New method for the synthesis of 2-substituted *N,N'*-diacylimidazolidines

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A method for the synthesis of 2-substituted *N,N'*-diacylimidazolidines was developed. The method is based on the reactions of acylating reagents (carboxylic acid chlorides and anhydrides, sulfonic acid chlorides, a carbamic acid chloride, and ethyl chlorocarbonate) with Schiff's bases prepared by the reaction of *N*-acylethylenediamines with aldehydes.

Key words: *N,N'*-diacylated imidazolidines, Schiff's bases, acylation, cyclization, acid chlorides.

In continuation of the studies into the synthesis of *N,N'*-diacylated imidazolidines (DAI) from *N*-monoacylated derivatives of ethylenediamine[1] (EDA) **1**, we developed a new method for the preparation of 2-substituted DAI **2**. Earlier,[2,3] some of the imidazolidines **2** (but only those with two identical acyl groups) were synthesized by the reaction of acid halides with Schiff's bases based on EDA. The new, much more general method proposed here allows one to obtain compound **2** containing both identical and different acyl fragments in various combinations. According to our method, the title compounds result from the reactions of acylating reagents with Schiff's bases **3** prepared from amines **1** and aliphatic or aromatic aldehydes **4** (Scheme 1, Table 1).

Table 1. Yields, physicochemical constants, and ^{1}H NMR spectral parameters for *N*-acyl-*N'*-alkylidene(arylidene)ethylenediamines $R^1NHCH_2CH_2N{=}CHR^2$ (**3a–i**)

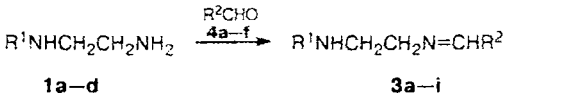
Compound	R^1	R^2	Yield (%)	M.p. /° C	^{1}H NMR ($CDCl_3$), δ (*J*/Hz)
3a	EtCO	*p*-$Me_2NC_6H_4$	65	120–122	1.15 (t, 3 H, Me, *J* = 6.2); 2.0 (q, 2 H, CH_2, *J* = 6.2); 3.00 (s, 6 H, 2 Me); 3.52–3.70 (m, 4 H, 2 CH_2); 6.05 (br.s, 1 H, NH); 6.70, 7.60 (both d, each 2 H, H arom., *J* = 8.6); 8.15 (s, 1 H, CH)
3b	Bz	*p*-$Me_2NC_6H_4$	72	73–75	3.03 (s, 6 H, 2 Me); 3.80 (br.s, 4 H, 2 CH_2); 6.70–7.75 (m, 9 H, H arom.)
3s	Ts	Pr^i	83	—[a]	0.95–1.05 (m, 6 H, 2 Me); 2.10 (m, 1 H, CH); 2.45 (s, 3 H, Me); 2.85–3.35 (m, 4 H, 2 CH_2); 4.35 (d, 1 H, CH, *J* = 6.5); 7.30, 7.70 (both d, each 2 H, H arom., *J* = 8.3); 7.35 (br.s, 1 H, NH)
3d	Ts	Ph	74	98–99	2.45 (s, 3 H, Me); 3.30–3.65 (m, 4 H, 2 CH_2); 4.90 (br.s, 1 H, NH); 7.30–7.65 (m, 9 H, H arom.); 8.20 (s, 1 H, CH)
3e	Ts	*m*-$MeOC_6H_4$	61	44–46	2.45 (s, 3 H, Me); 3.30, 3.60 (both t, each 2 H, 2 CH_2, *J* = 5.4); 3.75 (s, 3 H, MeO); 4.9 (br.s, 1 H, NH); 7.20–7.70 (m, 8 H, H arom.); 8.20 (s, 1 H, CH)
3f	Ts	*p*-$Me_2NC_6H_4$	83	120–122	2.45 (s, 3 H, Me); 3.05 (s, 6 H, 2 MeN); 3.30, 3.60 (both t, each 2 H, 2 CH_2, *J* = 5.4); 4.9 (br.s, 1 H, NH); 6.70, 7.25, 7.50, 7.75 (all d, each 2 H, H arom., *J* = 8.5); 8.05 (s, 1 H, CH)
3g	Ts	*m*-$NO_2C_6H_4$[b]	73	103–105	2.45 (s, 3 H, Me); 3.10, 3.65 (both t, each 2 H, 2 CH_2, *J* = 6.5); 7.32 (d, 2 H, H arom., *J* = 8.4); 7.65–8.25 (m, 5 H, H arom.); 8.35 (s, 1 H, SH); 8.50 (s, 1 H, H arom.)[c]
3h	Ts	*p*-$NO_2C_6H_4$[d]	77	144–146	2.45 (s, 3 H, Me); 3.10, 3.65 (both t, each 2 H, 2 CH_2, *J* = 6.5); 7.35, 7.65 (both d, each 2 H, H arom., *J* = 9.0); 7.95–8.25 (both d, each 2 H, H arom., *J* = 9.6); 8.35 (s, 1 H, CH)[c]
3i	*m*-O_2NTs	Pr^i	85	—[a]	0.95–1.05 (m, 6 H, 2 Me); 2.15 (m, 1 H, CH); 2.65 (s, 3 H, Me); 2.85–3.45 (m, 4 H, 2 CH_2); 4.40 (d, 1 H, CH, *J* = 6.9); 7.50 (s, 1 H, NH); 7.55, 7.95 (both d, each 1 H, H arom., *J* = 8.6); 8.40 (s, 1 H, H arom.)

[a] Oil.
[b] Found (%): C, 55.38; H, 4.90; N, 12.26; S, 8.96. $C_{16}H_{17}N_3O_4S$. Calculated (%): C, 55.32; H, 4.90; N, 12.10; S, 9.23.
[c] The spectrum was recorded in DMSO-d_6.
[d] Found (%): C, 55.44; H, 5.04; N, 12.19; S, 8.90. $C_{16}H_{17}N_3O_4S$. Calculated (%): C, 55.32; H, 4.90; N, 12.10; S, 9.23.

Translated from *Izvestiya Akademii Nauk. Seriya Khimicheskaya*, No. 5, pp. 896–900, May, 2000.

1066-5285/00/4905-0894 $25.00

Scheme 1



1a–d **3a–i**

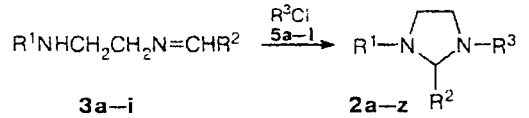
1: R^1 = EtCO (**a**), Bz (**b**), Ts (**c**), *m*-O_2NTs (**d**)
4: R^2 = Pr^i (**a**), Ph (**b**), *m*-$MeOC_6H_4$ (**c**), *p*-$Me_2NC_6H_4$ (**d**), *m*-$NO_2C_6H_4$ (**e**), *p*-$NO_2C_6H_4$ (**f**)
3: R^1 = EtCO (**a**), Bz (**b**), Ts (**c–h**), *m*-O_2NTs (**i**);
R^2 = *p*-$Me_2NC_6H_4$ (**a**, **b**, **f**), Pr^i (**c**, **i**), Ph (**d**), *m*-$MeOC_6H_4$ (**e**), *m*-$NO_2C_6H_4$ (**g**), *p*-$NO_2C_6H_4$ (**h**) (see Table 1)

The acyl substituents were propionyl, benzoyl, tosyl, and *m*-nitrotosyl. Isobutyraldehyde, benzaldehyde, *m*- and *p*-nitrobenzaldehyde, *m*-methoxybenzaldehyde, and *p*-dimethylaminobenzaldehyde were used as the starting aldehydes.

The structure of compounds **3** was mainly confirmed by ^{1}H NMR spectroscopy. Analytically pure samples of **3** were not obtained, except for products **3g**,**h**, because most of the azomethines **3** are unstable and partially decompose during purification. Yields, melting points, and ^{1}H NMR spectral data for compounds **3a–i** are presented in Table 1.

It was established that the reaction of organic acid chlorides **5** with Schiff's bases **3** in the presence of 1 equiv. of Et_3N usually yields a cyclization product, DAI **2** (Scheme 2, Table 2).

Scheme 2



3a–i **2a–z**

5: R^3 = Ac (**a**), EtCO (**b**), $ClCH_2CO$ (**c**), $PhOCH_2CO$ (**d**), Bz (**e**), *p*-$NO_2C_6H_4CO$ (**f**), 3,5-di(methylthio)isothiazol-4-ylcarbonyl (**g**), 1-piperidinocarbonyl (**h**), EtOCO (**i**), Ms (**j**), $PhSO_2$ (**k**), Ts (**l**)
2: R^1 = EtCO (**a**), Bz (**b**, **c**), Ts (**d–y**), *m*-O_2NTs (**z**);
R^2 = *p*-$Me_2NC_6H_4$ (**a–c**, **x**, **y**), Pr^i (**d–m**, **z**), Ph (**n–u**), *p*-$NO_2C_6H_4$ (**v**), *m*-$MeOC_6H_4$ (**w**);
R^3 = Ts (**a**, **c**, **m**, **u–w**, **y**), Bz (**b**, **g**, **q**, **z**), Ac (**d**, **x**), EtCO (**e**, **n**), $ClCH_2CO$ (**f**, **o**), *p*-$NO_2C_6H_4CO$ (**h**), 3,5-di(methylthio)isothiazol-4-ylcarbonyl (**i**), EtOCO (**j**), Ms (**k**, **s**), $PhSO_2$ (**l**, **t**), $PhOCH_2CO$ (**p**), 1-piperidinocarbonyl (**r**) (see Table 2)

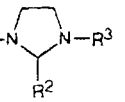
In our experiments, the acylating reagents were carboxylic acid (aliphatic, aromatic, and heterocyclic) chlorides, sulfonic acid chlorides, carbamic acid chlorides, and ethyl chlorocarbonate. Most of the reactions were carried out in acetonitrile. However, a few reactions conducted in benzene gave similar results. DAI **2** can be synthesized from azomethines **3** (which are formed *in situ* in acetonitrile) by treating this solution with Et_3N and acid chloride **5**. This approach is advisable with oily compounds **3**, *e.g.*, **3c**,**i**. The yields and some characteristics of DAI **2** are given in Table 2.

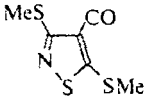
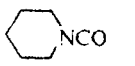
The compounds obtained were characterized by elemental analysis and ^{1}H NMR spectroscopy. The ^{1}H NMR spectra show signals from four protons of DAI in positions 4 and 5, one proton in position 2, and the protons of the corresponding substituents R^1, R^2, and R^3. For R^2 = Pr^i, the protons of DAI in position 2 usually appear as doublets at 5.0–6.5, while for R^2 = Ar, they are singlets in the range from 6.5 (for sulfonamides) to 7.0–7.5 (for carboxamides). When R^1 or R^3 are aliphatic carboxyl radicals, the protons of DAI in position 2 show themselves as two singlets of different intensity for R^2 = Ar (compounds **2a**,**n**,**o**,**x**) and, correspondingly, two doublets for R^2 = Pr^i (compounds **2d**,**e**,**f**). The protons of acyl substituents also exhibit two sets of signals. Although this phenomenon caused by rotational isomerism is known both for aliphatic and aromatic amides, the spectra of compounds **2** containing aroyl groups show no doubling of the corresponding signals. Probably, this can be attributed to steric factors, *i.e.*, the presence of a bulky aromatic ring makes the formation of the second isomer unfavorable. ^{1}H NMR spectral parameters for compounds **2a–z** are presented in Table 3.

The nature of acyl substituent R^1 in azomethine **3** has almost no influence on the course of the reaction with acylating reagents. The reactivity of compound **3** is mainly determined by the character of substituent R^2. For R^2 = Alk, the compounds easily react with various acid chlorides at room temperature to give DAI. The reactivity of azomethines **3** with an aromatic R^2 is decreased. In particular, compound **3d** reacts with carboxylic acid chlorides at ~20 °C, while in the case of sulfonic and carbamic acid chlorides, heating is required. The character of substituents in the aromatic ring of R^2 is significant. The presence of electron-releasing groups in both *meta*- (**3e**) and *para*-positions (**3a**,**b**,**f**) hardly influences the course of reaction, whereas strong electron-withdrawing substituents appreciably hinders the formation of DAI **2**. For example, azomethine **3h** reacts with TsCl and Et_3N in boiling acetonitrile to give product **2v** only in 10% yield, and compound **3g** affords no corresponding DAI at all. In both cases, *N,N'*-bistosylethylenediamine was formed in ~60% yield. In the other reactions, *N,N'*-diacylated EDA was detected in trace amounts or not at all.

Although compounds **3c**,**i** bearing an aliphatic R^2 react with acid chlorides more vigorously, *N,N'*-diacylated EDA are also formed in some amounts, probably, because of the lower stability of these azomethines.

Apart from the reactions with acid chlorides, several reactions of compounds **3** with anhydrides were carried out. It was shown that the reactions of azomethines **3c**,**f** with Ac_2O in acetonitrile at room temperature result in the corresponding DAI **2d**,**x** in high yields (Scheme 3).

Table 2. Yields and physicochemical constants of 2-substituted DAI **2a—z** 

Compound	R^1	R^2	R^3	Yield (%)	M.p. /C	Found / Calculated (%) C	H	N	S	Molecular formula
2a	EtCO	*p*-$Me_2NC_6H_4$	Ts	59	157—159	62.63 62.83	6.81 6.78	10.89 10.47	7.78 7.98	$C_{21}H_{27}N_3O_3S$
2b	Bz	*p*-$Me_2NC_6H_4$	Bz	74	170—172	75.11 75.15	6.51 6.31	—	—	$C_{25}H_{25}N_3O_2$
2c	Bz	*p*-$Me_2NC_6H_4$	Ts	85	158—160	66.34 66.79	6.09 6.06	9.60 9.35	7.13 7.12	$C_{25}H_{27}N_3O_3S$
2d	Ts	Pr^i	Ac	58[a]	140—142	57.66 58.04	7.00 7.14	—	10.52 10.33	$C_{15}H_{22}N_2O_3S$
2e	Ts	Pr^i	EtCO	56	99—101	58.81 59.23	7.08 7.46	—	9.52 9.88	$C_{16}H_{24}N_2O_3S$
2f	Ts	Pr^i	$ClCH_2CO$	42	129—131	52.14 52.24	6.35 6.14	—	9.05[b] 9.30	$C_{15}H_{21}ClN_2O_3S$
2g	Ts	Pr^i	Bz	54	110—111	64.00 64.49	6.75 6.49	7.65 7.52	8.12 8.61	$C_{20}H_{24}N_2O_3S$
2h	Ts	Pr^i	*p*-$NO_2C_6H_4CO$	49	139—141	57.32 57.54	5.54 5.55	—	7.86 7.68	$C_{20}H_{23}N_3O_5S$
2i	Ts	Pr^i		80	155—157	48.46 48.38	5.49 5.34	—	26.63 27.19	$C_{19}H_{25}N_3O_3S_4$
2j	Ts	Pr^i	EtOCO	45	60—62	56.54 56.45	7.31 7.11	8.31 8.23	9.43 9.45	$C_{16}H_{24}N_2O_4S$
2k	Ts	Pr^i	Ms	47	143—146	48.23 48.53	6.31 6.40	—	18.62 18.51	$C_{14}H_{22}N_2O_4S_2$
2l	Ts	Pr^i	$PhSO_2$	52	92—94	56.00 55.87	6.12 5.93	—	15.33 15.68	$C_{19}H_{24}N_2O_4S_2$
2m	Ts	Pr^i	Ts	59	138—140	56.39 56.84	6.01 6.20	6.22 6.63	14.83 15.17	$C_{20}H_{26}N_2O_4S_2$
2n	Ts	Ph	EtCO	59	123—124	63.78 63.66	5.97 6.12	—	9.05 8.93	$C_{19}H_{22}N_2O_3S$
2o	Ts	Ph	$ClCH_2CO$	58	145—148	57.00 57.06	5.02 5.05	—	8.46[c] 8.46	$C_{18}H_{19}ClN_2O_3S$
2p	Ts	Ph	$PhOCH_2CO$	25	159—161	66.06 66.04	5.62 5.54	—	7.30 7.34	$C_{24}H_{24}N_2O_4S$
2q	Ts	Ph	Bz	60	166—168	67.49 67.97	5.83 5.46	6.57 6.89	7.84 7.89	$C_{23}H_{22}N_2O_3S$
2r	Ts	Ph		53	136—138	63.58 63.90	6.97 6.58	—	7.50 7.75	$C_{22}H_{27}N_3O_3S$
2s	Ts	Ph	Ms	53	134—136	53.65 53.67	5.04 5.30	—	16.36 16.85	$C_{17}H_{20}N_2O_4S_2$
2t	Ts	Ph	$PhSO_2$	64	145—147	60.22 59.71	5.18 5.01	—	14.08 14.49	$C_{22}H_{22}N_2O_4S_2$
2u	Ts	Ph	Ts	68	134—136	60.22 60.51	5.11 5.30	—	13.95 14.04	$C_{23}H_{24}N_2O_4S_2$
2v	Ts	*p*-$NO_2C_6H_4$	Ts	10	199—200	54.64 55.08	4.77 4.62		12.37 12.78	$C_{23}H_{23}N_3O_6S_2$
2w	Ts	*m*-$MeOC_6H_4$	Ts	47	112—114	58.83 59.24	5.06 5.39	—	12.82 13.18	$C_{24}H_{26}N_2O_5S$
2x	Ts	*p*-$Me_2NC_6H_4$	Ac	67, 65[a]	157—159	61.59 61.99	6.12 6.50	—	8.63 8.27	$C_{20}H_{25}N_3O_3S$
2y	Ts	*p*-$Me_2NC_6H_4$	Ts	69	174—176	60.35 60.10	5.83 5.85	8.55 8.41	12.81 12.83	$C_{25}H_{29}N_3O_4S_2$
2z	*m*-O_2NTs	Pr^i	Bz	46	161—163	57.19 57.54	5.54 5.55	—	7.65 7.68	$C_{20}H_{23}N_3O_5S$

[a] In the reaction with Ac_2O.
[b] Found: Cl, 9.92%. Calculated: Cl, 9.25%.
[c] Found: Cl, 9.23%. Calculated: Cl, 9.25%.

Table 3. ^{1}H NMR spectra (in $CDCl_3$) of 2-substituted DAI **2a—z**

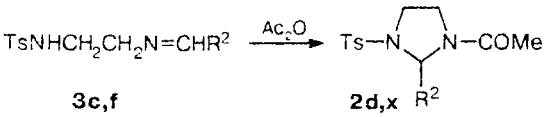
Compound	δ (*J*/Hz)			
	CH_3	CH_2	CH	H arom.
2a	1.00 (m, 3 H); 2.40 (s, 3 H); 2.95, 2.98 (both s, each 3 H, 2 MeN)	1.70—2.00 (m, 2 H); 3.40—4.00 (m, 4 H)	6.30, 6.50 (both s, 1 H)	6.70—7.80 (m, 6 H)
2b	3.00 (s, 6 H, 2 MeN)	3.85—4.50 (m, 4 H)		6.70 (d, 2 H, *J* = 10.0); 7.35—7.45 (m, 13 H, CH + H arom.)
2c	2.45 (s, 3 H); 3.00 (s, 6 H, 2 MeN)	3.45—3.90 (m, 4 H)	6.50 (s, 1 H)	6.70—7.80 (m, 14 H)
2d	1.00 (m, 6 H); 1.55, 1.90 (both s, 3 H); 2.45 (s, 3 H)	2.70—3.90 (m, 4 H)	1.95 (m, 1 H); 5.05, 5.50 (both d, 1 H, *J* = 9.7)	7.30, 7.70 (both d, each 2 H, *J* = 10.3)
2e	0.95—1.05 (m, 9 H); 2.45 (s, 3 H)	1.55—1.96 (m, 2 H); 2.80—3.90 (m, 4 H)	1.95 (m, 1 H); 5.10, 5.50 (both d, 1 H, *J* = 8.9)	7.25, 7.70 (both d, each 2 H, *J* = 11.1)
2f	1.00 (br.s, 6 H); 2.45 (s, 3 H)	2.85—3.95 (m, 6 H)	2.00 (m, 1 H); 5.15, 5.50 (both d, 1 H, *J* = 9.1)	7.30, 7.70 (both d, each 2 H, *J* = 10.0)
2g	1.05—1.15 (m, 6 H); 2.45 (s, 3 H)	2.85—3.85 (m, 4 H)	2.05 (m, 1 H); 6.85 (d, 1 H, *J* = 9.2)	7.10—7.75 (m, 9 H)
2h	1.05—1.15 (m, 6 H); 2.45 (s, 3 H)	2.75—3.90 (m, 4 H)	2.00 (m, 1 H); 5.80 (d, 1 H, *J* = 9.2)	7.25, 7.35, 7.75, 8.50 (all d, each 2 H, *J* = 10.0)
2i	1.10 (m, 6 H); 2.40 (s, 3 H); 2.53, 2.57 (both s, each 3 H, 2 MeS)	3.00—3.60 (m, 4 H)	2.25 (m, 1 H, CH); 5.75 (d, 1 H, *J* = 5.6)	7.35, 7.80 (both d, each 2 H, *J* = 11.0)
2j	1.00 (m, 6 H); 2.45 (s, 3 H)	2.80—3.90 (m, 6 H)	1.90 (m, 1 H); 5.20 (m, 1 H)	7.25, 7.70 (both d, each 2 H, *J* = 9.0)
2k	1.00 (m, 6 H); 2.45 (s, 3 H); 2.75 (s, 3 H, MeS)	3.50—3.75 (m, 4 H)	2.10 (m, 1 H); 5.20 (d, 1 H, *J* = 6.7)	7.40, 7.80 (both d, each 2 H, *J* = 9.2)
2l	1.00 (m, 6 H); 2.45 (s, 3 H)	3.30 (m, 4 H)	2.25 (m, 1 H); 5.30 (d, 1 H, *J* = 5.7)	7.25—7.75 (m, 9 H)
2m	1.00 (d, 6 H, *J* = 7.3); 2.45 (s, 6 H)	3.30 (m, 4 H)	2.30 (m, 1 H); 5.30 (d, 1 H, *J* = 5.6)	7.30, 7.55 (both d, each 4 H, *J* = 10.0)
2n	1.00 (m, 3 H); 2.45 (s, 3 H)	1.70—2.00 (m, 2 H); 3.00—4.00 (m, 4 H)	6.40, 6.70 (both s, 1 H)	7.25—7.80 (m, 9 H)
2o	2.45 (s, 3 H)	3.15—4.00 (m, 6 H)	6.55, 6.80 (both s, 1 H)	7.30—7.75 (m, 9 H)
2p	2.45 (s, 3 H)	3.20—4.40 (m, 6 H)		6.75—7.80 (m, 15 H, CH + H apom.)
2q	2.45 (s, 3 H)	3.05—3.90 (m, 4 H)		6.35—7.80 (m, 15 H, SH + H apom.)
2r	2.45 (s, 3 H)	1.30—1.60 (m, 6 H); 3.00—3.70 (m, 8 H)	6.60 (s, 1 H)	7.40—7.75 (m, 9 H)
2s	2.45 (s, 3 H); 2.75 (s, 3 H, MeS)	3.45 (m, 4 H)	6.40 (s, 1 H)	7.40—7.80 (m, 9 H)
2t	2.45 (s, 3 H)	3.45 (m, 4 H)	6.40 (s, 1 H)	7.30—7.70 (m, 11 H)
2u	2.45 (s, 6 H)	3.42 (s, 4 H)	6.40 (s, 1 H)	7.30—7.60 (m, 13 H)
2v	2.45 (s, 6 H)	3.45 (s, 4 H)	6.40 (s, 1 H)	7.30—8.20 (m, 12 H)
2w	2.45(s, 6 H); 3.75 (s, 3 H, MeO)	3.45 (m, 4 H)	6.40 (s, 1 H)	6.85—7.60 (m, 12 H)

(to be continued)

Table 3. (*continued*)

Compound	δ (*J*/Hz)			
	CH_3	CH_2	CH	H arom.
2x	1.70, 1.90 (both s, 3 H); 2.45 (s, 3 H); 2.94, 2.97 (both s, each 3 H, 2 MeN)	3.20—4.00 (m, 4 H)	6.30, 6.75 (both s, 1 H)	6.65—7.80 (m, 8 H)
2y	2.45 (s, 6 H); 2.95 (s, 6 H, 2 MeN)	3.45 (s, 4 H)	6.32 (s, 1 H)	6.65—7.60 (m, 12 H)
2z	1.10 (m, 6 H); 2.72 (s, 3 H)	3.00—3.85 (m, 4 H)	2.00 (m, 1 H, CH); 5.85 (d, 1 H, J = 10.0)	7.15—8.42 (m, 8 H)

Scheme 3



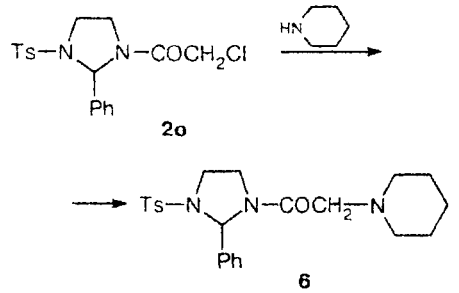
3c,f **2d,x**

R^2 = Pr^i (**3c**, **2d**), *p*-$Me_2NC_6H_4$ (**3f**, **2x**)

These compounds are also formed in close yields in the reactions of azomethines **3c,f** with acetyl chloride.

The use of functionalized acylating reagents extends the possibilities for the synthesis of DAI because functional groups can be involved in reactions (*e.g.*, according to Scheme 4).

Scheme 4



2o **6**

Thus, the method developed can be recommended for the preparation of various 2-substituted DAI.

Experimental

^{1}H NMR spectra were recorded on a Bruker AM-300 instrument. Melting points were determined on a Kofler stage. Before use, commercial solvents and reagents were distilled or recrystallized. The starting *N*-monoacylated EDA **1** was prepared according to the known procedures.[4,5]

***N*-Acyl-*N'*-alkylidene(arylidene)ethylenediamines (3a—i) (general procedure).** Aldehyde **4** (1 mmol) was added to a solution of compound **1** (1 mmol) **1** in 3—5 mL of EtOH. The reaction mixture was kept at 20 °C for 1 h and then allowed to stand overnight at −10 °C. The precipitate that formed was filtered off; azomethines **3c** and **3i** were isolated as oils upon removing the EtOH *in vacuo*.

***N,N'*-Diacyl-2-arylimidazolidines (2a—c,n—y) (general procedure).** Et_3N (1 mmol) and a corresponding acylating reagent (1 mmol) were added to a solution (or suspension) of compound **3** (1 mmol) in 5 mL of anhydrous MeCN. The reaction mixture was stirred at 20 °C for 2—6 h or refluxed for 1—2 h and then diluted with water. The precipitate that formed was filtered off, dried in air, and purified by recrystallization from EtOH or by preparative TLC with a 4 : 1 PhH—Me_2CO mixture as an eluent. Compound **2j** was reprecipitated with hexane from ether.

***N,N'*-Diacyl-2-isopropylimidazolidines (2d—m,z) (general procedure).** Isobutyraldehyde (1 mmol) was added to a solution of *N*-tosylethylenediamine (1 mmol) in 5 mL of anhydrous MeCN. The reaction mixture was kept at 20 °C for 1 h. After Et_3N (1 mmol) and a corresponding acylating reagent (1 mmol) were added, the synthesis was carried out as described above.

Reactions of azomethines 3 with Ac_2O. Ac_2O (1.2 mmol) was added to a solution (or suspension) of compound **3** (1 mmol) in 5 mL of anhydrous MeCN. The reaction mixture was kept at 20 °C for 1 h and then treated as described above.

2-Phenyl-1-*N*-piperidinoacetyl-3-tosylimidazolidine (6). A solution of DAI **2o** (0.54 g, 1.4 mmol) and piperidine (0.3 mL, 0.26 g, 3.0 mmol) in 5 mL of benzene was refluxed for 3 h. Then, the reaction mixture was cooled, washed with water, dried over Na_2SO_4, and concentrated; an oily residue crystallized upon trituration with ether. The precipitate that formed was filtered off, washed with ether, and dried in air to give compound **6** (0.47 g, 77%), m.p. 99—101 °C. Found (%): C, 64.65; H, 6.85; N, 9.63; S, 7.05. $C_{23}H_{29}N_3O_3S$. Calculated (%): C, 64.61; H, 6.84; N, 9.83; S, 7.48. ^{1}H NMR ($CDCl_3$), δ: 1.40 (m, 2 H, C—CH_2—C); 1.55 (m, 4 H, 2 C—CH_2—C); 2.20—2.35 (m, 4 H, 2 CH_2N); 2.42 (s, 3 H, Me); 2.70—3.90 (m, 6 H, 3 CH_2N); 6.80, 7.06 (both s, 1 H, CH); 7.25—7.80 (m, 7 H, H arom.).

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Received October 26, 1999;
in revised form December 29, 1999