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HYDROGEN AND SULFUR FROM HYDROGEN SULFIDE V.
ANODIC OXIDATION OF SULFUR ON ACTIVATED GLASSY CARBON

by

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Submitted to

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HYDROGEN AND SULFUR FROM HYDROGEN SULFIDE. V

ANODIC OXIDATION OF SULFUR ON ACTIVATED GLASSY CARBON

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Abstract- Fast, ambient-temperature, steady electrolysis of H_2S from solutions of NaSH in aqueous NaOH was achieved using suitably activated glassy carbon anodes. Thus electrolyses of H_2S can probably be carried out at high rates with modest cell voltages in the range 1-1.5 V. The variation in anode current densities with $[NaSH]$, $[NaOH]$, and anode rotational speed is presented.

INTRODUCTION

Previous studies have demonstrated benefits which might be realized by processing H_2S from mineral sources to recover H_2 and S and

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suggested thermochemical¹⁻⁸ and electrochemical⁹⁻¹² means to achieve this recovery.

Cathodes for the reduction of H^+ are well known. But the electrochemical literature, provides little help in solving two pressing problems: the low conductivities⁸ of the electrolytes and fouling of the anode by sulfur.¹²

Hamilton and Woods¹⁵ observed that the formation of multilayers of sulfur leads to passivation of a gold electrode in borate solutions in 1 M NaOH. Faroque and Fahidy¹⁶ reported that Pt electrodes are totally passivated by sulfur within 30 seconds after the application of a potential of 0.27 V with respect to a saturated calomel electrode, [V(SCE)]. To avoid this problem, they used a rotating tripolar wiper blade electrode.

The work of Lessner et al¹⁷ and Giggenbach¹⁸ suggests that the formation of polysulfides in strongly basic solutions should help avoid fouling, but that the use of strongly basic solutions at high temperatures (about 100°C) would lead to an undesirable disproportionation of zero valent sulfur to thiosulfate and sulfide. Dandapani, Scharifker, and Bockris¹² avoided passivation by using a basic solvent at 85°C. Using an H_2S -saturated 6M NaOH solution, they conducted electrolyses for extended periods at current densities of 100 mA/cm², producing H_2 at the cathode and elemental S and polysulfides at the anode, both at high faradaic efficiencies. At 25°C, the current density was about 25 mA/cm². Their anode was crucible-type graphite. They attribute their success to the solubilization of sulfur and the production of polysulfides at the anode and the sub-

sequent precipitation of sulfur from the electrolyte solution elsewhere. They make the point, however, that the use of catalytic anodes has (heretofore) been of no avail in solving the problem of sulfur passivation. In this paper we describe some observations we made and the results of a series of electrochemical studies of activated glassy carbon anodes in alkaline sulfide solutions at room temperature.

EXPERIMENTAL PART

Our preliminary studies confirmed many of the previous pessimistic observations, but we obtained encouraging results with activated glassy carbon anodes. We therefore explored the anodic oxidation of sulfur in aqueous NaOH with these electrodes. Our principal exploratory technique was linear scan voltammetry. The anode potentials are all given with respect to a saturated calomel electrode (SCE). The counterelectrode was a platinum cathode. The anode was a rotating-disc, glassy carbon working electrode. Figure 1 shows schematically the kinds of voltammograms we observed. All scans started at the open circuit potential of about about -0.65 V(SCE).

With an ordinary glassy carbon anode, the current stays very low until the anode potential is of the order of 1 V(SCE). Then the cell current starts to increase due to O_2 evolution at the anode. On the reverse sweep, the current closely retraces the path it took on the positive sweep, as is shown in Fig. 1a. However, if the positive sweep is extended to higher voltages, of the order of 2.5 V(SCE), and

held at this value momentarily, the reverse sweep retraces the positive sweep down to about 0.75 V(SCE). Then, the current increases abruptly, as is shown by the curve in Fig. 1b. It is much higher in the range -0.25 to +0.75 V(SCE) than it was before the anode had been activated. The activation of the anode is accompanied by the formation of a black film, which peels from the glassy carbon to reveal a yellow white substrate layer. The proportional increase in current with voltage is characteristic of an electrolytic cell in which the predominant overvoltage is associated with the resistance of the electrolytic solution.

If the voltage is held constant in the range of about -0.25 to +0.75 V(SCE), there is a steady flow of the augmented current, accompanied by electrolysis. It is apparent that activated glassy carbon anodes can oxidize sulfur species at much higher rates than untreated ones. In a comparable experiment graphite gave much lower currents than activated glassy carbon.

Increasing the voltage above 0.75 V(SCE) again leads to an abrupt decrease in the current density. This decrease is similar to those which have been attributed to passivation of the anode by the deposition of sulfur.¹²

Our cell has been described elsewhere.²⁰ The rotating disc anode assembly consisted of a teflon shaft with a brass inner core and an interchangeable teflon-coated disc electrode. Grade V10 glassy carbon was obtained from Atomergic Chemicals, Inc. Anodes were made by press-fitting glassy carbon disks and a brass shaft, connected by a conducting silver epoxy resin, into a teflon sleeve. Their surfaces were

polished to a mirror-like finish with Buehler, Ltd., microcloth polishing cloths on a polishing wheel using a slurry of 0.05 micron alumina in deionized water. The electrodes were then rinsed thoroughly with deionized water, activated, and used.

Solutions were made from (MCB Manufacturing Chemists Inc.) brand reagent grade NaOH and (Aldrich Chemical Company) brand reagent-grade NaHS. Deionized H₂O was used for all preparations. Air Products ultra-pure, carrier-grade N₂ was used for deaerating the deionized H₂O as well as for purging the cell. The experimental details have been described elsewhere.¹⁹

Anode potentials were measured with respect to a Fisher brand SCE. The principal experimental variables were [NaOH], [NaHS], and the rotational speed of the electrode. The linear scan experiments were followed by longer steady runs on activated electrodes at a fixed applied potential, in which the behavior of the anodes over extended periods of time was observed.

Glassy carbon activation apparently requires a high pH, which, in aqueous solutions, implies a high concentration of OH⁻. Attempts to activate the glassy carbon electrode in 1.0M NaSH were unsuccessful. The linear scan voltammogram looked like that of Fig. 1a. Bringing the NaOH concentration to 0.71M after the attempted activation in 1.0M NaSH did not enhance the current at 0.25 V(SCE). In 1M NaSH, with a measured pH of 11.2, the (calculated) pH is 9.98 and the calculated [OH⁻]=9.54x10⁻⁵.

In contrast, an activation sweep in 1M NaOH which contains no NaSH, in which the (calculated) [OH⁻] is four orders of magnitude

higher, results in activation of the glassy carbon. If, after such a sweep in 1M NaOH, NaSH is added to bring the concentrations to $[\text{NaOH}] = 0.71$ and $[\text{NaSH}] = 1$, the current density immediately jumps to the same value it achieves when the activation is effected in a solution which already contains both NaOH and NaSH.

Table 1 shows the results of some selected exploratory experiments in which we examined the effect of solution composition during the activation step on the subsequent current densities at 0.25 V(SCE). In general, the observations suggest that NaOH is necessary for anode activation and that the presence of NaSH in the electrolyte during activation does not affect the activation process insofar as subsequent current densities at a fixed potential (0.25 V) are concerned.

RESULTS

Figures 2-8 were generated by a microcomputer using a spline-fitting program on our experimental points. The original data can be found in Ref. 19. The anode potential in all of these experiments was 0.25 V(SCE). The figures show the variations of current density with rotational speed and $[\text{NaSH}]$ for $[\text{NaOH}] = 0.05, 0.1, 0.5, 1.0, 2.0, 4.0$, and 6.0 . The NaSH concentration ranged from 0.05 to 1.55. Although the anode exhibited activation at NaOH concentrations less than 1M, activation was most effective when $[\text{NaOH}] \geq 1\text{M}$. In the more basic solutions, current densities in the range 250-300 ma/cm² were the rule. They decreased dramatically as the NaOH concentration

dropped below about 1.0M. In the strongly basic solutions, the current densities increased monotonically with [NaSH], but in the weakly basic solutions, the variation of current density with [NaSH] was more erratic. It is possible that this behavior results from the fouling of the anode by sulfur. In strongly basic solutions, the high current densities were also virtually independent of the rotational speeds, but in the more weakly basic solutions, with [NaOH]=0.05, the lower current densities showed a tendency to decrease with increasing rotational speeds. We speculate that the anode reaction may require a sequence of steps and that a crucial intermediate is swept away too fast in the weakly basic solutions.

Long-term, steady electrolyses were conducted to observe how the activated glassy carbon anodes might behave in practical applications. In one set of experiments, the starting electrolyte was 0.71M NaOH and 1.0M NaSH. The activated glassy carbon anode was rotated at 900 rpm. It was brought to a potential of 0.25 V, whereupon electrolysis at an anode-current density of 89 mA/cm² was observed. The anode functioned well for almost 13 hours. Then the current density dropped. After in-place reactivation, it carried 128 mA/cm² for almost 9 additional hours. After a second in-place reactivation, it carried 137 mA/cm² for over 5 hours. It was then polished and reactivated, whereupon it again carried 129 mA/cm² for almost 20 hours before it again experienced a substantial drop in current density.

In a second experiment with 0.71M NaOH and 1.0M NaSH to which 0.5 M/l S had been added, the glassy carbon anode carried 106 mA/cm² for almost 12 hours before it became deactivated. After reactivation

without polishing, it successfully carried 102 mA/cm² for over 28 hours before the experiment was terminated with the anode still functioning well. In additional experiments, in which we moved the reference electrode as close as we could (about 1 mm) to the working electrode, we observed that the current densities increased by 50-100% at a given voltage. This observation is consistent with a linear ohmic drop in the solution for the region from about -0.25 to +0.75 V(SCE) in Fig. 1b.

CONCLUSIONS

The electrolysis of H₂S can be carried out at high rates in strongly alkaline solutions. We observed our highest rates with high concentrations of NaSH in 6M NaOH. With activation we observed, at room temperature, anodic current densities substantially greater than those reported in a related study,¹² although it should be noted that the experimental conditions were different. Both studies suggest that the electrolysis can be carried out at high rates with a modest cell voltage of 1-1.5V.

In Table 2 we summarize and compare, insofar as a comparison between the two sets of experiments is possible, some pertinent results. The electrochemistry of anodic oxidation of sulfur is complicated. We cannot as yet describe with confidence the anode process, but thermodynamic arguments suggest that S²⁻ is the principal reactant and S₂²⁻ is the dominant polysulfide product.

We calculated, using the thermodynamic data of Ref. 13 and as-

suming activity coefficients of one, the concentrations (activities) of the species of interest in both studies. In Ref. 12, the solutions used were aqueous NaOH saturated with H_2S at 1 atm. The variations with $[\text{NaOH}]$ of the base 10 logarithms of the concentrations of the most significant species, excluding Na^+ , in such solutions are given in Fig. 9. The pH ranges from about 8 to 9. The dominant sulfur containing anion is SH^- . Its concentration is 4-5 orders of magnitude greater than $[\text{S}^{2-}]$. Figures 10-12 show the variations of the logarithms of the concentrations of the same species with $[\text{NaSH}]$ for three representative NaOH concentrations (0.1M, 1M, and 6M). The concentration of aqueous undissociated H_2S is also presented. The activity of H_2S in the concentrated solutions, which we here identify with the concentration, will be the same at all NaOH concentrations in Fig. 9 (namely, 0.102 mol/l) because they are all saturated with H_2S at one atmosphere. In the solutions that we found to be of greatest interest, the OH^- concentrations are all reasonably high. They are substantially more strongly basic than those used in Ref. 12. The pH ranged from about 12 to 15. The dominant sulfur-containing species is S^{2-} ; its concentration is two orders of magnitude greater than that of SH^- in 6M NaOH and ranges downward, in 1M NaOH, from one order of magnitude greater to nearly the same value as $[\text{NaSH}]$ ranges from 0.1 to 1.6. In 0.1M NaOH, the SH^- concentration goes above that of S^{2-} over a wide range of $[\text{NaSH}]$. Depletion of the surface concentration of S^{2-} would be expected at very high current densities. In the more concentrated solutions, a consideration of the diffusion of S^{2-} to the glassy carbon surface suggests that the limiting cur-

rent density would be about 1 amp/cm² if this species were the primary reactant.

It appears that passivation of the anode was avoided because of the anodic production of polysulfides from S²⁻ and their solubility in strongly basic solutions.

The interelectrode ohmic drop controlled the cell operation and limited the performance in both studies. In practice, minimizing the interelectrode spacing to reduce the ohmic drop will have to be balanced by the need to prevent soluble polysulfide anode products from reaching the cathode to be reduced parasitically. Either a cell separator or electrolyte flow could be used in a practical cell design. Tangential flow between the electrodes could be used to control the thickness of the boundary layer at the anode, but evolution of H₂ at the cathode would also cause mixing. The use of a separator would minimize mixing, but it would increase the ohmic drop between the electrodes.¹²

In any case, it is clear that the oxidation of sulfide is no longer the limiting factor in the electrolysis of hydrogen sulfide, since activated glassy carbon electrodes will support high oxidation rates. Further studies of the activation suggest that injection of solution species into the glassy carbon surface catalyzes the oxidation process.^{19, 21}

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Table 1. Activation procedures and associated current densities.

Procedure ^a	i^b , mA/cm ²
Activated in 0.5M NaOH. Then, at 0.25V(SCE), NaSH was added to make [NaSH]=1.55	29
Activated in 0.5M NaOH + 1.55M NaSH	58
Activated in 1.0M NaOH. Then, at 0.25V(SCE), NaSH was added to make [NaSH]=1.55	275
Activated in 1.0M NaOH + 1.55M NaSH	275
Activated in 2.0M NaOH. Then, at 0.25V(SCE), NaSH was added to make [NaSH]=1.55	263
Activated in 2.0M NaOH + 1.55M NaSH	307
a. The sweep rate was 5mV/s. The activation sweep went from -0.65V(SCE) to +2.25V(SCE) on the positive sweep and from +2.25V(SCE) to +0.25V(SCE) on the negative sweep. b. At +0.25V(SCE).	

Table 2. Anode current densities in various experiments.

[NaOH]	[S ²⁻] ^c	[SH ⁻] ^c	pH ^c	i^d , amps/cm ²
1.0 ^a	10 ⁻⁵	≈1	7.98	0.012
1.0 ^b	0.1-0.9	0.01-0.6	14-13	0.033-0.265
6.0 ^a	3.5x10 ⁻⁴	≈6	8.76	0.025
6.0 ^b	0.1-1	0.001-0.03	15.8-14.6	0.031-0.270
a. Ref. 12. aqueous NaOH, saturated with hydrogen sulfide. b. Present work. aqueous NaOH containing 0.05-1.55M NaSH. c. calculated value d. At 25°C and 0.25 V(SCE).				

Figure Captions

1. Schematic representation to show essential features of our linear scan voltammograms. (a) Voltammogram in which the the maximum potential is not high enough to produce activation. (b) Voltammogram in which the anode is activated.
2. Variation of current density with [NaSH] and rotational speed; [NaOH]=0.05.
3. Variation of current density with [NaSH] and rotational speed; [NaOH]=0.1.
4. Variation of current density with [NaSH] and rotational speed; [NaOH]=0.5.
5. Variation of current density with [NaSH] and rotational speed; [NaOH]=1.
6. Variation of current density with [NaSH] and rotational speed; [NaOH]=2.
7. Variation of current density with [NaSH] and rotational speed; [NaOH]=4.
8. Variation of current density with [NaSH] and rotational speed;

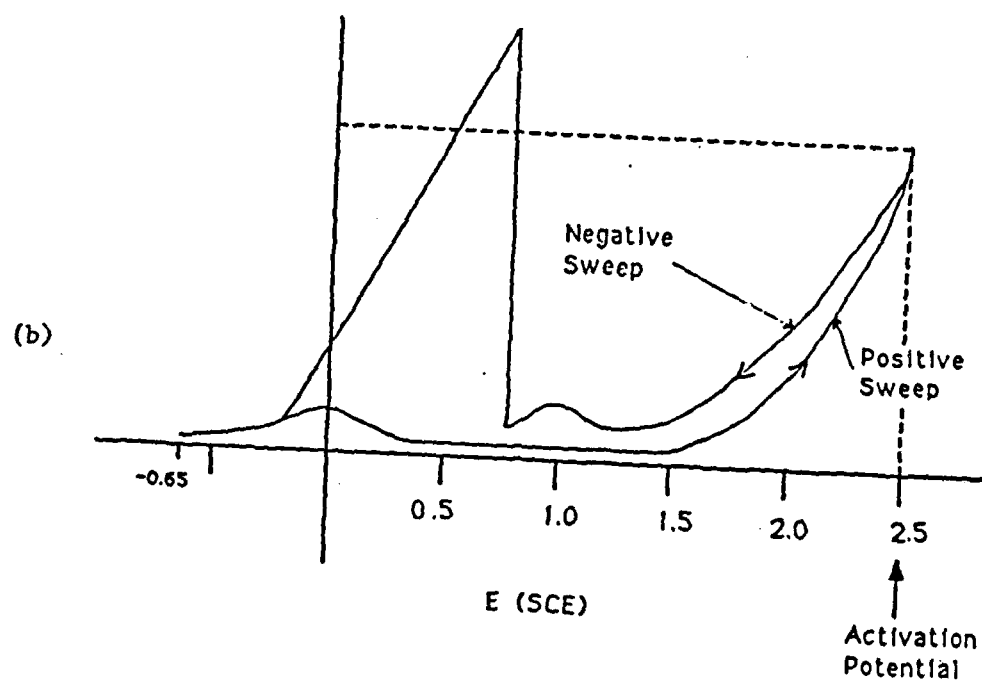
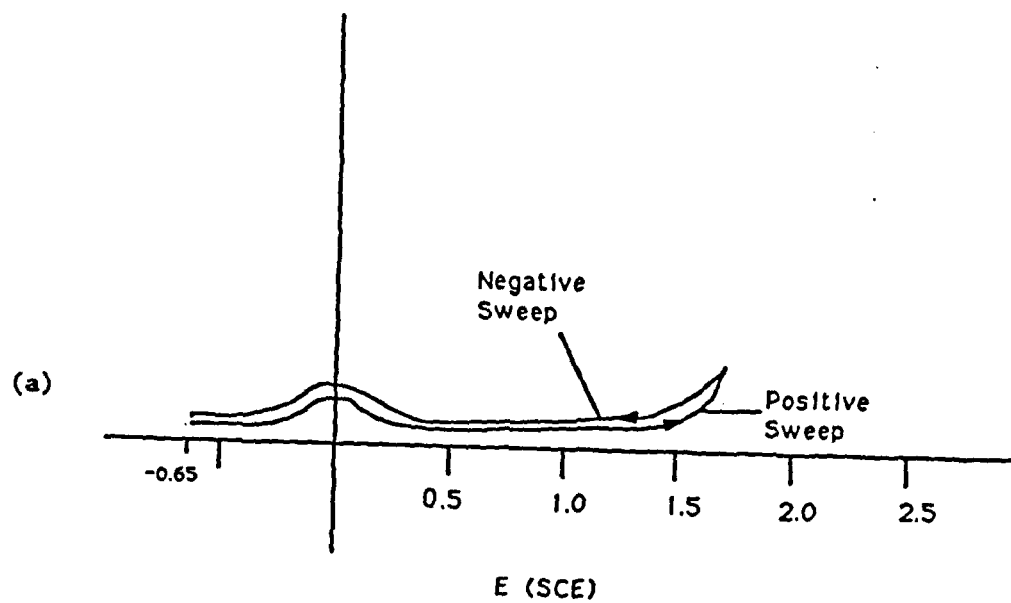
[NaOH]=6.

9. Variation of base 10 logarithms of the concentrations of the principal species of interest in saturated aqueous solutions of H_2S in NaOH with [NaOH]. The activity of $\text{H}_2\text{S}(\text{aq})$ is constant at all [NaOH] and is calculated to be 0.102 mol/l at 25°C .

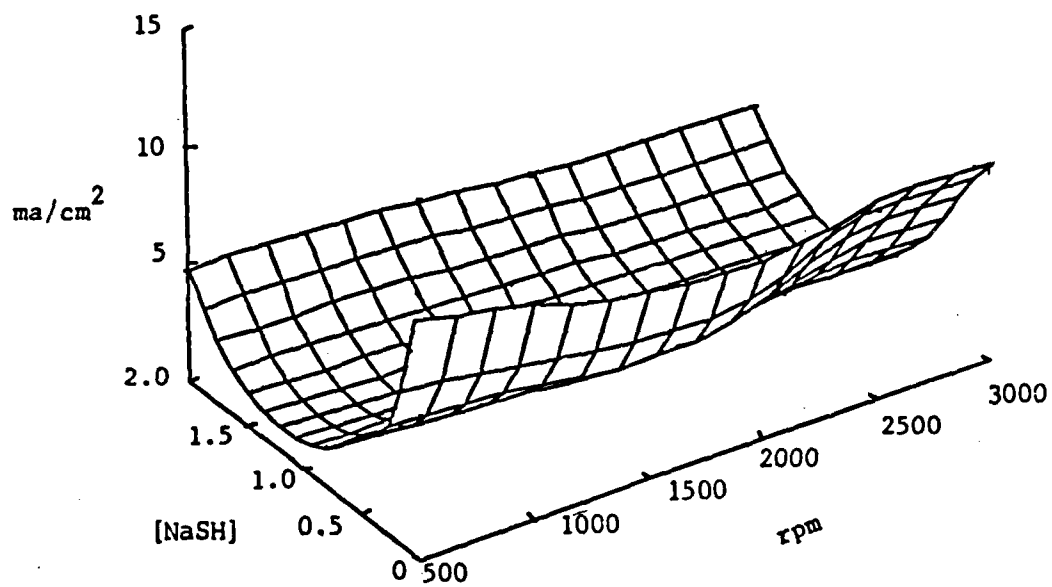
10. Variation of base 10 logarithms of the concentrations of the principal species of interest in aqueous 0.1M NaOH with [NaSH].

11. Variation of base 10 logarithms of the concentrations of the principal species of interest in aqueous 1.0M NaOH with [NaSH].

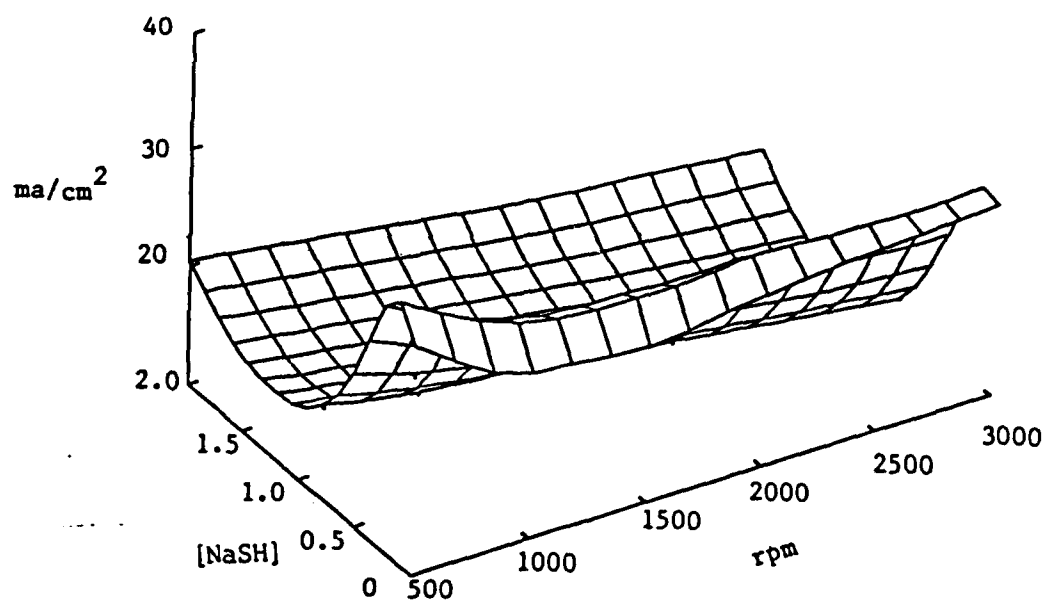
12. Variation of base 10 logarithms of the concentrations of the principal species of interest in aqueous 6.0M NaOH with [NaSH].



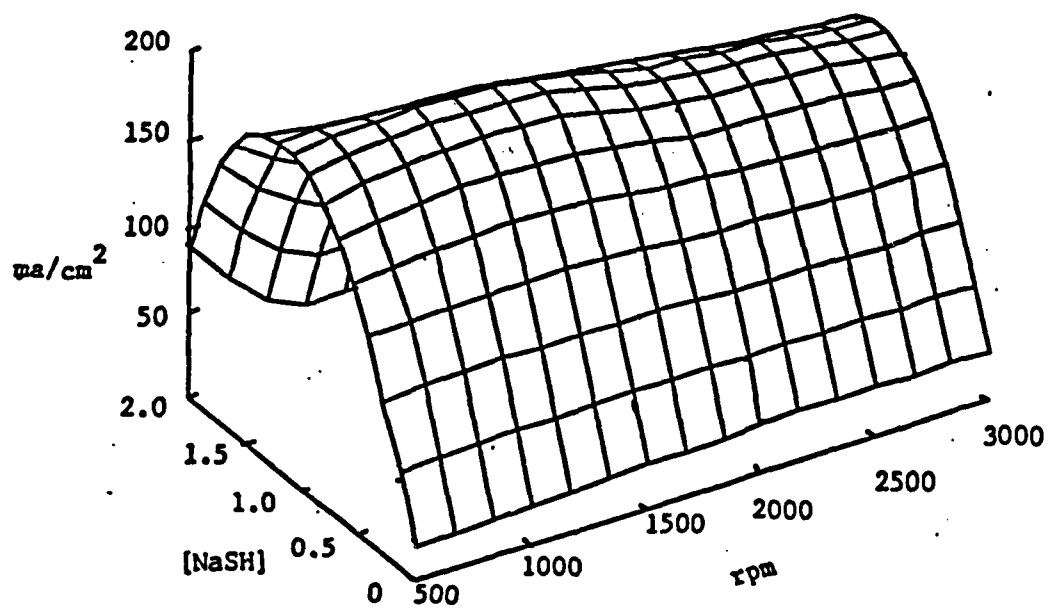
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Figure 1



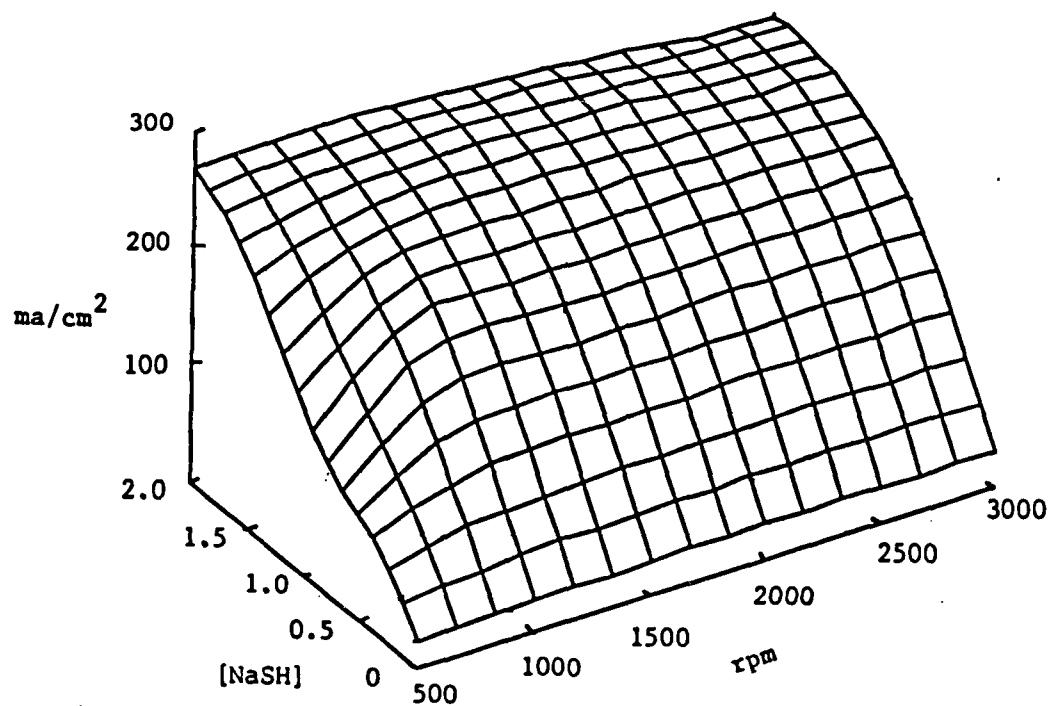
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Figure 2



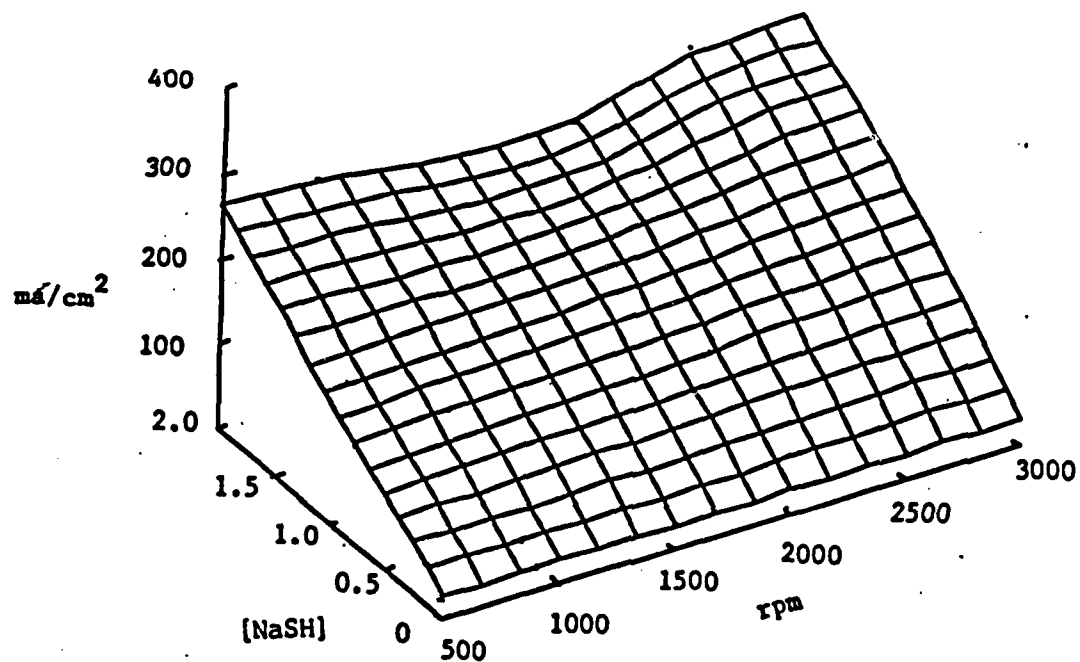
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Figure 3



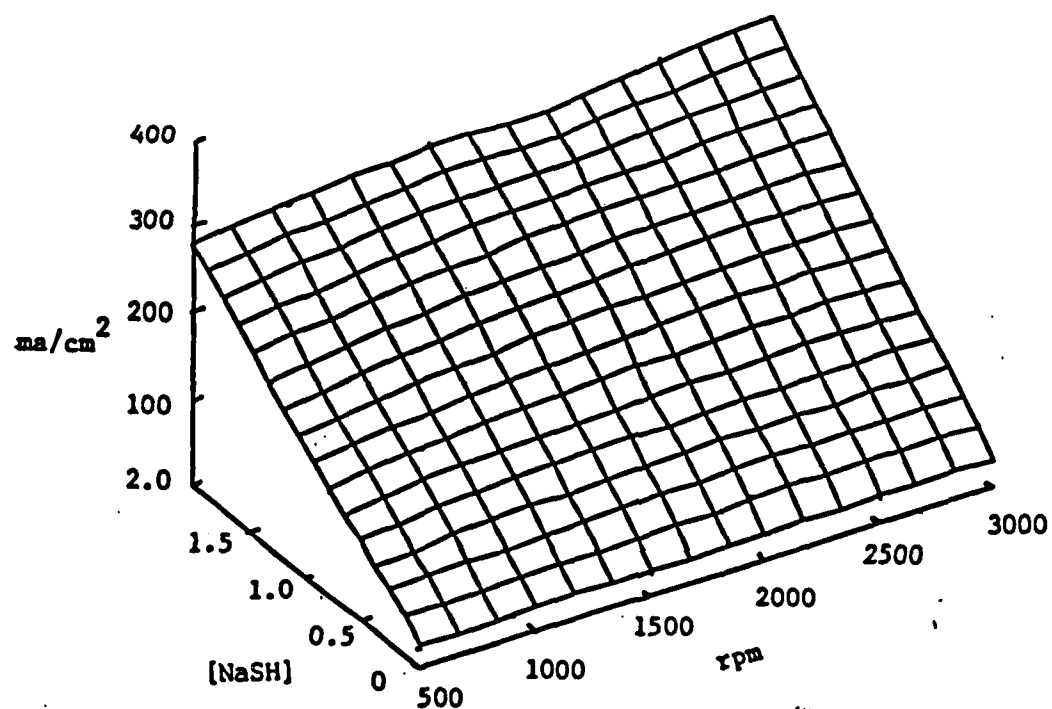
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Figure 4



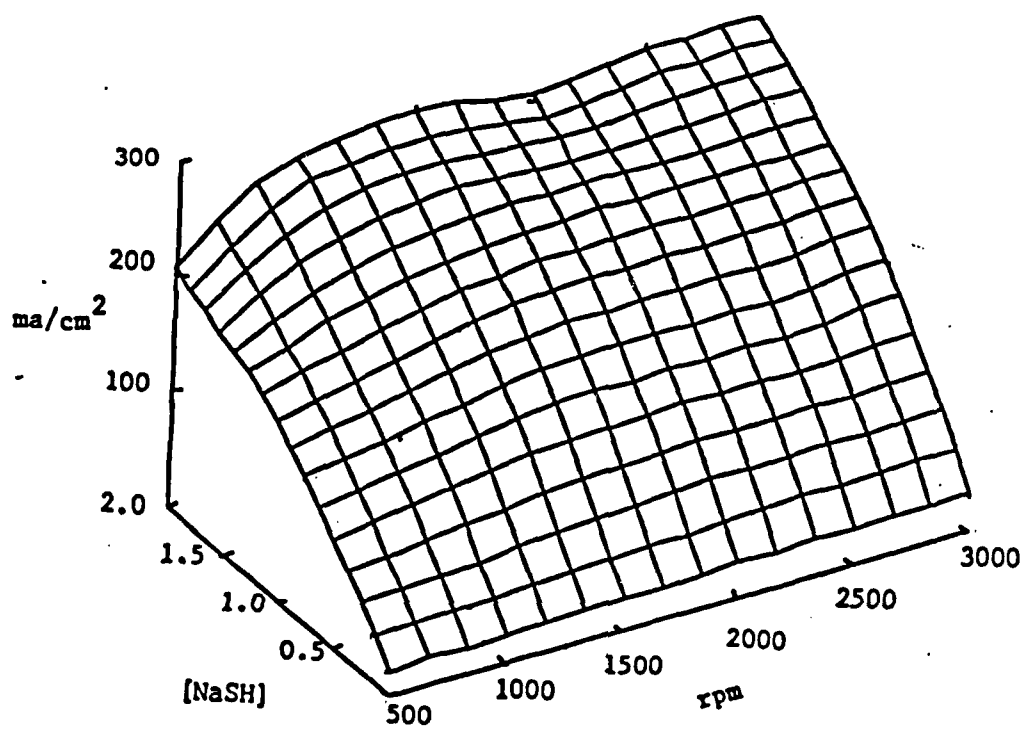
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Figure 5



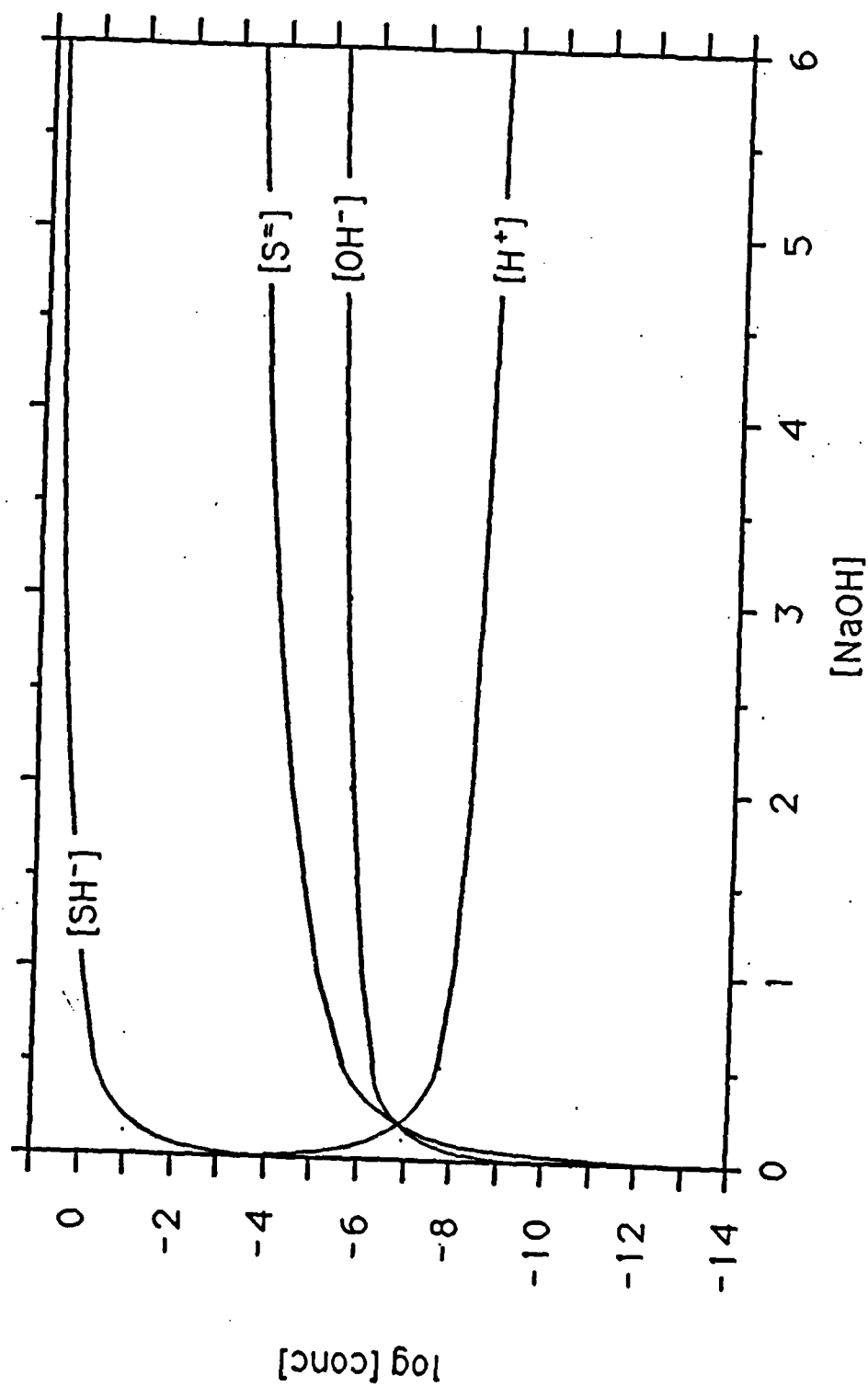
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Figure 6

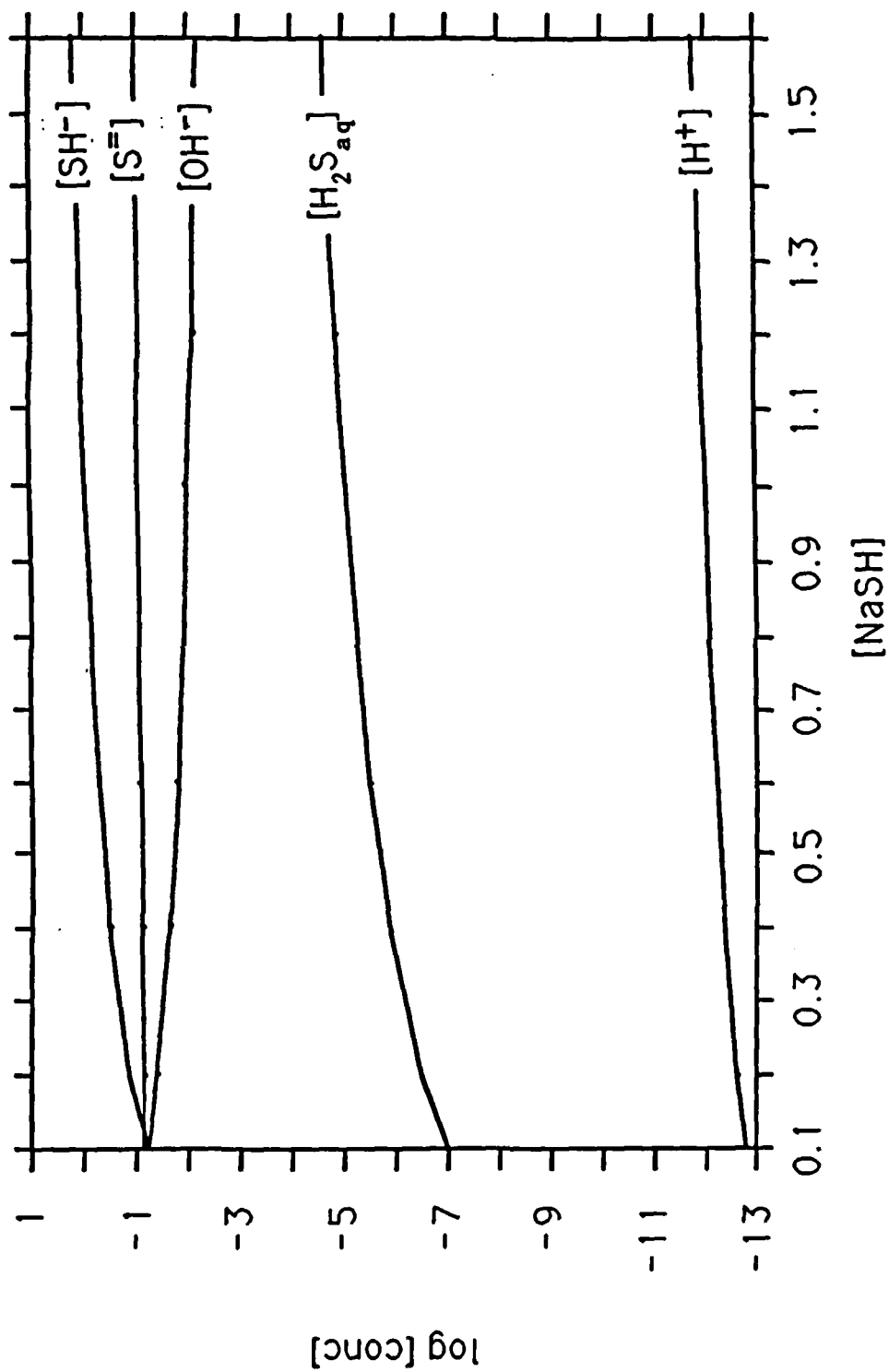


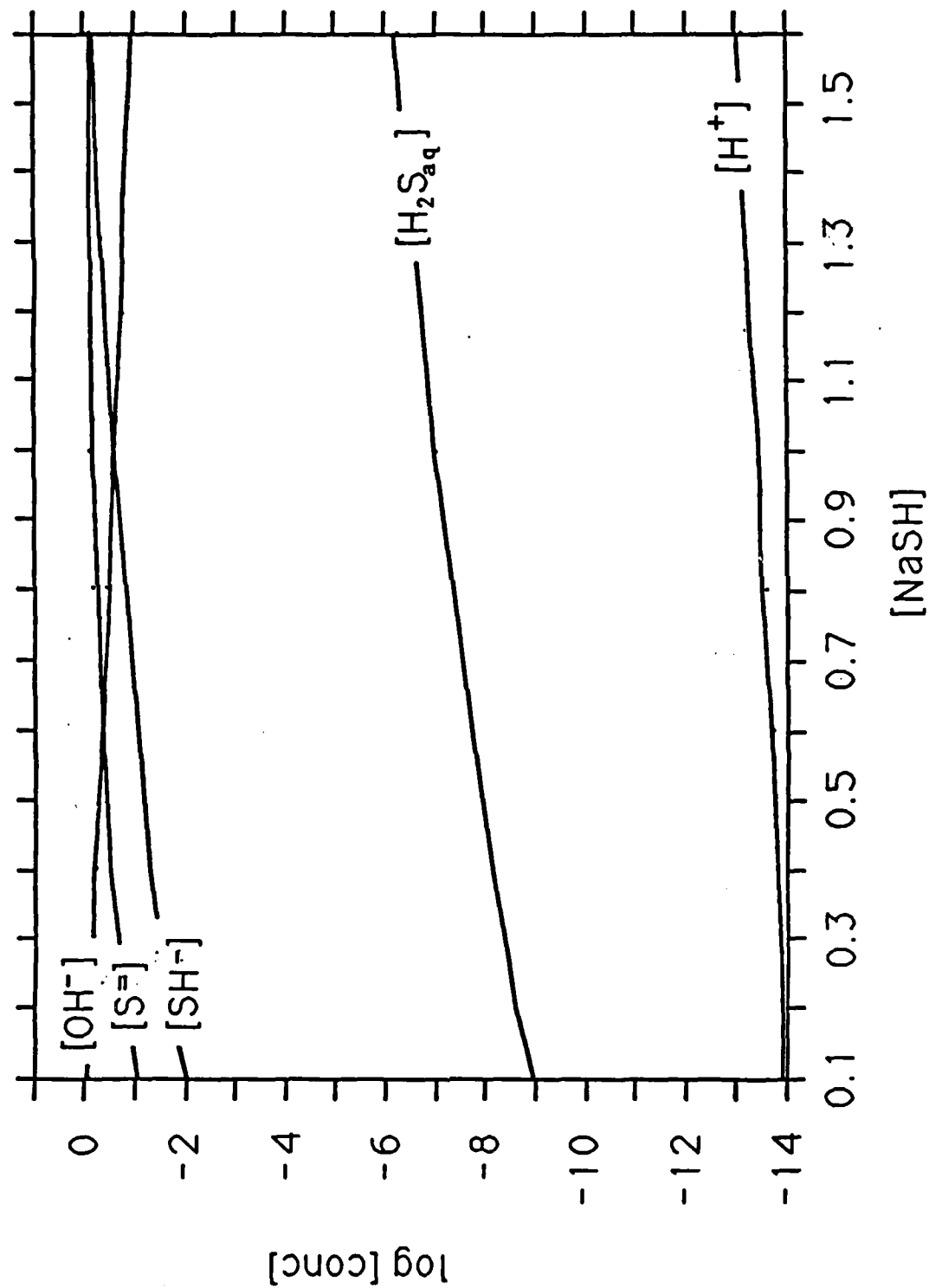
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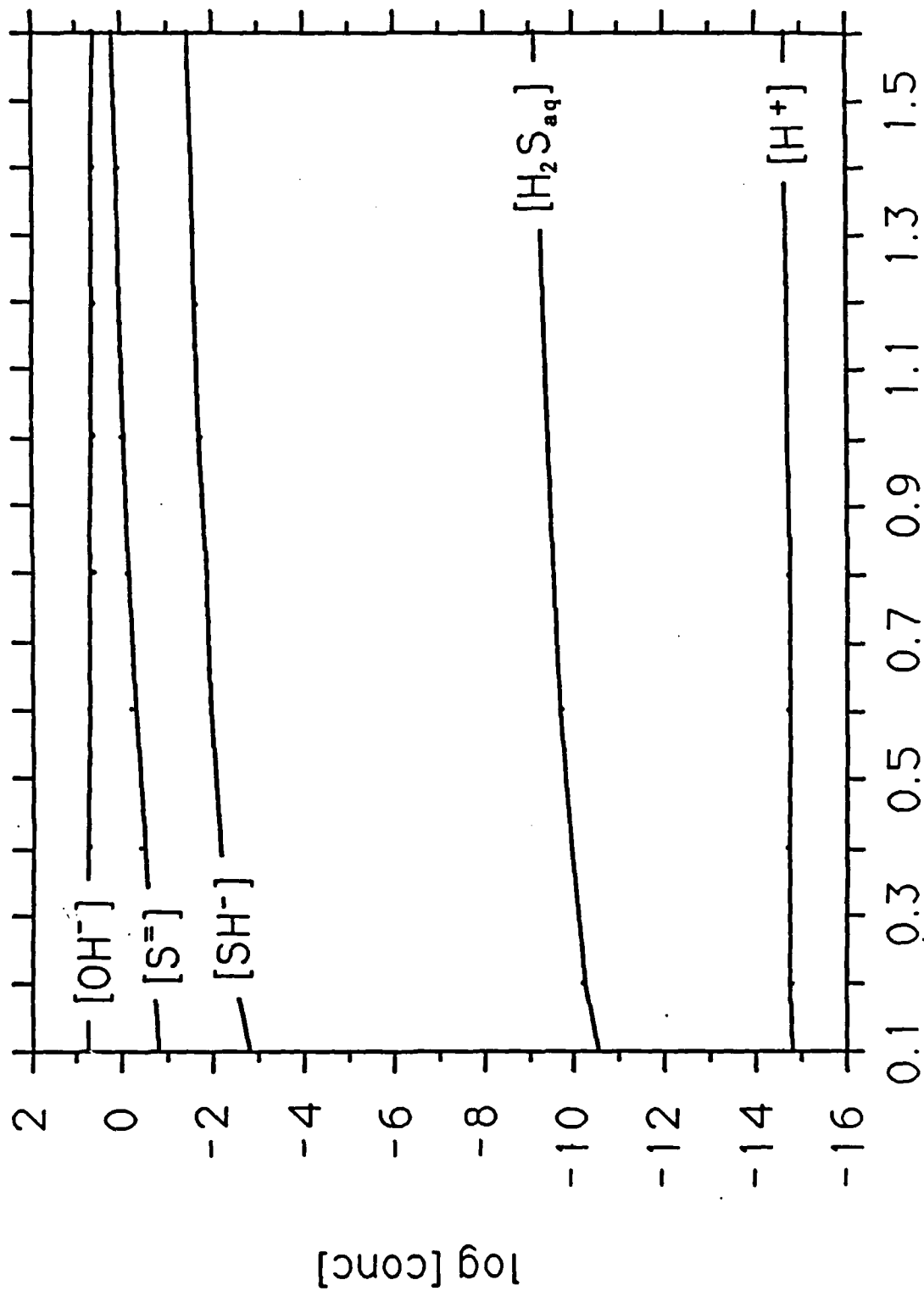
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Figure 8







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Figure 11



[NaSH]

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