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Studies in Sulphonamides—Part XIII : Synthesis of some new 1-methyl-3-aryl-2-(arylazo/N-substituted p-sulphamylbenzeneazo)propane-1,3-diones as potential antibacterials

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Abstract. Two 1-methyl-3-arylpropane-1,3-diones viz., 1-methyl-3-(2′,4′-dimethoxyphenyl)- and 1-methyl-3-(3′,4′-dimethoxyphenyl) propane-1,3-diones have been synthesised and coupled with diazotised simple and sulphonamide bases in presence of sodium acetate to furnish the corresponding 1-methyl-3-(2′,4′-/3′,4′-dimethoxyphenyl)-2-(arylazo/*N*-substituted *p*-sulphamylbenzeneazo) propane-1,3-diones. All these compounds were later subjected to *in vitro* screening against *S. aureus*, *E. coli* and *P. pyocyanea* when these showed considerable activity.

1. Introduction

Encouraged by the findings of our earlier work on the structure activity relationship of 1-methyl-3-aryl- and 1,3-diaryl-2-(*N*-substituted *p*-sulphamylbenzeneazo) propane-1,3-diones[1–5] we have now extended this work by synthesising 1-methyl-3-aryl propane-1,3-diones having two methoxyl groups in different positions of the phenyl ring and their coupling with different diazotised simple and sulphonamide bases. This work is expected to provide an opportunity to evaluate the effect of introducing and additional methoxyl group on the rate of coupling reaction and antibacterial properties of the synthesised compounds.

The present paper describes the synthesis of 1-methyl-3-(2′,4′-dimethoxyphenyl)- and 1-methyl-3-(3′,4′-dimethoxyphenyl) propane-1,3-diones and their coupling with simple and sulphonamide bases to furnish the corresponding 1-methyl-3-(2′,4′-/3′,4′-dimethoxyphenyl)-2-(arylazo/*N*-substituted *p*-sulphamylbenzeneazo) propane-1,3-diones of the type as shown in Fig. 1(a, b). The homogeneity and purity of all the compounds were checked by TLC and elemental analysis and structure assigned on the basis of IR and NMR spectral studies. All the synthesised products were later

subjected to *in vitro* screening against *S. aureus*, *E. coli* and *P. pyocyanea* at two different concentrations of 50 μg/ml and 100 μg/ml.

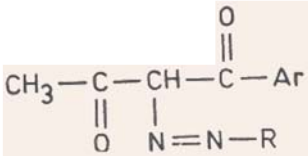

Fig. 1 (a) *Ar* = 2,4- and 3,4-dimethoxyphenyl; *R* = C_6H_4R'
(b) *Ar* = 2,4- and 3,4-dimethoxyphenyl; *R* = $C_6H_4SO_2NHR'$

2. Experimental Procedure

1-Methyl-3-(2',4'-dimethoxyphenly)- and 1-methyl-3-(3',4'-dimethoxyphenyl) propane-1,3-diones required for this work were prepared following the standard method of condensing the appropriate ketone with ethyl acetate in presence of sodium in dry ether. Both the β-diketones were purified through their copper complexes.

Synthesis of 1-methyl-3-(2',4'-/3',4'-dimethoxyphenyl)-2-(arylazo/N-substituted p-sulphamylbenzeneazo) propane-1,3-diones

To an ice-cold and well stirred solution of 1-methyl-3-(2',4'-/3',4'-dimethoxyphenyl)-propane-1,3-dione (0.002 mol) in ethanol (50 ml) containing sodium acetate (6 g) was slowly added a diazotised solution of sulphonamide or simple base (0.002 mol) with continuous stirring and cooling (0-5°). The yellow coloured azo compound was precipitated by the addition of water and after further stirring for five minutes, the separated solid was filtered, washed well with water and dried.

Pure 1-methyl-3-(2',4'-3',4'-dimethoxyphenyl)-2-(arylazo/N-substituted p-sulphamylbenzeneazo)propane-1,3-dione was crystallised from ethanol, acetic acid or DMF or from a mixture of any two of the above solvents (Table 1).

The NMR spectrum ($CDCl_3$) of 1-methyl-3-aryl propane-1,3-diones indicate that 1-methyl-3-(3',4'-dimethoxyphenyl) propane-1,3-dione is 99% in the enolic (chelated) form, because the signal at 4.15 δ, ($-CO-CH_2-\overset{\overset{O}{\|}}{C}-$) is very week and

$=CH-\overset{\overset{O}{\|}}{C}$ gives a signal at 6.3 δ, which is not the position in case of 1-methyl-3-(2',4'-dimethoxyphenyl) propane-1,3-dione (Table 2). Further, a hump in the region 16.6 δ, (*OH* protons) has moved downfield (18.9 δ) in the case of 1-methyl-3-(3',4'-dimethoxyphenyl)propane-1,3-dione which can be attributed to its involvent in hydrogen bonding.

In the NMR spectrum of 1-methyl-3-aryl-2-(arylazo/*N*-substituted *p*-sulphamylbenzeneazo)propane-1,3-diones, a peak in the region 2.57 ± 0.10 δ, characterstic of methyl group attached to a keto group is observed (Table 3). There are two peaks for two methoxyl groups in the region 3.75–4.1 δ; the one due to methoxyl group near the electron attracting group ($>C=O$) will be observed little down field as compared to that due to methoxyl group attached at the para position with respect to the keto

group. Two humps in the region 15.18–15.40 δ and 13.5–14.5 δ in case of 2-arylazo derivatives and ~ 12.8 δ and ~ 11.3 δ in 2-(*N*-substituted *p*-sulphamylbenzeneazo)-derivatives can be attributed to *OH* and *NH* protons respectively which indicates the presence of azo and hydrazono form in equilibrium.

The IR spectral studies of 1-methyl-3-aryl-2-(*N*-arylazo/*N*-substituted *p*-sulphamylbenzeneazo)propane-1,3-diones show a peak in the region ~ 1650 cm^{-1} and sharp peaks in the region 1565–1595 cm^{-1} and 1156–1170 cm^{-1}, which could be assigned to keto, azo and $CH_3-\overset{\overset{O}{\|}}{C}-$groupings (Table 4). Further a sharp peak in the range 808 to 844 cm^{-1} and peaks of medium or weak intensity in the region 833–890 cm^{-1} are indicative of the presence of 1,2,4-substituted phenyl ring; in the bands in the region 685–790 cm^{-1} represent differently substituted phenyl rings. IR spectra of 2-(*N*-substituted *p*-sulphamylbenzeneazo)propane-1,3-diones show the symmetrical and asymmetrical stretching frequencies due to $S=O$ group in the range 1090–1120 cm^{-1} and 1260–1350 cm^{-1} respectively. However, no bands in the region 3200–3400 cm^{-1} in the IR spectrum (KBr pellets) of different aryl azo derivatives are observed thereby supporting the arylazo structure.

During the coupling reaction, it has been observed that by the introduction of another methoxy group in the phenyl ring of the propane-1,3-dione, the rate of coupling in general decreases thereby giving lower yields. This is in confirmation with our earlier observation[4] that introduction of an electron releasing group decreases the rate of coupling reaction. However, when compared among themselves it was observed that shifting of one of the methoxyl group from ortho to the meta position in phenyl ring of the dione, with respect to one already present, the rate of coupling increases.

3. Evaluation of antibacterial activity

The results of the antibacterial tests at 50 μg/ml indicates that a fairly large number of compounds exhibit considerable activity against all the three micro-organism.

The azo compounds synthesised from sulphonamide bases are far more active than their corresponding simple azo analogues with few exceptions. When the results of screening of these compounds were compared with those of 1-methyl-3-(4′methoxyphenyl)-2-(*N*-substituted *p*-sulphamylbenzeneazo)propane-1,3-dione[3], it became clear that the introduction of an additional methoxyl group at ortho or meta position to the one already present caused an overall increase in the activitiy.

On comparing the results of 2-(arylazo/*N*-substituted *p*-sulphamylbenzeneazo) derivatives of the two series among themselves, it is observed that if a methoxyl group is changed from ortho to the meta position, the azo compounds by and large show considerable increase in activity against all the three micro-organisms.

Again, if the results are compared on the basis of changes made in the substitution pattern of the phenyl ring attached through an azo linkage, keeping all the other substituents unaltered, it is observed that the replacement of methyl group by a halogen atom, the activity against all the three increases but relatively more against

Table 1. 1-Methyl-3-(2′,4′-dimethoxyphenyl)-2-(arylazo)propane-1,3-diones
(Fig. 1a : *Ar* = 2,4-Dimethoxyphenyl)

Sl. No.	*R′*	M.P. (°C)	Colour	Yield (%)	Crystallisation solvent	Molecular formula	Percentage Found *C*	Percentage Found *H*	Percentage Requires *C*	Percentage Requires *H*	Antibacterial activity *S. aureus*	Antibacterial activity *E. coli*	Antibacterial activity *P. Pyocyanea*
1.	*H*	105	Y	70	EtOH	$C_{18}H_{18}O_4N_2$	66.2	5.3	66.2	5.5			++
2.	*o*-Methyl	131–2	YF	65	AcOH/DMF	$C_{19}H_{20}O_4N_2$	67.0	6.0	67.1	5.9	+		++
3.	*m*-Methyl	116	O	63	EtOH	$C_{19}H_{20}O_4N_2$	67.3	5.9	67.1	5.9	+		
4.	*p*-Methyl	122	Y	60	EtOH	$C_{19}H_{20}O_4N_2$	67.0	6.1	67.1	5.9			
5.	*o*-Chloro	160	YN	72	AcOH/DMF	$C_{18}H_{17}O_4N_2Cl$	60.1	4.9	60.0	4.7			
6.	*m*-Chloro	166	YN	70	EtOH/AcOH	$C_{18}H_{17}O_4N_2Cl$	59.8	4.6	60.0	4.7	+		
7.	*p*-Chloro	134	YF	75	EtOH/AcOH	$C_{18}H_{17}O_4N_2Cl$	60.0	4.7	60.0	4.7	+		
8.	*o*-Bromo	171	Y	82	DMF	$C_{18}H_{17}O_4N_2Br$	53.5	4.2	53.3	4.2			
9.	*p*-Bromo	146	Y	75	EtOH/AcOH	$C_{18}H_{17}O_4N_2Br$	53.2	4.4	53.3	4.2			++
10.	*o*-Nitro	189	YN	74	AcOH/DMF	$C_{18}H_{17}O_6N_3$	58.0	4.6	58.2	4.5	+		++
11.	*m*-Nitro	133	Y	75	EtOH/AcOH	$C_{18}H_{17}O_6N_3$	58.1	4.6	58.2	4.5	+		
12.	*p*-Nitro	164	YF	73	AcOH/DMF	$C_{18}H_{17}O_6N_3$	58.4	4.5	58.2	4.5	++		+++
13.	*o*-Methoxy	135–6	O	68	EtOH/AcOH	$C_{19}H_{20}O_5N_2$	63.8	5.3	64.0	5.6	++	+	+
14.	*m*-Methoxy	103	B	65	EtOH/AcOH	$C_{19}H_{20}O_5N_2$	64.1	5.5	64.0	5.6	++	++	+
15.	2,5-Dichloro	178–9	Y	75	AcOH/DMF	$C_{18}H_{16}O_4N_2Cl_2$	55.0	4.1	54.8	4.1	+		—

Table 2. 1-Methyl-3-(3′,4′-dimethoxyphenyl)-2-(arylazo)propane-1,3-diones
(Fig. 1a : *Ar* = 3,4-Dimethoxyphenyl)

Sl. No.	*R*	M.P. (°C)	Colour	Yield (%)	Crystallisation solvent	Molecular formula	Percentage				Antibacterial activity		
							Found		Requires				
							C	*H*	*C*	*H*	*S aureus*	*E. coli*	P. *Pyocyanea*
1.	*H*	168	Y	70	EtOH/AcOH	$C_{18}H_{18}O_4N_2$	66.2	5.5	66.2	5.5		—	+
2.	*o*-Methyl	117	YF	63	EtOH/AcOH	$C_{19}H_{20}O_4N_2$	67.0	6.1	67.1	5.9		—	+
3.	*m*-Methyl	164	YF	67	EtOH/AcOH	$C_{19}H_{20}O_4N_2$	67.1	6.0	67.1	5.9			++
4.	*p*-Methyl	153	YO	67	EtOH/AcOH	$C_{19}H_{20}O_4N_2$	67.3	5.9	67.1	5.9	—		+
5.	*o*-Chloro	118	Y	76	EtOH/AcOH	$C_{18}H_{17}O_4N_2Cl$	60.0	4.9	60.0	4.7	+		—
6.	*m*-Chloro	131	YN	73	EtOH/AcOH	$C_{18}H_{17}O_4N_2Cl$	59.8	4.8	60.0	4.7	+	+	—
7.	*p*-Chloro	160	Y	78	EtOH/AcOH	$C_{18}H_{17}O_4N_2Cl$	60.1	4.5	60.0	4.7	+	—	—
8.	*o*-Bromo	138	YF	78	EtOH/AcOH	$C_{18}H_{17}O_4N_2Br$	53.3	4.2	53.3	4.2		+	—
9.	*p*-Bromo	163	YN	75	EtOH/AcOH	$C_{18}H_{17}O_4N_2Br$	53.5	4.5	53.3	4.2			+
10.	*o*-Nitro	171	PY	75	EtOH/AcOH	$C_{18}H_{17}O_6N_3$	58.1	4.7	58.2	4.5			—
11.	*m*-Nitro	204	LY	75	AcOH/DMF	$C_{18}H_{17}O_6N_3$	58.0	4.5	58.2	4.5		+	
12.	*p*-Nitro	210	Y	79	AcOH/DMF	$C_{18}H_{17}O_6N_3$	58.3	4.5	58.2	4.5		+	
13.	*o*-Methoxy	148	RO	72	EtOH/AcOH	$C_{19}H_{20}O_5N_2$	64.2	5.4	64.0	5.6			
14.	*m*-Methoxy	172	O	63	EtOH/AcOH	$C_{19}H_{20}O_5N_2$	64.1	5.7	64.0	5.6	+		+
15.	2,5-Dichloro	188	Y	76	AcOH/DMF	$C_{18}H_{16}O_4N_2Cl_2$	55.0	4.3	54.8	4.1	+		

Table 3. 1-Methyl-3-(2′,4′-dimethoxyphenyl)-2-(N-substituted p-sulphamylbenzeneazo)propane-1,3-diones
(Fig. 1b : *Ar* = 2,4-Dimethoxyphenyl)

Sl. No.	*R*	M. P. (°C)	Colour	Yield (%)	Crystallisation solvent	Molecular formula	Percentage				Antibacterial activity		
							Found		Requires				
							C	*H*	*C*	*H*	*S. aureus*	*E. coli*	*P pyocyanea*
1.	*H*	203	YF	73	AcOH/DMF	$C_{18}H_{19}O_6N_3S$	53.1	4.9	53.3	4.7	+	++++	++
2.	Acetyl	205	YF	74	AcOH/DMF	$C_{20}H_{21}O_7N_3S$	53.7	4.7	53.7	4.7	+	+	—
3.	Phenyl	183	Y	76	EtOH/AcOH	$C_{24}H_{23}O_6N_3S$	60.0	5.0	59.8	4.8	+	—	++
4.	*o*-Methylphenyl	160	YO	65	EtOH/AcOH	$C_{25}H_{25}O_6N_3S$	60.7	5.3	60.6	5.5	—	++++	+++
5.	*m*-Methylphenyl	169	B	63	EtOH/AcOH	$C_{25}H_{25}O_6N_3S$	60.6	5.8	60.6	5.5	+++	++	—
6.	*p*-Methylphenyl	165	Y	60	EtOH/AcOH	$C_{25}H_{25}O_6N_3S$	60.8	5.4	60.6	5.5	+++	++	+++
7.	*o*-Chlorophenyl	168	Y	75	AcOH/DMF	$C_{24}H_{22}O_6N_3SCl$	55.8	4.2	55.7	4.2	++++	++	++
8.	*m*-Chlorophenyl	169	YN	76	AcOH/DMF	$C_{24}H_{22}O_6N_3SCl$	55.9	4.1	55.7	4.2	++++	+	+
9.	*p*-Chlorophenyl	167	LY	74	EtOH/AcOH	$C_{24}H_{22}O_6N_3SCl$	55.7	4.4	55.7	4.2	++	++	++
10.	*p*-Bromophenyl	171	B	76	EtOH/AcOH	$C_{24}H_{22}O_6N_3SBr$	51.0	4.1	51.0	3.8	+++	+++	+++
11.	Guanidyl	218	LY	75	AcOH/DMF	$C_{19}H_{21}O_6N_5S$	51.3	4.7	51.0	4.7	++	++	+
12.	α-Pyridyl	170	Y	65	EtOH/AcOH	$C_{23}H_{22}O_6N_4S$	57.5	4.8	57.4	4.6	++	++	+
13.	Pyrimidyl	206	PY	73	EtOH/AcOH	$C_{22}H_{21}O_6N_5S$	55.0	4.3	54.9	4.3	++++	++	+++
14.	2,6-Dimethyl-pyrimidyl	188	Y	70	EtOH/AcOH	$C_{24}H_{25}O_6N_5S$	56.5	5.0	56.3	4.9	+++++	+++	++
15.	4,6-Dimethyl-pyrimidyl	140	YF	74	EtOH	$C_{24}H_{25}O_6N_5S$	56.3	5.0	56.3	4.9	+++	+	+
16.	2,6-Dimethyl-pyrimidyl	145	Y	76	EtOH/AcOH	$C_{24}H_{25}O_6N_5S$	53.2	5.1	53.0	4.8	+++++	—	+
17.	5-Methyl-1,3,4-thiadiazol-2-yl	122	PY	63	EtOH/AcOH	$C_{21}H_{21}O_6N_5S_2$	50.0	4.2	50.1	4.2	+	—	++++++
18.	*m*-Nitrophenyl	178	LY	76	EtOH/AcOH	$C_{24}H_{22}O_8N_4S$	54.6	4.0	54.5	4.1	—	+	+++++
19.	*p*-Nitrophenyl	186	B	78	EtOH/AcOH	$C_{24}H_{22}O_8N_4S$	54.7	4.3	54.5	4.1	—	—	+++
20.	*o*-Methoxyphenyl	163	RB	74	EtOH/AcOH	$C_{25}H_{25}O_7N_3S$	58.9	5.0	58.7	4.5	—	—	+++++
21.	4-Methylpyrimidyl	196	B	75	EtOH/AcOH	$C_{23}H_{23}O_6N_5S$	55.3	4.6	55.5	4.6	+	—	+
22.	1-Phenyl-pyrazol-4-yl	145	B	65	EtOH/AcOH	$C_{27}H_{25}O_6N_5S$	59.1	4.3	59.2	4.5	—	++	+
23.	Thaizol-2-yl	192	Y	68	EtOH/AcOH	$C_{21}H_{20}O_6N_4S_2$	51.4	4.0	51.6	4.1	—	—	+

Table 4. 1-Methyl-3-(3′,4′-dimethoxyphenyl)-2-(*N*-substituted *p*-sulphamylbenzeneazo)propane-1,3-diones
(Fig. 1b: *Ar* = 3,4-Dimethoxyphenyl)

Sl. No.	*R*	M. P. (°C)	Colour	Yield (%)	Crystallisation solvent	Molecular formula	Percentage Found *C*	Percentage Found *H*	Percentage Requires *C*	Percentage Requires *H*	Antibacterial activity *S. aureus*	Antibacterial activity *E. coli*	Antibacterial activity *P. phyocyanea*
1.	*H*	202	Y	65	AcOH/DMF	$C_{18}H_{19}O_6N_3S$	53.3	4.5	53.3	4.7			
2.	Acetyl	204	Y	56	AcOH/DMF	$C_{20}H_{21}O_7N_3S$	53.9	4.7	53.7	4.7			+
3.	Phenyl	156	YF	62	EtOH/AcOH	$C_{24}H_{23}O_6N_3S$	60.0	4.7	59.8	4.8			—
4.	*o*-Methylphenyl	191	LY	60	EtOH/AcOH	$C_{25}H_{25}O_6N_3S$	60.6	5.5	60.6	5.5		+	
5.	*m*-Methylphenyl	186	LY	62	EtOH/AcOH	$C_{25}H_{25}O_6N_3S$	60.8	5.5	60.6	5.5	++		
6.	*p*-Methylphenyl	189	PY	64	EtOH/AcOH	$C_{25}H_{25}O_6N_3S$	60.4	5.4	60.6	5.5			
7.	*o*-Chlorophenyl	172	YF	68	EtOH/AcOH	$C_{24}H_{22}O_6N_3SCl$	55.9	4.3	55.7	4.2	++	+	
8.	*m*-Chlorophenyl	175	Y	65	EtOH/AcOH	$C_{24}H_{22}O_6N_3SCl$	55.7	4.5	55.7	4.2	++	—	
9.	*p*-Chlorophenyl	222	Y	66	DMF/EtOH	$C_{24}H_{22}O_6N_3SCl$	55.6	4.1	55.7	4.2	++	+	—
10.	*p*-Bromophenyl	225	YN	70	DMF/EtOH	$C_{24}H_{22}O_6N_3SBr$	58.8	4.0	58.9	3.9			—
11.	Guanidyl	193	B	72	AcOH/DMF	$C_{19}H_{21}O_6N_5S$	51.2	4.6	51.0	4.7		+	
12.	α-Pyridyl	213	Y	63	AcOH/DMF	$C_{23}H_{22}O_6N_4S$	57.4	4.8	57.4	4.6		—	
13.	Pyrimidyl	247	YN	70	AcOH/DMF	$C_{22}H_{21}O_6N_5S$	55.0	4.2	54.9	4.3		+	
14.	2,6-Dimethyl-pyrimidyl	165	LY	66	DMF/EtOH	$C_{24}H_{25}O_6N_5S$	56.5	5.0	56.3	4.9		+	
15.	4,6-Dimethyl-pyrimidyl	198	PY	71	DMF/EtOH	$C_{24}H_{25}O_6N_5S$	56.4	5.	56.3	4.9		++	
16.	2,6-Dimethoxy pyrimidyl	194	Y	70	EtOH/AcOH	$C_{24}H_{25}O_8N_5S$	52.9	4.6	53.0	4.8			
17.	5-Methyl-1,3,4-thiadiazol-2-yl	160	Y	65	DMF/EtOH	$C_{21}H_{21}O_6N_5S_2$	50.0	4.2	50.1	4.2			
18.	*o*-Nitrophenyl	168	PY	70	DMF/EtOH	$C_{24}H_{22}O_8N_4S$	54.5	4.4	54.5	4.1			++
19.	*m*-Nitrophenyl	218	BrR	72	DMF/EtOH	$C_{24}H_{22}O_8N_4S$	54.5	4.0	54.5	4.1			
20.	*p*-Nitrophenyl	201	LY	73	EtOH/AcOH	$C_{24}H_{22}O_8N_4S$	54.6	4.3	54.5	4.1			+++
21.	*o*-Methoxyphenyl	210	OY	69	AcOH/DMF	$C_{25}H_{25}O_7N_3S$	59.0	5.0	58.7	4.9	+++	++	
22.	4-Methyl pyrimidyl	205	Y	70	DMF/EtOH	$C_{23}H_{23}O_6N_5S$	55.3	4.6	55.5	4.6			
23.	3-Methoxy-pyrizine-6-yl	198	Y	68	EtOH/AcOH	$C_{23}H_{23}O_7N_5S$	53.7	4.5	53.8	4.5		++	++
24.	1-Pheny;pyra-zol-4-yl 130		BrR	62	AcOH/DMF	$C_{27}H_{25}O_6N_5S$	59.1	4.7	59.2	4.5	—	++	
25.	Thaizol-2-yl	211	Y	63	DMF/EtOH	$C_{21}H_{20}O_6N_4S_2$	51.7	4.1	51.6	4.1	—	+	+

B = Brown; Br = Brick; F = Flakes; L = Lemmon; N = Needles; O = Orange; P = Pale; R = Red; S = Shining and Y = Yellow.

S. aureus. The interchange from halogen to the nitro group further enhances the activity against *S. aureus*.

In 1-methyl-3-aryl-2-(*N*-substituted *p*-sulphamylbenzeneazo)propane-1,3-diones the replacement of the phenyl ring attached at *N'*-position of the sulphonamide residue by a six membered heterocyclic ring caused an increase in activity particularly against *S. aureus* and *E. coli*, while a five membered hetrocyclic ring decreases the activity. The order of activity against the three micro-organism is *P. pyocyanea* > *S. aureus* > *E. coli* in 2-arylazo derivatives and is *S. aureus* > *E. coli* > *P. pyocyanea* in case of 2-(*N*-substituted *p*-sulphamylbenzeneazo derivatives.

References

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