Electrochemical investigation of some potential antibacterials, II

Rajeev Jain, P. Padmaja, and Seema Gupta

Abstract: The electrochemical behaviour of 2-(4′-sulphonamoyl)hydrazonobutyrate-1,3-diones and sulphonamoylazoaminobenzenes has been studied over a wide range of pH at dropping mercury as well as glassy carbon electrodes. Both types of compounds exhibited a $4e^-$ reduction reaction at both electrodes. At pH > 4.5, 2-(4′-sulphonamoyl)hydrazonobutyrate-1,3-diones exhibited a $2e^-$ reduction wave at higher potentials. Both compounds undergo a $2e^-$ oxidation reaction. On the basis of polarography, linear and cyclic voltammetry, controlled potential electrolysis, coulometry, and spectral analysis, a detailed mechanism has been postulated for the reduction as well as the oxidation.

Key words: electrochemistry, sulphonamides, polarography, cyclic voltammetry, coulometry.

Résumé : Opérant sur une très large plage de pH et utilisant une électrode tombante de mercure ainsi qu'une électrode de carbone vitreux, on a étudié le comportement électrochimique de 2-(4′-sulfonamoyl)hydrazonobutyrate-1,3-diones et de sulfonamoylazoaminobenzènes. Les deux types de composés présentent une réaction de réduction à $4e^-$ avec les deux électrodes. À des pH supérieurs à 4,5, les 2-(4′-sulfonamoyl)hydrazonobutyrate-1,3-diones présentent une réduction à $2e^-$ à des potentiels plus élevés. Les deux types de composés subissent une réaction d'oxydation à $2e^-$. Sur la base des résultats obtenus par polarographie, voltammétrie linéaire et cyclique, électrolyse à potentiel contrôlé, coulométrie et analyse spectrale, on propose un mécanisme détaillé tant pour la réduction que pour l'oxydation.

Mots clés : électrochimie, sulfonamides, polarographie, voltammétrie cyclique, coulométrie.

[Traduit par la rédaction]

Sulphonamides were the first effective chemotherapeutic agents to be employed systematically for the prevention and cure of bacterial infections. Sulphonamides are widely used in the treatment of urinary tract infection, burn therapy, conjunctivitis, and chloroquine-resistant malaria and are also drugs of choice for the treatment of nocardiosis, toxoplasmosis, severe travellers diarrhoea, and meningococcal infections (1, 2).

Keeping in view the medicinal importance of sulphonamides (3), some new derivatives of sulphonamides have been synthesized. The redox behaviour (4, 5) of hydrazones has attracted considerable attention due to their importance as precursors (6) of various antineoplastic and antidiabetic compounds. However, no reference has been found dealing with those compounds bearing a -NH-N=N- group. This group of compounds having a -NH-N- as well as a -N=N- grouping in a single molecule deserve an extensive electrochemical study. In view of the importance of redox phenomena in reactions of physiological importance, it is considered worthwhile to study the mechanism and nature of the end products resulting from the reduction/oxidation of compounds of potential medicinal importance.

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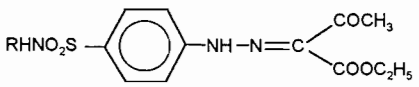
In continuation of our work (7–9) in exploring the redox behaviour of potential antibacterials, we have selected some hydrazono derivatives of sulphonamides, viz., 2-(4′-sulphonamoyl)hydrazonobutyrate-1,3-diones (A) and sulphonamoylazoaminobenzenes (B) for a comprehensive and comparative study taking into consideration such pertinent aspects of redox phenomena as the nature of the waves, characterization of the end products, and mechanism of the electrode process. These studies have been augmented, wherever possible, by results of controlled potential electrolysis, coulometry, and product analysis.

Experimental

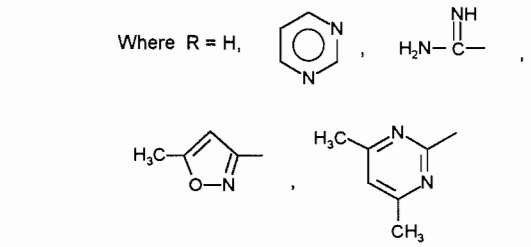
Some representatives of both classes of compounds, viz., 2-(4′-sulphonamoyl)hydrazonobutyrate-1,3-diones (A) and sulphonamoylphenyl azoaminobenzenes (B), as represented in Scheme 1, were selected.

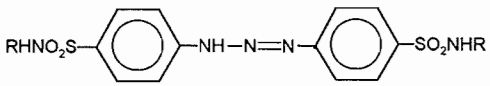
Compounds A and derivatives were synthesized by the literature method (10). Compounds B were synthesized and characterized in the following manner. Sulphanilamide (0.01 mol, 1.72 g) was dissolved in a mixture of concentrated hydrochloric acid (5 mL) and water (5 mL) and cooled to 0°C in an ice bath. To this, a cold aqueous solution of sodium nitrite (0.01 mol, 0.69 g) was added. The diazonium salt so obtained was filtered into an already cooled solution mixture of sodium acetate (8.0 g) and sulphanilamide in alcohol. A yellow precipitate of sulphonamoylphenylazoaminobenzene appeared immediately. The precipitate was filtered, washed with water, and recrystallized from light petroleum, mp 91°C, yield 68%. IR ν_{max}^{KBR}: 3370 ($NH\text{-}C_6H_5$), 3220, 3180 (NH_2SO_2), 1520

Scheme 1.

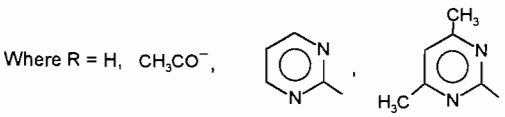


(A)





(B)



(-N=N-), 1340 (SO_2) cm^{-1}; ^1H NMR δ ($CDCl_3$): 2.2 (2H, -NH_2), 7.4 (5H, pH), 9.3 (H, NH). Anal. calcd. for $C_{12}H_{13}N_5S_2O_4$: C 40.56%, H 3.66%, N 19.70%; found: C 40.20%, H 3.5%, N 19.40%.

Similarly, other substituted sulphonamoylphenylazoaminobenzenes were synthesized and duly characterized.

Apparatus and technique

Polarographic measurements were carried out on an ELICO DC CL-25 polarograph. Potentiometric studies were carried out on an expanded-scale pH meter with a glass electrode that was previously standardized with buffer of known pH. All potentials reported are with reference to the SCE. Triply distilled mercury was used for the dropping mercury electrode (DME).

Cyclic voltammetric studies were carried out on a BAS CV-27 cyclic voltammograph in connection with a digital electronic 2000 Omnigraph X-Y/t recorder. A three-electrode cell was used for the purpose. The cell consists of a glass cylinder with a cap having holes for introducing electrodes and tubing for nitrogen gas. Oxygen was removed from the solution by passing nitrogen gas through the solution for about 10 min and then a blanket of nitrogen was maintained throughout the experiment. In the present study, glassy carbon (GC) was used as the working electrode, 1.0 M Ag/AgCl as the reference electrode, and platinum wire was used as the auxillary electrode. The working electrode was polished with fine-grade emery paper, followed by polishing with alumina (0.5 μm), then was washed and activated by triangular voltage sweeps from +1.0 V to −1.6 V at a rate of 5–200 mV/s for 15 min. The activity of the electrode was tested (11) using a ferricyanide–ferrocyanide redox couple in 0.01 M KCl.

For the purpose of cyclic voltammetric and polarographic measurements, solutions were prepared by mixing a 2.0 mL solution of the compound, 2.0 mL of DMF (which is necessary to keep the compound in solution), 1.0 mL of 1.0 M KCl, and 5.0 mL of appropriate Britton–Robinson buffers (12). Britton–Robinson buffers in the pH range 2.0–11.8 were prepared by adding suitable amounts of 0.2 M NaOH solution to a stock B.R. buffer solution (pH 1.8). B.R. buffer stock was prepared by mixing boric acid, phosphoric acid, and glacial acetic acid (mixture contains 2.472 g of boric acid, 2.14 mL of phosphoric acid, 2.30 mL of glacial acetic acid made up to 1000 mL).

For the coulometric determination (13) of the number of electrons transferred in the electrode process, 2.0 mL of the solution of the compound containing buffer, solvent, and supporting electrolyte in the same ratio as used for CV studies was electrolyzed at a slightly more negative potential than the peak potential for the respective peaks. From the decrease in current with time, the number of electrons transferred was calculated. The progress of electrolysis was monitored by recording voltammograms at regular intervals of time. The products of electrolysis were identified by comparison of their properties with authentic products using TLC.

Results and discussion

DC polarography

All the 2-(4′-sulphonamoyl)hydrazonobutyrate-1,3-diones (A) reduced in a single, $4e^-$, well-defined wave in the pH range 2.0–12.0. However, an additional $2e^-$ wave was observed at pH > 4.5 (Table 1). The first wave over the entire pH range was found to be the diffusion-controlled, irreversible reduction of hydrazono group. The diffusion-controlled nature of the limiting current was established on the basis of linear plots of i_d vs. concentration and i_d vs. $h^{1/2}$ (where h is the height of the mercury column). Sulphonamoylphenylazoaminobenzenes (B) exhibited one four-electron, diffusion-controlled irreversible reduction wave over the entire pH range 2.0–12.0 (Table 2). In these compounds, no additional wave was observed at pH > 4.5. The number of electrons involved in the polarographic wave was found, by coulometry, to be four. The value of n was also determined by controlled potential electrolysis using the following formula (14).

$$n = \frac{i_0}{2.30\, C_0 F u(\Delta \log i/\Delta t)}$$

where i_0 is the current at $t = 0$ when $C = C_0$, u is the volume of the solution and $\Delta\log i/\Delta t$ is the slope of the plot of log i against log t. This estimate provided support for the value obtained by the millicoulometric method. The controlled potential electrolysis was carried out at a potential corresponding to the plateau of the wave.

The half-wave potentials of both compounds A and B are a linear function of pH. The plot of $E_{1/2}$ vs. pH was linear up to 7.5 for A and to 9.2 for B and thereafter there is little change in $E_{1/2}$ with pH (Fig. 1). This suggests that protons are involved in the reduction process. However, in alkaline solution, the half-wave potential did not change with pH. Irreversibility of

Table 1. Voltammetric characteristics of 2-(4′-sulphonamoyl)hydrazonobutyrate-1,3-diones at pH 4.5, concentration = 1.0×10^{-4} M.

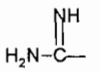
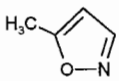
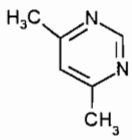
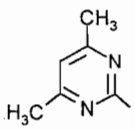
Scan no.	R	$-E_{1/2}$, V	i_d, μA	$-E_p$, V	i_p, μA	Current density $i_p/ACv^{1/2}$
1.	H	0.82	0.80	1.25	5.9	0.12×10^3
2.		0.84	0.88	1.10	5.7	0.11×10^3
3.		0.78	0.92	1.17	5.3	0.11×10^3
4.		0.92	0.92	1.12	6.2	0.12×10^3
5.		0.76	0.80	1.13	6.8	0.18×10^3

Table 2. Polarographic characteristics of sulphonamoylphenylazoaminobenzene at pH 6.70, concentration 2.0×10^{-4} M.

Scan no.	Substituent	Melting point, °C	$-E_{1/2}$, V	Standard deviation	i_d, μA	Standard deviation
1.	H	91	0.82	±0.017	2.32	±0.014
2.	CH_3—CO—	98	0.84	±0.013	2.40	±0.018
3.		101	0.83	±0.015	2.45	±0.020
4.		107	0.81	±0.016	2.41	±0.012

the electrode process was confirmed by plotting $E_{d.e.}$ against $(i/i_d - i)$. It was found that the value of slope exceeded $59.2/n$ mV. The values of αn_a (product of transfer coefficient and number of electrons transferred in the rate-determining step) and p (number of protons involved in rate-determining step) were determined by using the following expressions (15, 16). The values of αn_a determined by logarithmic analysis of the polarographic wave are comparable with those values determined from the expressions given below. The value of p obtained from the expressions was nearly equal to 1.0.

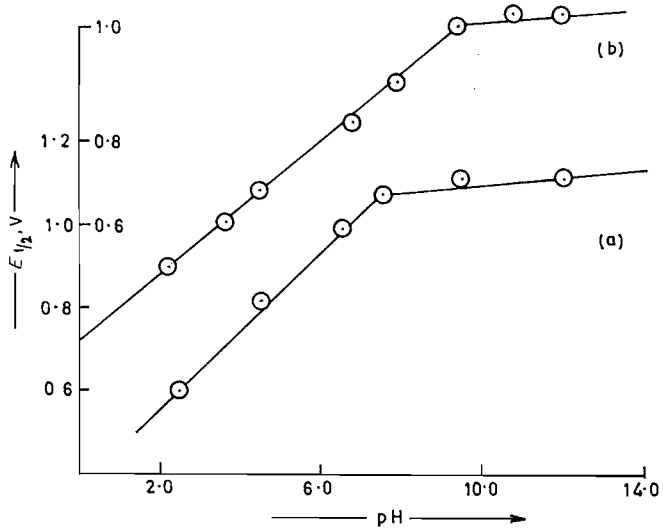
$$E_{3/4} - E_{1/4} = \frac{0.0517}{\alpha n_a}$$

$$\frac{dE_{1/2}}{dpH} = \frac{0.0591}{\alpha n_a} p$$

Linear sweep and cyclic voltammetry

Cyclic voltammograms of 2-(4-sulphonamoyl)hydrazonobutyrate-1,3-diones (A) and sulphonamoylazoaminobenzenes (B) were recorded at different scan rates and concentrations using the glassy carbon electrode. Typical cyclic voltammograms are incorporated in Fig. 2. The cyclic voltammogram of compound A exhibited one well-defined cathodic peak in the pH range 2.5–12.0 (Table 1). One more peak, II_c, at higher negative potentials was observed at pH > 4.5. In sulphonamoylazoaminobenzene (B) two well-defined two-electron

Fig. 1. Plot of $-E_{1/2}$ vs. pH for (*a*) 2-(4′-sulphonamoyl)-hydrazonobutyrate-1,3-diones; (*b*) sulphonamoylphenyl-azoaminobenzene.



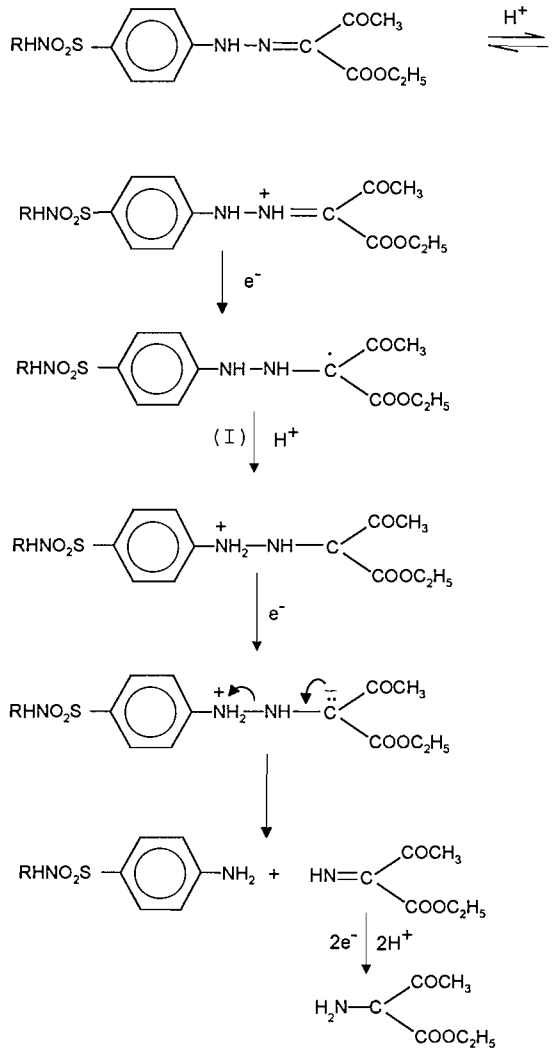
reduction peaks were observed over the entire pH range (Table 3). The peak currents were measured from the direct reading shown on the digital display of the potentiostat. A cyclic voltammogram of the solvent and the supporting electrolyte was recorded after flushing with nitrogen gas for about 10 min. The current of the blank was considered as the base line (c) as indicated in Fig. 2. The peak currents (i_p) of the compound A were comparable to those of the compound B. The linear plots of peak current i_p vs. the square root of the scan rate (Fig. 3) and i_p vs. concentration of the depolarizer show a slight deviation from the origin, indicating adsorption (17) of the depolarizer in both waves, in acidic as well as in alkaline media. The peak potential was found to shift cathodically with scan rate. The peak potentials also shifted to negative values with the concentration of the compound.

The peak potential was dependent on pH and shifted towards more negative potentials with an increase in pH up to 7.5 for compound A and 9.2 for compound B. On reversing the scan, only a small peak was observed at very high positive potentials, indicating that the processes corresponding to both peaks are irreversible. However, this small hump (I_a) in Fig. 2 at positive potentials indicates that these hydrazones and azoaminobenzenes also undergo oxidation at the glassy carbon electrode. To ascertain whether the peak is due to oxidation of hydrazones, voltammograms were initiated in the positive direction. It was found that the oxidation peak appeared at the same potential. Thus it can be safely concluded that the oxidation peak is due to the independent oxidation of hydrazones and azoaminobenzenes. No corresponding peak was observed in the reverse scan, indicating that the oxidation process in both cases is irreversible. The number of electrons involved in the oxidation process was calculated from coulometric measurements and found to be 2.0.

Coulometry and controlled potential electrolysis

At pH 2.5–4.0 at −1.0 V, 2-(4′-sulphonamoyl)hydrazonobutyrate-1,3-diones (A) undergo a four-electron reduction reac-

Scheme 2.



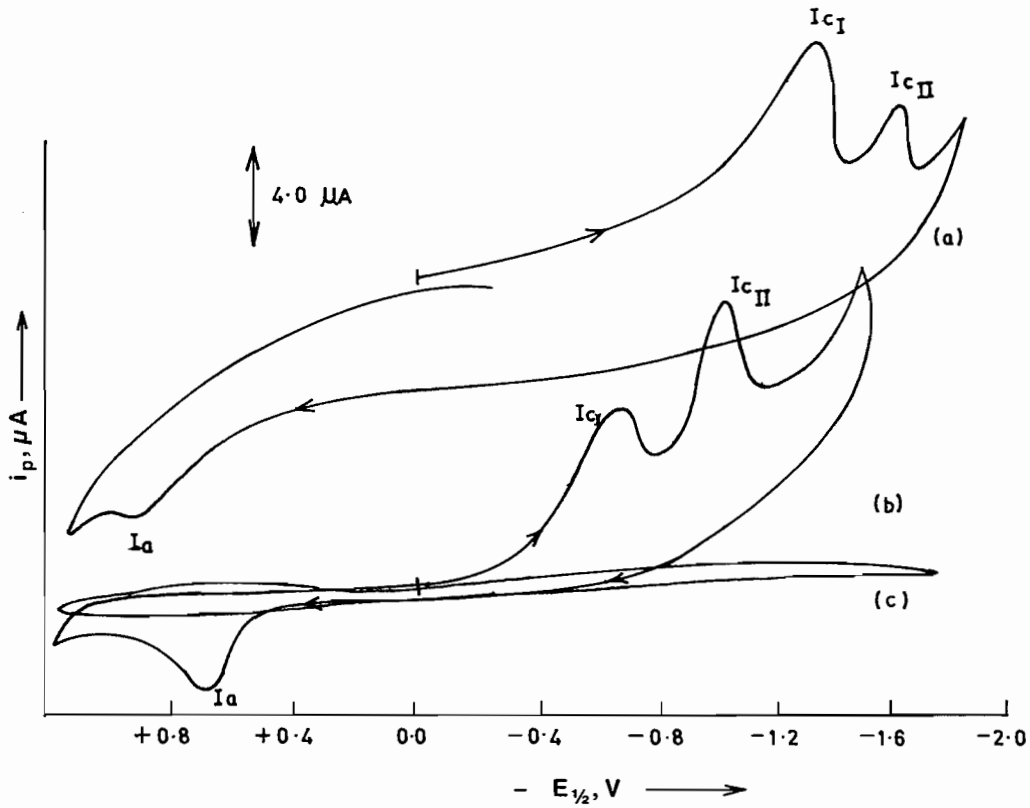
tion. The TLC of the electrolyzed solution on a silica-gel G-plate resulted in two spots. One of the separated compounds exhibited bands corresponding to primary amino stretching bands at 3210 cm^{-1}, C—H linkage at 2650 cm^{-1}, and a peak for the C=O group at 1695 cm^{-1}. The other compound was identified as sulphanilamide by comparison with standard sulphanilamide. It was also verified using different solvent mixtures.

Controlled potential electrolysis at pH 6.2 and 10.5 at −1.5 V and subsequent product analysis confirms the presence of sulphanilamide, ethyl acetoacetate, and ammonia. The presence of ethyl acetoacetate was proved by comparing with authentic ethyl acetoacetate using GLC, and ammonia was detected by Nessler's reagent. The number of electrons involved in the entire overall process was found to be 6.0 + 0.25. A detailed mechanism is given in Scheme 2 in the pH range 2.0–12.0 and at pH > 4.0 in Scheme 3, while azoaminobenzenes over the entire pH range studied consumed four electrons. Each peak required a two-electron process. Controlled potential electrolysis resulted in two products, as separated by TLC on silica gel G plates of 0.25 cm thickness

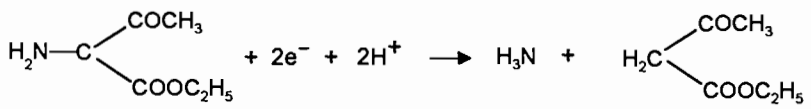
Table 3. Voltammetric characteristics of sulphonamoylphenylazoaminobenzene at GC electrode. Conc. 2.0×10^{-4} M; scan rate (v) 50 mV s^{-1}.

Scan no.	pH	$-E_{pc}$, I	i_{pc}, I	Current density $i_p/Acv^{1/2}$ $\times 10^2$	$-E_{pc}$, II	i_{pc}, II	Current density $i_p/Acv^{1/2}$ $\times 10^2$	E_{pa}I, V	i_{pa}, μA	Current density $i_p/Acv^{1/2}$ $\times 10^2$
1.	2.30	0.70	4.6	0.46	0.82	4.5	0.46	0.66	4.5	0.46
2.	4.60	0.75	5.2	0.52	0.88	5.2	0.52	0.67	4.8	0.48
3.	6.70	0.84	4.8	0.48	0.98	4.9	0.48	0.38	5.0	0.50
4.	7.75	0.80	4.9	0.49	1.03	5.3	0.53	0.30	4.5	0.46
5.	9.20	1.10	5.3	0.53	1.18	5.0	0.50	0.26	5.2	0.52
6.	11.40	1.11	5.0	0.50	1.18	5.2	0.52	0.25	5.0	0.50

Fig. 2. Cyclic voltammograms of (*a*) 2-[4′-(4″-6″-dimethyl)pyrimidinyl sulphonamoyl)hydrazonobutyrate-1,3-dione at pH 7.2, $v = 50$ mV/s, concentration = 1.0×10^{-4} M; (*b*) sulphonamylphenylazoaminobenzene at pH 4.7, $v = 50$ mV/s, concentration = 1.0×10^{-4} M; (*c*) base line in DMF.

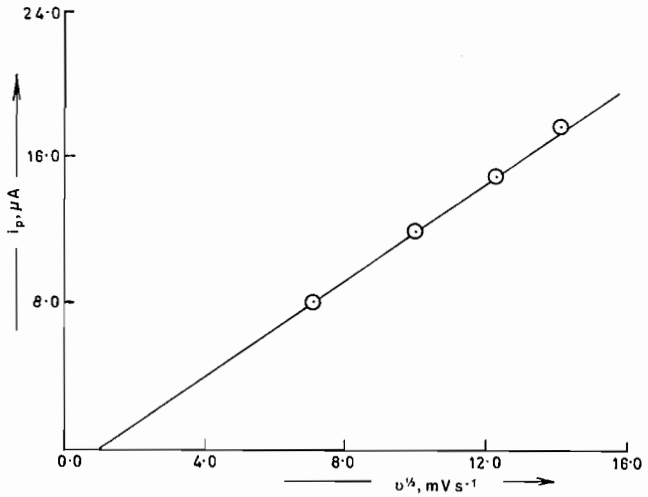


Scheme 3.

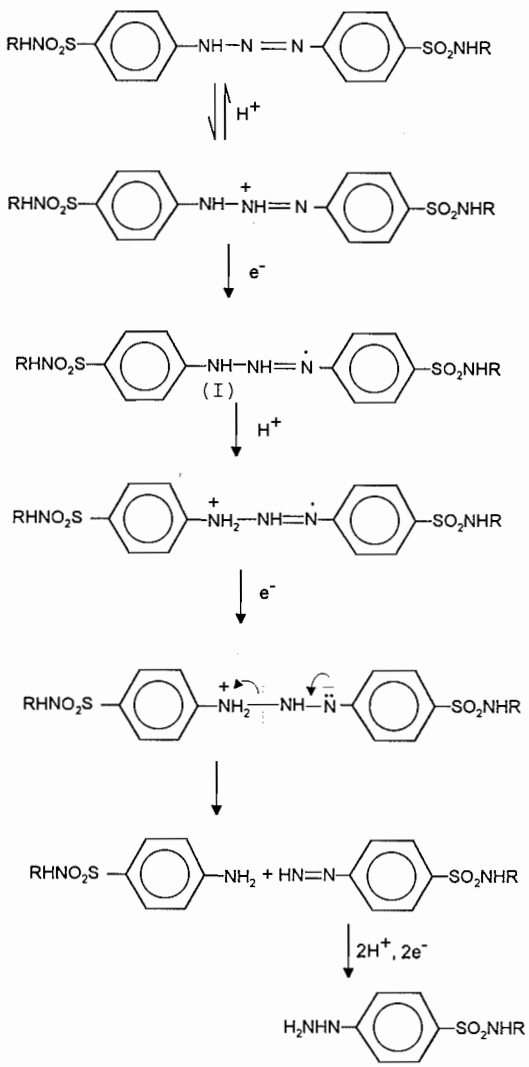


(chloroform and carbon tetrachloride in the ratio 1:1), $R_f = 0.8$ and 0.2. One of the spots is matched with an authentic sample of sulphanilamide (Anal. calcd. for $C_6H_8N_2SO_2$: C 41.86, H 4.65, N 16.27%; found: C 42.52, H 4.92, N 16.5%. IR ν_{max}^{KBr}: 1260 (C-N), 1340 (SO_2); 3180, 3210 (NH_2SO_2) cm^{-1}). The IR spectra could be superimposed on the IR spectra of an authentic sample, confirming that one of the products formed after electrolysis is sulphanilamide. The other product is identified as sulphonamoylphenylhydrazine by comparison of its R_f values with those of related compounds. Keeping these facts in view, the reduction may be proposed to be as given in Scheme 4 for compounds B.

Fig. 3. Plot of i_p (μA) vs. $\nu^{1/2}$ for 2-[4′-(4″-6″-dimethyl)pyrimidinyl sulphonamoyl]hydrazonobutyrate-1,3-dione at pH 4.5, concentration = 1.0×10^{-4} M.



Scheme 4.



The above mechanism is supported by the shift of $E_{1/2}$ towards more negative values with pH as protons are consumed in the reduction. As the equilibrium shifts towards the unprotonated forms, $E_{1/2}$ becomes constant. In this mechanism (Schemes 2 and 4), the radical (I) could also combine with an electron instead of a proton, but this possibility is ruled out because such a mechanism involves only two electrons, whereas coulometric data clearly indicate a four-electron reduction process.

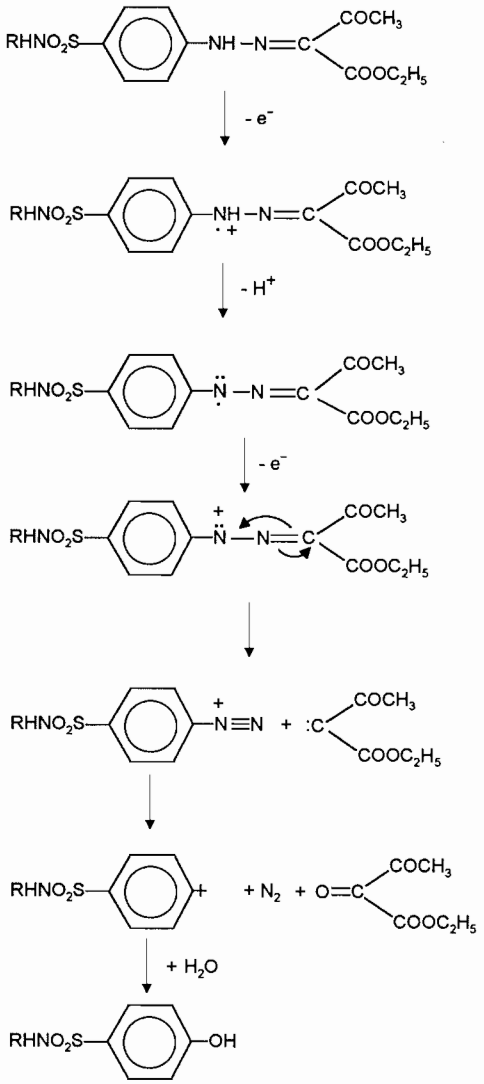
However, in compounds A, further reduction of the product formed at the four-electron stage takes place in a second 2e reduction wave at pH > 4.5. The peak/wave has been assigned to the reduction of the activated C—N bond, as shown in Scheme 3. Other workers have also reported a similar mechanism for the activated C—N bond (18, 19). Coulometric data for compounds A and B at the oxidation peak potential resulted in two electrons for each compound. Controlled potential electrolysis of these compounds resulted in two spots, indicating the formation of the different compounds.

Controlled potential electrolysis (CPE) of hydrazono compounds A was carried out to determine the number of products. For CPE, hydrazone was electrochemically oxidized at the GC electrode using 1.0 M tetraethylammonium bromide at 1.2 V. A cyclic voltammogram was recorded using the solvent and supporting electrolyte without depolarizer to determine whether or not oxidation is due to depolarizer. No peak was observed in the range of the hydrazone group. This observation ruled out the possibility of oxidation of any other component than the hydrazono group.

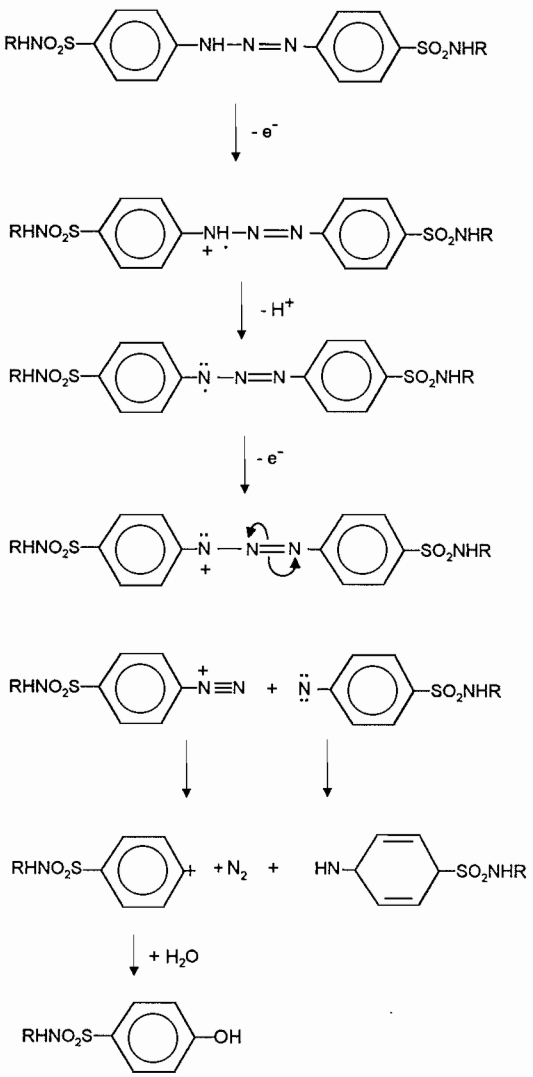
UV–visible spectra of the solution were recorded at different time intervals during electrolysis. λ_{max} at 390 nm due to the -NH-N=C- grouping showed a systematic decrease in the absorption maxima after each time interval and finally disappeared. Disappearance of the peak indicated cleavage of the NH-N=C- group during oxidation. After complete CPE, the analyte was subjected to product analysis. The electrolyzed solution, on separation by TLC using hexane + methanol (70:30 V/V), gave two products. One of the products was identified by TLC as *p*-hydroxy benzenesulphonamide by comparison with an authentic sample. The second product was characterized by spectral analysis, i.e., IR ν_{max}^{KBr} : 1720 cm^{-1}, characteristic of the >C=O group, and ^{1}H NMR δ ($CDCl_3$): 1.39 (t, 3H, $-CH_3$), 4.24 (q, 2H, $-CH_2$), clearly indicating that the second product is an α,β-diketoester. The α,β-diketoester was obtained in fairly good yield (20–45%).

The product was confirmed by its reaction with 2,4-dinitrophenylhydrazine. When the product of the electrooxidation was treated with an excess of 2,4-dinitrophenylhydrazine, it readily formed hydrazone, which was isolated and identified by elemental and spectral analysis. Keeping these products in view, a probable mechanism may be proposed, as depicted in Scheme 5. This mechanism is also supported by the work of others (20, 21), whereas in sulphonamoylazoaminobenzene (B), controlled potential electrolysis of the compound resulted in two spots on separation by TLC on alumina using benzene + chloroform (60:40 V/V). The products were identified by comparing with authentic compounds. The probable mechanism for the oxidation of this compound is given in Scheme 6. The products are expected to result from the cleavage of the diazonium cation and by the rearrangement of nitrene formed during the oxidation. The radical cation obtained after

Scheme 5.



Scheme 6.



abstraction of an electron undergoes two main reactions, viz., loss of a proton from the α-position to the azo group and cleavage of the arylazo group, forming a diazonium cation. The diazonium cation disproportionates into nitrogen and an aryl cation, which loses a proton when a water molecule is added. This compound is matched with an authentic sample. The nitrene formed undergoes tautomerism. This type of mechanism has been proposed by Fasani et al. (22) for the oxidation of azo benzenes.

Finally, it was observed that both reduction and oxidation proceed through the same pathways, although the products are different. In the case of hydrazones at higher pH, i.e., pH > 4.5, the product obtained at the $4e^-$ stage undergoes further reduction due to the activated C—N bond. Lack of electron-withdrawing groups in azoaminobenzene may be the reason why further reduction of hydrazine is hindered, whereas oxidation of hydrazones, as well as azominobenzenes, follows the same path. In hydrazones, carbene formed is flanked by electron-withdrawing groups and hence readily adds a water molecule and is converted into a ketone, while in azoaminobenzene the nitrene obtained readily undergoes tautomerism.

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