2$ requires: C, 65.26; H, 3.13; N, 5.44); ^{1}H n.m.r. δ ($CDCl_3$, 200 MHz): 7.31 - 7.37 (m, 4H), 7.73 (td, 1H, Jt = 7.3, Jd = 1.0), 7.82 (td, 1H, Jt = 7.4, Jd = 1.3), 7.95 (d, 1H, J = 7.3), 8.04 (d, 1H, J = 7.3); ^{13}C n.m.r. δ ($CDCl_3$, 50.3 MHz): 123.71, 125.43, 126.13, 127.62, 129.04, 132.04, 133.28, 135.52, 136.96, 142.30, 147.45, 164.65; m.s., m/z (%): 257 (M^+, 19), 213 (29), 178 (23), 104 (30), 76 (100), 50 (66); i.r.: 1818, 1703, 1491, 1279, 1213, 1096, 918, 826, 772, 702 cm^{-1}.

N-(2-Naphthyl)phthalisoimide (**3l**)

(91%), yellow leaves (aq acetone) mp 151-152 °C. (Found: C, 78.98; H, 4.06; N, 5.15. $C_{18}H_{11}NO_2$ requires: C, 79.11; H, 4.06; N, 5.13); ^{1}H n.m.r. δ ($CDCl_3$, 200 MHz): 7.42 -8.06 (m, 11H); ^{13}C n.m.r. δ ($CDCl_3$, 75.4 MHz): 122.70, 123.50, 123.80, 125.15, 125.89, 126.29, 127.45, 127.53, 128.11, 128.53, 131.81, 132.93, 133.53, 135.25, 136.98, 141.21, 147.00, 164.86; m.s., m/z (%): 273 (M^+, 17), 229 (12), 228 (8), 140 (25), 127 (43), 126 (92), 104 (23), 76 (100), 50 (43); i.r.: 1790, 1701, 1277, 1248, 1211, 926, 828, 777, 702 cm^{-1}.

N-(1-Naphthyl)phthalisoimide (**3m**)

(89%), yellow needles (aq acetone) mp 132-133 °C. (Lit[3], mp 132-133 °C). (Found: C, 79.38; H, 4.03; N, 5.11. $C_{18}H_{11}NO_2$ requires: C, 79.11; H, 4.06; N, 5.13); ^{1}H n.m.r. δ ($CDCl_3$, 200 MHz): 7.46 - 7.54 (m, 4H), 7.67 - 7.75 (m, 2H), 7.79-7.86 (m, 2H), 7.95 (d, 1H, J = 7.4), 8.19 (d, 1H, J = 7.5), 8.28 - 8.32 (m, 1H); ^{13}C n.m.r. δ ($CDCl_3$, 50.3 MHz): 118.95, 123.76, 125.39, 125.72, 126.06, 126.30, 126.39, 127.92, 127.97, 129.09, 133.24, 134.11, 135.48, 136.98, 140.06, 147.69, 165.02; m.s., m/z (%): 273 (M^+, 16), 229 (20), 228 (24), 140 (29), 114 (26), 76 (100), 50 (47); i.r.: 1789, 1699, 1258, 1103, 924, 793, 772, 766, 700 cm^{-1}.

N-(2-Terbutylphenyl)phthalisoimide (**3n**)

(92%), pale yellow leaves (aq acetone) mp 114-115 °C. (Found: C, 77.08; H, 6.12; N, 5.05. $C_{18}H_{17}NO_2$ requires: C, 77.40; H, 6.13; N, 5.01); ^{1}H n.m.r. δ ($CDCl_3$, 300 MHz): 1.44 (s, 9H), 7.13 - 7.23 (m, 3H), 7.43

(d, 1H, J = 7.8), 7.77 (t, 1H, J = 7.2), 7.87 (t, 1H, J = 7.5), 7.99 (d, 1H, J = 7.2), 8.10 (d, 1H, J = 7.5); ^{13}C n.m.r. δ($CDCl_3$, 75.4 MHz): 30.16, 35.43, 123.53, 123.62, 125.34, 125.83, 126.32, 126.37, 128.00, 133.09, 135.41, 136.94, 142.68, 143.14, 145.43, 165.10; m.s., m/z (%):279 (M^+, 26), 264 (12), 246 (43), 220 (100), 133 (34), 104 (35), 76 (47); i.r.: 1788, 1713, 1485, 1272, 1218, 1100, 927, 760, 696 cm^{-1}.

N-(2-Pyridyl)phthalimide (**4**)

(84%), white needles (acetonitrile) mp 225-226 °C. (Lit[26], mp 225-226 °C). (Found: C, 69.36; H, 3.63; N, 12.53. $C_{13}H_8N_2O_2$ requires: C, 69.64; H, 3.60; N, 12.49); ^1H n.m.r. δ (DMSO-d_6, 200 MHz): 7.51 - 7.59 (m, 2H), 7.92 - 8.06 (m, 5H), 8.66 (dd, 1H, J = 4.8, J = 1.5); ^{13}C n.m.r. δ (DMSO-d_6, 50.3 MHz): 122.99, 123.59, 124.00, 131.28, 134.91, 138.60, 145.83, 149.32, 166.39; m.s., m/z (%): 224 (M^+, 23), 196 (63), 168 (15), 104 (47), 76 (100), 50 (63); i.r.: 1714, 1588, 1441, 1115, 1086, 883, 781, 717, 646 cm^{-1}.

N-(4-Pyridyl)phthalimide (**5**)

(77%), white needles (acetonitrile) mp 230-231 °C. (Lit[26], mp 231-232 °C). (Found: C, 69.52; H, 3.59; N, 12.45. $C_{13}H_8N_2O_2$ requires: C, 69.64; H, 3.60; N, 12.49); ^1H n.m.r. δ ($CDCl_3$, 200 MHz): 7.62 (d, 2H, J = 1.6), 7.64 (d, 2H, J = 1.6), 7.82 - 7.87 (m, 2H), 7.98 - 8.02 (m, 2H); ^{13}C n.m.r. δ ($CDCl_3$, 50.3 MHz): 119.38, 124.15, 131.40, 134.97, 139.83, 150.67, 166.28; m.s., m/z (%): 224 (M^+, 28), 180 (34), 104 (38), 76 (100), 50 (69); i.r.: 1713, 1588, 1373, 1227, 1107, 1065, 883, 806, 725, 620 cm^{-1}.

2-(4H-3,1-Benzoxazin-2-yl)benzoic acid (**15a**)

(78%), pale brown needles (acetonitrile) mp 178-179 °C. (Found: C, 71.35; H, 4.41; N, 5.51. $C_{15}H_{11}NO_3$ requires: C, 71.14; H, 4.38; N, 5.53); ^1H n.m.r. δ (DMSO-d_6, 200 MHz): 5.35 (s, 2H), 7.14 - 7.36 (m, 4H), 7.61 - 7.65 (m, 2H), 7.74 - 7.80 (m, 2H), 13.17 (br s, 1H); ^{13}C n.m.r. δ (DMSO-d_6, 75.4 MHz): 66.51, 122.43, 123.72, 124.36, 126.64, 128.78, 129.53, 130.81, 131.03, 132.95, 133.58, 139.14, 158.07, 168.94; m.s., m/z (%): 253 (M^+, 6), 209 (75), 149 (40), 105 (100), 104 (69), 77 (46), 76 (39), 51 (37), 50 (31); i.r.: 1705, 1622, 1607, 1491, 1323, 1222, 799, 770, 733, 691 cm^{-1}.

2-(6-Methyl-4H-3,1-benzoxazin-2-yl)benzoic acid (**15b**)

(86%), pale brown needles (acetonitrile) mp 180-182 °C. (Found: C, 71.66; H, 4.88; N, 5.22. $C_{16}H_{13}NO_3$ requires: C, 71.90; H, 4.90; N, 5.24); ^1H n.m.r. δ (DMSO-d_6, 200 MHz): 2.30 (s, 3H), 5.31 (s, 2H), 6.95 (s, 1H), 7.04 - 7.15 (m, 2H), 7.59 - 7.64 (m, 2H), 7.73 - 7.79 (m, 2H), 13.16 (br s, 1H); ^{13}C n.m.r. δ (DMSO-d_6, 50.3 MHz): 20.82, 66.53, 122.32, 123.77, 124.81, 128.82, 129.22, 129.58, 130.75, 130.99, 132.89, 133.71, 136.18, 136.79, 157.70, 169.12; m.s., m/z (%): 267 (M^+, 40), 223 (100), 149 (46), 119 (52), 118 (62), 105 (87), 92 (53), 91 (54), 77 (48), 76 (65), 65 (92), 51 (44), 50 (51); i.r.: 1712, 1625, 1601, 1317, 1218, 1131, 820,

771, 735, 673 cm^{-1}.

N,N'-(1,2-Phenylene)bisphthalisomide (**16**)

(90%), yellow needles (acetonitrile) mp 190-192 °C. (Lit[6], mp 185-187 °C). (Found: C, 71.98; H, 3.30; N, 7.60. $C_{22}H_{12}N_2O_4$ requires: C, 71.74; H, 3.28; N, 7.61); ^1H n.m.r. δ ($CDCl_3$, 200 MHz): 7.26 - 7.30 (m, 2H), 7.32 - 7.44 (m, 2H), 7.65 - 7.81 (m, 4H), 7.85 - 8.01 (m, 4H); ^{13}C n.m.r. δ ($CDCl_3$, 50.3 MHz): 123.42, 123.59, 125.25, 126.31, 127.84, 133.15, 135.31, 136.55, 137.59, 147.99, 164.50; m.s., m/z (%): 368 (M^+, 2), 220 (10), 164 (6), 104 (42), 76 (100), 50 (49); i.r.: 1811, 1715, 1697, 1275, 1225, 927, 902, 763, 700 cm^{-1}.

N,N'-(1,2-Phenylene)bisphthalimide (**17**)

(94%), pale brown needles (acetonitrile) mp 307-309 °C. (Lit[6], mp 299-301 °C). (Found: C, 71.99; H, 3.26; N, 7.59. $C_{22}H_{12}N_2O_4$ requires: C, 71.74; H, 3.28; N, 7.61); ^1H n.m.r. δ ($CDCl_3$, 300 MHz):7.50 - 7.55 (m, 2H), 7.57 - 7.62 (m, 2H), 7.67 - 7.73 (m, 4H), 7.79 - 7.85 (m, 4H); ^{13}C n.m.r. δ($CDCl_3$, 75.4 MHz): 123.79, 128.86, 129.46, 129.60, 131.49, 134.29, 166.22; m.s., m/z (%): 368 (M^+, 95), 323 (21), 296 (30), 104 (33), 76 (100), 50 (32); i.r.: 1732, 1504, 1387, 1230, 1069, 884, 762, 723 cm^{-1}.

N,N'-(1,4-Phenylene)bisphthalisomide (**18**)

(91%), yellow needles (DMF) mp 292-294 °C. (Lit[6], mp 296-298 °C). (Found: C, 72.00; H, 3.27; N, 7.63. $C_{22}H_{12}N_2O_4$ requires: C, 71.74; H, 3.28; N, 7.61; n.m.r. spectra could not be recorded due to its extremely low solubility in the usual spectroscopic solvents; m.s., m/z (%): 368 (M^+, 5), 221 (10), 177 (9), 104 (42), 76 (100), 50 (39); i.r.: 1814, 1694, 1504, 1275, 1221, 1096, 911, 837, 772, 701 cm^{-1}.

N,N'-(1,5-Naphthylene)bisphthalisoimide (**19**)

(78%), yellow leaves (DMF) mp > 330 °C. (Found: C, 74.53; H, 3.38; N, 6.72. $C_{26}H_{14}N_2O_4$ requires: C, 74.64; H, 3.37; N, 6.70); n.m.r. spectra could not be recorded due to its extremely low solubility in the usual spectroscopic solvents; m.s., m/z (%): 418 (M^+, 13), 130 (12), 104 (42), 76 (100), 50 (41); i.r.: 1786, 1697, 1277, 1255, 1100, 943, 907, 779, 702 cm^{-1}.

N,N'-(1,8-Naphthylene)bisphthalisoimide (**23**)

(65%), yellow needles (chloroform-dichloromethane) mp 255-256 °C. (Found: C, 74.79; H, 3.37; N, 6.68. $C_{26}H_{14}N_2O_4$ requires: C, 74.64; H, 3.37; N, 6.70); n.m.r. spectra could not be recorded due to its extremely low solubility in the usual spectroscopic solvents; m.s., m/z (%): 418 (M^+, 24), 272 (40), 166 (22), 104 (27), 76 (100), 50 (31); i.r.: 1788, 1699, 1256, 1107, 922, 775, 768, 702 cm^{-1}.

N,N'-Ethylenebisphthalisoimide (**33a**)

(92%), white needles (ether-pet ether) mp 232-236 °C. (Found: C, 67.19; H, 3.77; N, 8.78. $C_{18}H_{12}N_2O_4$ requires: C, 67.50; H, 3.78; N, 8.75); ^{1}H n.m.r. δ ($CDCl_3$, 300 MHz): 4.08 (s, 4H), 7.71 (td, 2H, Jt = 8.1, Jd = 1.2), 7.78 (td, 2H, Jt = 7.5, Jd = 1.2), 7.92 - 7.98 (m, 4H); ^{13}C n.m.r. δ ($CDCl_3$, 75.4 MHz): 49.33, 123.29, 125.16, 128.30, 132.68, 135.23, 136.40, 149.43, 164.71; m.s., m/z (%): 320 (M^+, 1), 160 (100), 133 (9), 104 (11), 76 (13), 50 (7); i.r.: 1797, 1789, 1712, 1277, 1221, 1110, 1008, 917, 781, 706 cm^{-1}.

N,N'-Propylenebisphthalisoimide (**33b**)

(89%), white needles (pet ether) mp 148-150 °C. (Found: C, 68.57; H, 4.19; N, 8.40. $C_{19}H_{14}N_2O_4$ requires: C, 68.26; H, 4.22; N, 8.38); ^{1}H n.m.r. δ ($CDCl_3$, 200 MHz): 2.17 (qui, 2H, J = 6.7), 3.87 (t, 4H, J = 6.7), 7.70 (td, 2H, Jt = 7.2, Jd = 1.1), 7.79 (td, 2H, Jt = 7.4, Jd = 1.4), 7.89 - 7.97 (m, 4H); ^{13}C n.m.r. δ ($CDCl_3$, 50.3 MHz): 31.00, 46.56, 123.01, 125.05, 128.20, 132.52, 135.17, 136.36, 148.65, 164.72; m.s., m/z (%): 334 (M^+, 7.5), 187 (20), 174 (100), 161 (93), 160 (62), 132 (45), 130 (48), 104 (85), 76 (67), 50 (38); i.r.: 1817, 1713, 1471, 1266, 1116, 918, 778, 699 cm^{-1}.

N,N'-Ethylenebisphthalimide (**34a**)

(96%), pale brown needles (acetonitrile) mp 237-240 °C. (Found: C, 67.54; H, 3.75; N, 8.77. $C_{18}H_{12}N_2O_4$ requires: C, 67.50; H, 3.78; N, 8.75); ^{1}H n.m.r. δ ($CDCl_3$, 200 MHz): 4.01 (s, 4H), 7.66 -7.73 (m, 4H), 7.75 - 7.81 (m, 4H); ^{13}C n.m.r. δ ($CDCl_3$, 50.3 MHz): 36.74, 123.26, 131.85, 133.92, 168.12; m.s., m/z (%): 320 (M^+, 1), 173 (100), 160 (86), 133 (17), 104 (26), 77 (30), 76 (26); i.r.: 1714, 1445, 1403, 1062, 881, 722 cm^{-1}.

N,N'-Propylenebisphthalimide (**34b**)

(92%), yellow plates (acetonitrile) mp 201-203 °C. (Found: C, 68.33; H, 4.24; N, 8.42. $C_{19}H_{14}N_2O_4$ requires: C, 68.26; H, 4.22; N, 8.38); ^{1}H n.m.r. δ ($CDCl_3$, 200 MHz): 2.11 (qui, 2H, J = 7.3), 3.77 (t, 4H, J = 7.3), 7.69 - 7.76 (m, 4H), 7.80 - 7.86 (m, 4H); ^{13}C n.m.r. δ ($CDCl_3$, 50.3 MHz): 27.61, 35.69, 123.23, 132.02, 133.92, 168.16; m.s., m/z (%): 334 (M^+, 15), 174 (33), 161 (47), 160 (55), 133 (32), 130 (49), 104 (55), 76 (100), 50 (63); i.r.: 1765, 1713, 1435, 1395, 1336, 1177, 1020, 870, 723 cm^{-1}.

Acknowledgement: We gratefully acknowledge the financial support of the Dirección General de Investigación Científica y Técnica (project number PB94-1129). One of the authors (MCRA) thanks Ministerio de Educación y Cultura for a grant.

REFERENCES AND NOTES

1. See for example: Roderick, W.R; Bhatia, P.L. *J. Org. Chem.* **1963**, 28, 2018., and references cited therein.
2. Howe, R.K. *J. Org. Chem.*. **1973**, 38, 4164.
3. Ganin, E.V.; Makarov, V.F.; Nikitin, V.I. *Ukr. Khim. Zh.* **1987**, 53, 964.
4. Takeuchi, H.; Eguchi, H. *J. Chem. Soc., Perkin Trans.* 1, **1988**, 2149.
5. Kukolja, S.; Lammer, S.R. *J. Am. Chem. Soc.* **1975**, 97, 5582.
6. Awad, W.I.; Wasfi, A.S.; Ewad, M.J. *J. Iraqi Chem. Soc.* **1977**, 2, 5.
7. Pyriadi, M.; Hadi, S. *Arab. Gulf J. Sci. Res. A.* **1987**, 5, 341.
8. Roderick, W.R. *J. Org. Chem.* **1964**, 29, 745.
9. Sauers, C.K.; Gould, C.L.; Ioannou, E.S. *J. Am. Chem. Soc.* **1972**, 94, 8156.
10. Au, T.K.; Baydar, A.E.; Boyd, G.V. *J. Chem. Soc., Perkin Trans.* 1, **1981**, 2884.
11. Cotter, R.J.; Sauers, C.K.; Wheland, J.M. *J. Org. Chem.* **1961**, 26, 10.
12. Ernst, M.L.; Schmir, G.L. *J. Am. Chem. Soc.* **1966**, 88, 5001.
13. Curtin, D.Y.; Miller, L.L. *J. Am. Chem. Soc.* **1967**, 89, 637.
14. Erwin, O. *Organic Syntheses;* Blatt, A.H. Ed.; John Wiley and Sons, Inc.: London, 1946; Coll. Vol. II; p. 528.
15. Kunzek, H.; Rühlmann, K. *J. Organometal. Chem.* **1972**, 42, 391.
16. Guirado, A.; Barba, F.; Hursthouse, M.B.; Martínez, A.; Arcas, A. *Tetrahedron Lett.* **1986**, 27, 4063.
17. Naik, S.N.; Pandey, B.; Ayyangar, N.R. *Synthetic Commun.* **1988**, 18, 633.
18. Becher, J.; Simonsen, O.; Sæther H.; Hansen, T.K.; Jørgensen, T.; Bøwadt, S. *Heterocycles* **1993**, 35, 445.
19. Guirado, A.; Zapata, A.; Fenor, M. *Tetrahedron Lett.* **1991**, 32, 2633.
20. i.r. and high field ^{1}H n.m.r. spectra for crude and crystalline products were recorded and showed negligible differences.
21. Boyd, G.V.; Monteil, R.L. *J. Chem. Soc., Perkin Trans. 1.* **1978**, 1338.
22. *Comprehensive Heterocyclic Chemistry*; Katritzky, A.; Rees, C.; Potts, K. Eds. Pergamon Press: Oxford, 1984; Vol. 3; p. 1020
23. *Elsevier's Encyclopaedia of Organic Chemistry*, Radt, F. Ed.; Elsevier Publishing Company: New York, 1949; Series III, Vol. 12B; p. 830.
24. The structure of this compound was kindly confirmed by Prof. P.G. Jones. *Z. Kristallogr.* **1993**, 208, 341.

25. *The Chemistry of the Amino Group*, Patai, S. Ed.; Interscience Publishers: London, 1968; Chap. 4.

26. Suzuki, T.; Kenbou, N.; Mitsuhashi, K. *J. Heterocyclic Chem.* **1979**, 16, 645.

27. Sachs, F. *Liebigs Ann. Chem.* **1909**, 365, 53, 135.

28. Endres, W. *Arch. Pharm.* **1972**, 305, 571.

29. Verbicky, J.W.; Williams, L. *J. Org. Chem.* **1981**, 46, 175

30. SHELXTL, Siemens Analytical X-Ray Instruments, Madison, WI.

31. Since available spectroscopic data (specially ^{13}C n.m.r. data) for phthalisoimides are scarce all recorded spectra are given.

32. Hoogenwerff, S.; van Drop, W.A. *Rec. Trav. Chim.* **1902**, 21, 339.

33. Bhatia, P.L.; Gupta, S.N. *Indian J. Chem.* **1964**, 2, 295.

(*Received in UK* 20 *January* 1997; *revised* 14 *February* 1997; *accepted* 20 *February* 1997)