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On the corrosion characteristics of some commercial magnetic metallic glasses in NaCl solution

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The corrosion characteristics of six commercial magnetic metallic glasses, namely $Fe_{78}B_{13}Si_9$ (Metglas 2605-S-2), $Fe_{67}Co_{18}B_{14}Si$ (Metglas 2605 Co), $Fe_{40}Ni_{40}B_{20}$ (Vitrovac 0040), $Fe_{39}Ni_{39}Mo_2(SiB)_{20}$ (Vitrovac 4040), $Fe_{40}Ni_{38}Mo_4B_{18}$ (Metglas 2826 MB) and $Fe_{40}Ni_{40}P_{14}B_6$ (Metglas 2826) in which iron and boron are common base elements have been studied by immersion tests and potentiostatic techniques in various concentrations of NaCl solutions at room temperature. In exposure for up to six days, weight loss observations indicated that the corrosion rates in 3.5% NaCl were in the following increasing order: 2826 < 4040 < 0040 < 2826 MB < 2605 Co < 2605-S-2. During exposure for up to four hours under open-circuit conditions, 2826 alloy showed the noblest behaviour while 2605-S-2 was the least noble. All the alloys, except 2826, showed rapid dissolution without any passivation during anodic polarization. Phosphorus addition to Fe–Ni–B glass enhanced the active dissolution and resulted in the formation of a black prepassive film in the active region (0.2 V) which becomes protective in the passive region (0.3 to 1.25 V). The values of $I_{crit.}$ and $I_{pass.}$ (the critical and passive current densities) decreased with a decrease of concentration from 3.5 to 0.01% NaCl, while in 0.001% NaCl $I_{pass.}$ slightly increased and the passivation potential range decreased. The corrosion resistance of Fe–B metallic glasses increased with metal addition in the order Co < Mo < Ni and with metalloid addition in the order Si < P.

1. Introduction

Amorphous alloys or metallic glasses are solid solutions in a chemically homogeneous, single-phase and disordered atomic state which prevent the occurrence of initiation sites for corrosion. Structural faults like grain boundaries, segregates, dislocations and imperfections, which contain a higher free energy and are thermodynamically unstable, are not present in these alloys. Due to the absence of heterogeneous characteristics in amorphous alloys, the corrosion resistance increases. It has been observed [1, 2] that in the absence of alloying elements like chromium and/or phosphorus with iron or nickel or cobalt base metallic glasses, very poor corrosion resistance is found but when about 8 at % Cr and 13 at % P is added [3, 4] the corrosion resistance developed is quite high, while in stainless steel 17 at % Cr is essential to obtain good corrosion resistance.

It has been found that both metal and metalloid addition [5–7] play an active role in improving the corrosion resistance of metallic glasses. Naka *et al.* [6, 7] have investigated the effect of metal and metalloid addition on the corrosion resistance of Fe–Cr alloys in acidic and neutral solutions. In their review [8], Hashimoto and Masumoto summarize the previous work leading to a proposal for the effect of alloying metal and metalloid to iron-base metallic glasses. However, the effect of phosphorus and molybdenum (with or without chromium) on the formation of a passive film have been investigated by several researchers [6–10] but the exact role played by certain metals and metalloids with iron-base metallic glasses still remains unclear. The effects of alloying metallic elements nickel, molybdenum and cobalt and metalloid silicon and phosphorus on Fe–B alloys are of interest to the present investigation, which has been carried out in sodium chloride solution of various concentrations using weight loss and electrochemical techniques. All the investigated alloys are of commercial importance [11–15] and hence the study of the corrosion characteristics of these alloys is worth examining.

2. Experimental procedure

In the present investigation, six Fe–B commercial metallic glasses were used for studying their corrosion characteristics in sodium chloride solutions. The trade names, compositions, dimensions and important uses of the alloys are recorded in Table I. No surface treatment was given to the specimens, which were used in the as-received condition after washing thoroughly with acetone and distilled water. Sodium chloride used was of AR grade and solutions were prepared in double distilled water. Rectangular specimens of 2 to 4 cm^2 surface area were used in both weight loss and polarization studies.

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TABLE I Metallic glasses studied with commercial applications

Trade name of alloy	Composition (at %)	Dimensions (mm)	Application
Metglas 2605-S-2	$Fe_{78}B_{13}Si_9$	50 × 0.025	Distribution transformers, low-frequency magnetic devices and other magnetic uses.
Metglas 2605 Co	$Fe_{67}Co_{18}B_{14}Si$	50 × 0.030	Highest saturation induction alloy, used in a wide variety of transformers, audiotable heads, high frequency circuits, magnetic tape heads of tape recorders and other applications where light weight is required.
Vitrovac 0040	$Fe_{40}Ni_{40}B_{20}$	15 × 0.025	Magnetic devices, brazing foil, flywheels, different types of composite etc.
Vitrovac 4040	$Fe_{34}Ni_{39}Mo_2(SiB)_{20}$	15 × 0.035	As above.
Metglas 2826	$Fe_{40}Ni_{40}P_{14}B_6$	25 × 0.035	Small-diameter cable wrap, composite magnetic devices, brazing foil etc.
Metglas 2826 MB	$Fe_{40}Ni_{38}Mo_4B_{18}$	50 × 0.040	Magnetic devices, power switching, signal and current transformers, composites, brazing foil, flywheels etc.

2.1. Immersion tests

For all the six alloys, the immersion tests were performed in 3.5% NaCl solution and up to 144 h exposure period. The specimen was suspended with the help of a polythene thread in 100 ml NaCl solution placed in a 150 ml conical flask which was closed with a cork. The specimens were removed at different intervals, dipped in 10% HCl for about 10 sec and then rubbed with cotton to remove all the corrosion products, finally washed with distilled water, dried and then weighed.

2.2. Electrochemical studies

The electrochemical studies were carried out in a glass cell using a platinum counter electrode and a saturated calomel reference electrode. A Wenking Potentioscan (Model POS73) was used for polarization experiments. The reference electrode was always facing the shiny side of the specimen and the distance between the tip of the reference electrode and the specimen was fixed at a minimum which did not show an overload. Working electrodes were prepared by joining a copper wire to one edge of the metallic glass strip and covering the joint with an epoxy resin.

During potentiostatic polarization, 3.5, 1.0, 0.01 and 0.001% NaCl solutions were used and the polarization cell was always open to the atmosphere. Initially the open-circuit potential (OCP) was noted after it attained a constant value with the passage of time (ca. 30 min). The specimen was then cathodically polarized in 50 mV steps up to − 1500 mV. The polarization currents were recorded after a steady value as each potential was reached. After cathodic polarization the specimen was again left in the OCP condition for about 30 min and then the anodic polarization was carried out.

3. Results and discussion

3.1. Immersion tests

The corrosion rate for six commercial metallic glasses (2605-S-2, 2605 Co, 0040, 4040, 2826 MB and 2826) calculated from the immersion tests for various exposure time in 3.5% NaCl are plotted in Fig. 1. Metallic glass 2605-S-2 shows the highest corrosion rate, whereas 2826 alloys have the least corrosion rate. Taking $Fe_{78}B_{13}Si_9$ (26C5-S-2) and $Fe_{40}Ni_{40}B_{20}$ (0040) as the base alloys, the effect of metal and metalloid additions on the corrosion rate in 3.5% NaCl solution is as follows.

On the addition of 18 at % Co and a reduction of 8 at % Si while boron remains almost constant ($Fe_{67}Co_{18}B_{14}Si$), the corrosion rate decreases by a factor of about three compared with no cobalt addition. During immersion both alloys show about the same dissolution pattern with time. This suggests that the addition of cobalt as an alloying metallic element to Fe–B metallic glass increases its corrosion resistance.

In the case of $Fe_{40}Ni_{40}B_{20}$ alloy the corrosion rate also continuously decreases with time and is reduced to one-half that of $Fe_{78}B_{13}Si_9$ alloy. This indicates that Fe–Ni metallic glasses possess better corrosion resistance in comparison to iron-base alloys.

The addition of 2 at % Mo and the partial replacement of boron by silicon in $Fe_{40}Ni_{40}B_{20}$ glass slightly

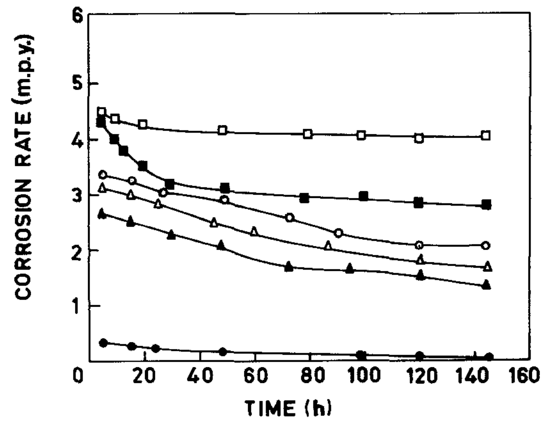

Figure 1 Corrosion rates obtained from the weight loss data for all six metallic glasses in 3.5% NaCl at 30° C: (●) $Fe_{40}Ni_{40}P_{14}B_6$ (2826), (○) $Fe_{40}Ni_{38}Mo_4B_{18}$ (2826 MB), (▲) $Fe_{39}Ni_{39}Mo_2$ $(SiB)_{20}$ (4040), (△) $Fe_{40}Ni_{40}B_{20}$ (0040), (■) $Fe_{67}Co_{18}B_{14}Si$ (2605 Co), (□) $Fe_{78}B_{13}Si_9$ (2605-S-2).

TABLE II Various corrosion parameters calculated from polarization curves

Corrosion parameter	NaCl (%)	$Fe_{78}B_{13}Si_9$ (2605-S-2)	$Fe_{67}Co_{18}B_{14}Si$ (2605 Co)	$Fe_{40}Ni_{38}Mo_4B_{18}$ (2826 MB)	$Fe_{40}Ni_{40}B_{20}$ (0040)	$Fe_{39}Ni_{39}$-$Mo_2(SiB)_{20}$ (4040)	$Fe_{40}Ni_{40}P_{14}B_6$ (2826)
$E_{corr.}$ mV (SCE)	3.5	−720	−670	−440	−405	−370	−250
	1.0	−705	−645	−425	−380	−360	−240
	0.1	−690	−635	−380	−360	−350	−210
	0.01	−680	−620	−360	−340	−330	−190
	0.001	−670	−565	−340	−315	−310	−175
$I_{corr.}$ ($A\,cm^{-2}$)	3.5	35.0	27.0	12.0	7.8	7.5	2.40
	1.0	26.5	22.0	9.5	6.5	6.0	2.0
	0.1	15.0	12.0	4.2	3.8	3.5	1.35
	0.01	11.0	9.0	3.0	2.6	2.2	0.90
	0.001	7.5	4.5	2.5	1.8	1.65	0.30
$I_{crit.}$ ($mA\,cm^{-2}$)	3.5	No passivation	No passivation	No passivation	No passivation	No passivation	4.5
	1.0	No passivation	No passivation	No passivation	No passivation	No passivation	3.0
	0.1	No passivation	No passivation	No passivation	No passivation	No passivation	0.95
	0.01	No passivation	No passivation	No passivation	No passivation	No passivation	0.55
	0.001	No passivation	No passivation	No passivation	No passivation	No passivation	0.40
$I_{pass.}$ ($mA\,cm^{-2}$)	3.5	No passivation	No passivation	No passivation	No passivation	No passivation	0.70
	1.0	No passivation	No passivation	No passivation	No passivation	No passivation	0.35
	0.1	No passivation	No passivation	No passivation	No passivation	No passivation	0.25
	0.01	No passivation	No passivation	No passivation	No passivation	No passivation	0.20
	0.001	No passivation	No passivation	No passivation	No passivation	No passivation	0.30
Passivation range (mV (SCE))	3.5	No passivation	No passivation	No passivation	No passivation	No passivation	+300 to +1200
	1.0	No passivation	No passivation	No passivation	No passivation	No passivation	+300 to +1250
	0.1	No passivation	No passivation	No passivation	No passivation	No passivation	+250 to +1350
	0.01	No passivation	No passivation	No passivation	No passivation	No passivation	+1200 to +1450
	0.001	No passivation	No passivation	No passivation	No passivation	No passivation	+200 to +600

decreases the corrosion rate. Naka *et al.* [6] have also reported that silicon addition decreases the corrosion rate of metallic glasses in NaCl solution, in comparison to the effect of boron.

The addition of 4 at % Mo to $Fe_{40}Ni_{40}B_{20}$ alloy, replacing 2 at % Ni and 2 at % B, increases the corrosion rate quite appreciably. This indicates that molybdenum addition is less effective in improving the corrosion resistance than nickel addition.

Similarly, replacement of a major part of boron by phosphorus in $Fe_{40}Ni_{40}B_{20}$ glass reduces the corrosion rate by about 20 times. This is perhaps due to the dissolution of phosphorus from the surface of the alloy as phosphate which accumulates at the alloy solution interface, which in turn is responsible for the decrease in the corrosion rate [9]. Though silicon and boron additions to the alloy lead to the formation of silicate and borate along with the oxides of the alloy they do not give good protection.

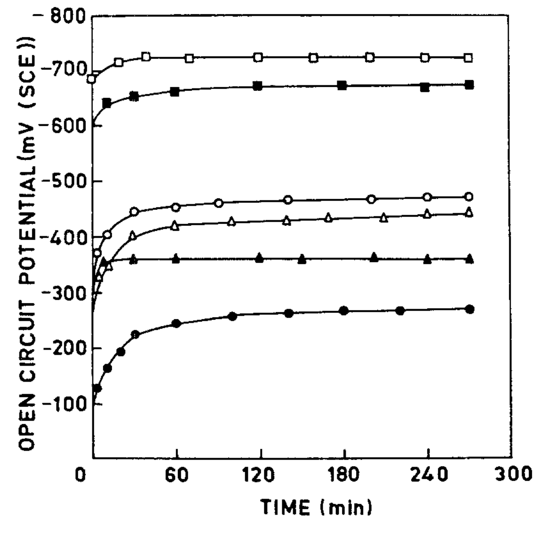

Figure 2 Variation of open-circuit potential with exposure time for all six metallic glasses in 3.5% NaCl solution at 30° C: (●) $Fe_{40}Ni_{40}P_{14}B_6$ (2826), (○) $Fe_{40}Ni_{38}Mo_4B_{18}$ (2826 MB), (▲) $Fe_{39}Ni_{39}Mo_2$ $(SiB)_{20}$ (4040), (△) $Fe_{40}Ni_{40}B_{20}$ (0040), (■) $Fe_{67}Co_{18}B_{14}Si$ (2605 Co), (□) $Fe_{78}B_{13}Si_9$ (2605-S-2).

In all the six alloys, initially the corrosion rate was high but after 20 to 30 h exposure in 2605-S-2 and 2605 Co and 40 to 50 h in 2826, 2826 MB, 0040 and 4040, a black to greenish-black film (2826 and 2605 Co) or a yellow film (2626 MB, 0040, 4040 and 2605-S-2) is formed on the surface. These are corrosion product films which reduce the further dissolution of the alloys, and the corrosion rate becomes almost constant after about 60 h immersion. The overall results also indicate that initial dissolution is an essential requirement for the formation of a protective film.

The corrosion rates of $Fe_{67}Co_{18}B_{14}Si$ (2605 Co) and $Fe_{78}B_{13}Si_9$ (2605-S-2) alloys are very high in comparison to those of Fe–Ni-base metallic glasses. However, these two alloys have interesting properties such as large saturation magnetization and Invar characteristics [11].

3.2. Open-circuit potentials

All the six alloys showed an almost constant OCP during a 250 min exposure study. Initially there was a slight shift towards the active direction but it stabilized after 40 to 50 min (Fig. 2). The stabilized OCP values for all the alloys in various concentrations of NaCl solution (3.5, 1.0, 0.1, 0.01 and 0.001%) are given in Table II. Based on the OCP values the metallic

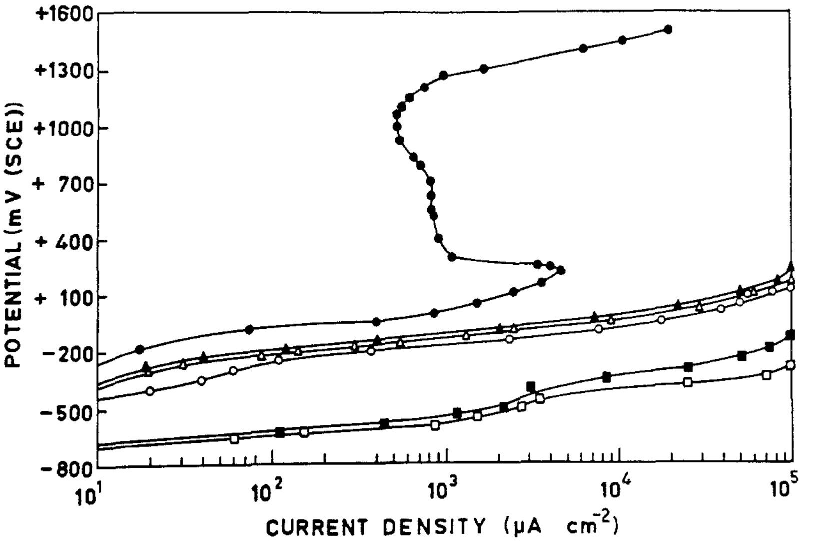

Figure 3 Anodic polarization curve for all the investigated alloys in 3.5% NaCl at 30° C: (●) $Fe_{40}Ni_{40}P_{14}B_6$ (2826), (○) $Fe_{40}Ni_{38}Mo_4B_{18}$ (2826 MB), (▲) $Fe_{39}Ni_{39}Mo_2$ $(SiB)_{20}$ (4040), (△) $Fe_{40}Ni_{40}B_{20}$ (0040), (■) $Fe_{67}Co_{18}B_{14}Si$ (2605 Co), (□) $Fe_{78}B_{13}Si_9$ (2605-S-2).

glasses may be graded as follows:

(Noblest) 2826 → 4040 → 0040 → 2826 MB →

2605 Co → 2605-S-2 (least noble)

3.3. Anodic polarization behaviour

Fig. 3 shows the anodic polarization behaviour of the six alloys in 3.5% NaCl solution. All the alloys except 2826 fail to show any transition from the active to the passive state. Various corrosion parameters obtained from the polarization curves in different concentrations of NaCl solution (3.5, 1.0, 0.1, 0.01 and 0.001%) are given in Table II. The corrosion current densities of the alloys ($I_{corr.}$) were calculated by extrapolating the anodic and cathodic Tafel lines.

In the case of 2826 alloy, the passivation state is observed in almost all the concentrations of NaCl solution (Fig. 4 and Table II). The passivation potential range for this alloy slightly decreases with an increase of concentration from 0.01 to 3.5% NaCl, whereas in the case of 0.001% NaCl solution the passivation potential range becomes very small. The magnitude of the passive current density ($I_{pass.}$) and the critical current density ($I_{crit.}$) for passivation decreases with the NaCl solution concentration. No crevice corrosion was observed on any specimen. Pitting was observed in all cases except 2826 alloy, even under open-circuit potential conditions. In 2826 alloy pits were observed only after increasing the potential beyond the passive region during anodic polarization.

For all the alloys, the open-circuit potential shifts towards the noble direction and the corrosion current decreases with a decrease of NaCl solution concentration from 3.5 to 0.001%. The influence of various alloying elements like molybdenum, cobalt, nickel, silicon and phosphorus added to Fe–B metallic glasses on the corrosion behaviour of the alloys may be explained as given below.

3.3.1. Effect of molybdenum addition

No passivation is observed in 2826 MB and 4040 alloys in any concentration of NaCl solution (Fig. 3 and Table II). However, in dilute H_2SO_4 solution, it has been found [7, 9, 16] that molybdenum addition produces passivation during anodic polarization by the formation of a diffusion barrier which prevents further dissolution of the alloy in the potential range from 0.5 to 1.5 V. It has also been reported [7] that molybdenum addition in the presence of chromium and phosphorus to Fe–Ni glasses produced a highly protective film and that molybdenum helps in the repair of the passive film.

The passive film of pure crystalline molybdenum is composed of MoO_2 and the reaction product of the transpassivation is MoO_4^{2-} ions [17–18]. Accordingly, the transpassive reaction for molybdenum in a neutral

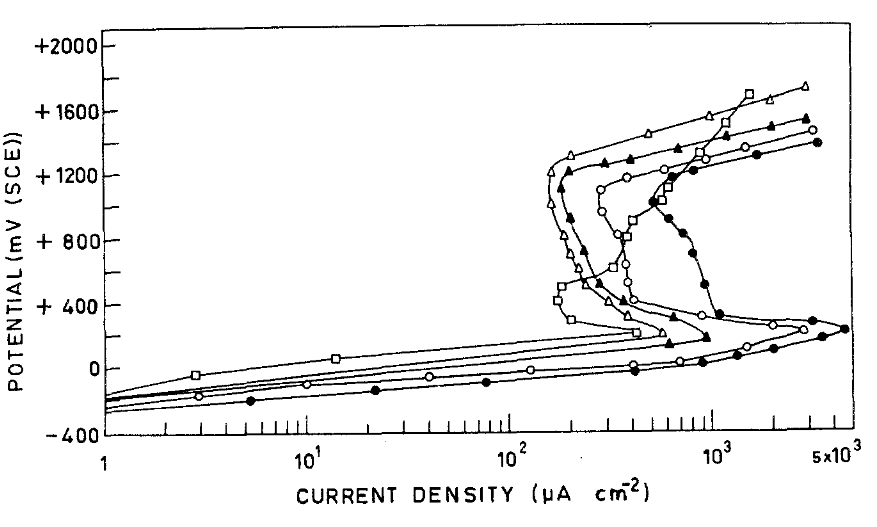

Figure 4 Anodic polarization curve for 2826 alloy at 30° C in sodium chloride solutions of various concentrations: (●) 3.5%, (○) 1.0%, (▲) 0.1%, (△) 0.01%, (□) 0.001%.

solution of pH higher than 6, and its equilibrium potential, may be represented as follows [19]:

$$MoO_2 + 2H_2O = MoO_4^{2-} + 4H^+ + 2e^- \qquad (1)$$

$$E = 0.606 - 0.118 + 0.295 \log a \, MoO_4^{2-} \qquad (2)$$

From the above equations, it is clear that in a near-neutral solution ($pH > 6$) MoO_4^{2-} ions should be more stable than MoO_2 and there is no possibility of passivation.

Another possibility is the formation of a passive film containing MoO_3, but it has also been reported [19] that when the pH is higher than 3.7, the MoO_3 dissolves as $HMoO_4^-$ or MoO_4^{2-} ions in solution. The present results show that the addition of molybdenum to Fe–Ni metallic glasses does not improve the corrosion resistance in sodium chloride, neither during polarization at higher anodic potentials nor under open-circuit condition.

3.2.2. Combined effect of molybdenum and silicon addition

The presence of silicon is not effective in the formation of a passive film with molybdenum (Fig. 3), but it slightly decreases the corrosion rate (Table II) in comparison to that of the alloy not containing silicon. Though molybdenum tends to increase the corrosion, in the presence of silicon the dissolution is reduced due to the formation of silicon oxide. It is known [9, 10, 16] that molybdenum helps the passivation of Fe–Ni glasses in the presence of metalloids like phosphorus, silicon and boron by synergistic interaction among polyanions of these elements in H_2SO_4, but this type of interaction is not possible in NaCl solution because molybdenum itself is not stable. The corrosion rates of 0040, 2826 MB and 4040 alloys are in the following order:

$$4040 < 0040 < 2826\ \mathrm{MB}$$

It can, therefore, be concluded that silicon is more effective in increasing the corrosion resistance in comparison to boron in NaCl solution.

3.3.3. Effect of phosphorus addition

Fig. 4 shows the anodic polarization curve for $Fe_{40}Ni_{40}P_{14}B_6$ (2826) alloy, where phosphorus replaces a major part of boron in the $Fe_{40}Ni_{40}B_{20}$ alloy. As seen from Fig. 1 and Table II, the phosphorus-containing alloy corrodes at about a one order of magnitude lower rate than Fe–Ni–B alloy under open-circuit condition. $I_{\mathrm{crit.}}$ and $I_{\mathrm{pass.}}$ decrease with the concentration of NaCl solution from 3.5 to 0.01%, but in 0.001% NaCl solution $I_{\mathrm{pass.}}$ slightly increases and the passivation potential range is also smaller.

A black prepassive film was observed during anodic polarization of 2826 alloy until the $I_{\mathrm{crit.}}$ stage in NaCl solution. Such a film has been observed by many workers [9, 16, 20, 21] in H_2SO_4 as well. It has been found that this prepassive film on 2826 alloy consists of oxides of iron and nickel and it is enriched with oxidized phosphorus:

$$3Ni^{2+} + 4H_2O = Ni_3O_4 + 8H^+ + 2e^- \qquad (3)$$

$$3Fe^{2+} + 4H_2O = Fe_3O_4 + 8H^+ + 2e^- \qquad (4)$$

Recently Flis and Duquette [22], on the basis of energy-dispersive X-ray analysis (EDXA) for Ni–P metallic glasses in NaCl solution, observed that precipitation of a phosphorus-containing corrosion product occurs at the surface of the alloys. Sorensen *et al.* [21] found that the active–passive transition is the result of some process other than passivation of the metal surface. In the active region, the surface rapidly becomes black. This black film cannot be removed either by the stirring of the solution or by rubbing the surface with cotton, indicating that it is the product of a chemical change. At the more noble potential (passive region) the current drops markedly and is associated with the formation of a white film. Further, analysis [9, 21] of the black and white films revealed that the nickel content of the film increases at a more active potential (0.2 V) and this film changes to a thin white film; the nickel content decreases and the iron content increases over the passivation range (0.3 to 1.25 V). The prepassive black film may consist of nickel phosphate, $Ni_3(PO_4)_2 \cdot 8H_2O$, which is less protective, and the passive white film of iron phosphate, $Fe_3(PO_4)_2 \cdot 8H_2O$, which is more protective in nature.

This shows that phosphorus addition to an Fe–Ni–B alloy increases the corrosion resistance under open-circuit potential conditions and provides a good passive film during anodic polarization.

It has earlier been reported [23, 24] that the effect of phosphorus addition to Fe–Cr glasses is to stimulate the active dissolution of chromium as Cr^{3+} ion which is directly responsible for the passive film formation. Similar results have also been reported [22] for Ni–P metallic glasses in NaCl solution, that phosphorus enhanced the anodic dissolution of nickel. Rapid anodic dissolution of the alloy in the active region is essential for the accumulation of film-forming elements near the alloy surface to form a good passive film. On this basis, it is assumed that in 0.001% NaCl solution the lower dissolution rate does not allow the formation of a uniform passive film on the surface. Surface analysis techniques have also been employed to evaluate the composition of the passive film formed on Fe–Ni–Cr–P–B amorphous alloys [25, 26]. Asami *et al.* [25] observed that the passive film consists of hydrated chromium oxyhydroxide, ferrous and ferric ions, trace amounts of nickel and PO_4^{3-} with bound water, while other workers [26] showed a substantial enrichment of phosphorus. The polarization data from Table II suggest that the higher rate of active dissolution of the alloy in higher concentrations of NaCl solution leads to the enrichment of passivating species at noble potentials and produces a better passivation.

3.3.4. Effect of cobalt and silicon addition

Fig. 3 shows the anodic polarization curve of $Fe_{78}B_{13}Si_9$ (2605-S-2) and $Fe_{67}Co_{18}B_{14}Si$ (2605 Co) metallic glasses in 3.5% NaCl solution, while their corrosion parameters in various concentrations of NaCl solution are given in Table II. These two commercial metallic glasses, produced for their superior magnetic properties, have been found to dissolve actively in NaCl solution during polarization and immersion test investigations. These two alloys show about a two orders of magnitude lower corrosion rate in 3.5% NaCl solution than in 0.1 M H_2SO_4 solution, the results for which have been published elsewhere [16]. The lower corrosion rate in NaCl may be due to the precipitation of a corrosion product film, but in the case of H_2SO_4 solution such a protective film cannot be formed at all because of the lower pH (< 6). During the anodic polarization no passivation occurs, but the corrosion current decreases for both alloys with a decrease in NaCl solution concentration. $I_{\text{corr.}}$ value for these two alloys have been found to be about three to four times higher than those of Fe–Ni–B metallic glasses.

4. Conclusions

1. The addition of metallic elements to Fe–B metallic glasses improves the corrosion resistance in the order Co < Mo < Ni.

2. Phosphorus is more effective than silicon in decreasing the corrosion rate of Fe–B metallic glasses.

3. All the metallic glasses, except 2826, fail to show passivation in NaCl solution.

4. Initial active dissolution of 2826 alloy is essential for the formation of a passive film.

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