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Effect of Aluminum Addition to Iron-Phosphorus Alloys Used For Soft Magnetic Applications.

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ABSTRACT

The present investigation deals with designing Fe-P based ternary alloys produced by in-house developed powder metallurgical technique based on 'Hot Powder Preform Forging'. Proper soaking of preforms at high temperature (1050°C) eliminates iron-phosphide eutectic and brings entire phosphorus into solution in iron. Attempting hot forging thereafter completely eliminates hot as well as cold shortness and thereby helps to form these preforms into very thin sheets of 0.5mm. The use of costly hydrogen atmosphere during sintering has been eliminated by use of addition of carbon. The glassy ceramic coating applied over the compact serves as a protective coating to avoid atmospheric oxygen attack over the compact held at high temperature. The alloy so formed was subjected to density examination at various stages. Microstructural study has been carried out to estimate the grain size, volume percentage porosity in the alloy and uniform distribution of phosphorus in iron matrix. Fe-0.28wt%O-0.07wt%C-0.5wt%P-0.5wt%Al alloy so formed yielded coercivity as low as 0.33 Oe, resistivity as high as 28.4 $\mu\Omega\text{cm}$ and total loss as low as 0.142 W/Kg. The alloy which is drawn to thin sheets could find its possible application in manufacturing transformer cores.

Keywords: aluminium, alloy, magnetic applications.

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INTRODUCTION

Iron–phosphorous alloy system for the production of magnetic materials is not known in wrought processing route in spite of the fact that phosphorous as an alloying element has all the favourable characteristics to enhance the magnetic properties of iron [1]. This because of the fact that in wrought route it is not possible to bring phosphorous in a homogeneous solid solution with iron due to its segregation tendency during solidification of the melt. Powder metallurgical processing, due to its predominantly solid state processing approach, has been very successful in exploiting the potential of Fe–P alloy system for magnetic applications [2]. Conventional powder metallurgy processes enable iron powder to be compacted to 7.1 g/cm³ or 93% of the theoretical density [3]. Compaction and sintering of these alloys result in heavy volume shrinkage [4]. Double pressing achieves higher density but at increased cost. Existence of prior particle boundaries (PPBs) renders Hot Isostatic Pressing (HIP) unsuitable for magnetic applications. In view of this, in the present investigation, densification is carried out by a cost effective hot powder preform forging technique. The process renders highest possible densification without resorting to hydrogen as sintering atmosphere. It is essentially the process where shaping and consolidation are deformation based. This causes redistribution of segregants if at all these remained at the particle surfaces (deformation can displace these from grain boundary and disintegrate them to fine particles which easily dissolve inside the ferritic grains). Such a processing route provides superior magnetic properties in comparison to existing sintering approach.

MATERIALS AND METHOD

The iron powder consists of ASC 100. 29 atomized iron powder supplied by M/S Hognas India Ltd., with a nominal carbon 0.01 wt% and hydrogen loss value 0.1% with -100 mesh size. Iron-phosphide powder (C-0.01 wt %) (size -100 mesh) was prepared by reacting iron powder with ortho-phosphoric acid and a subsequent thermal treatment (800° C) to yield Fe₃P coating over iron powder (Fig.1). The chemical reactions are:

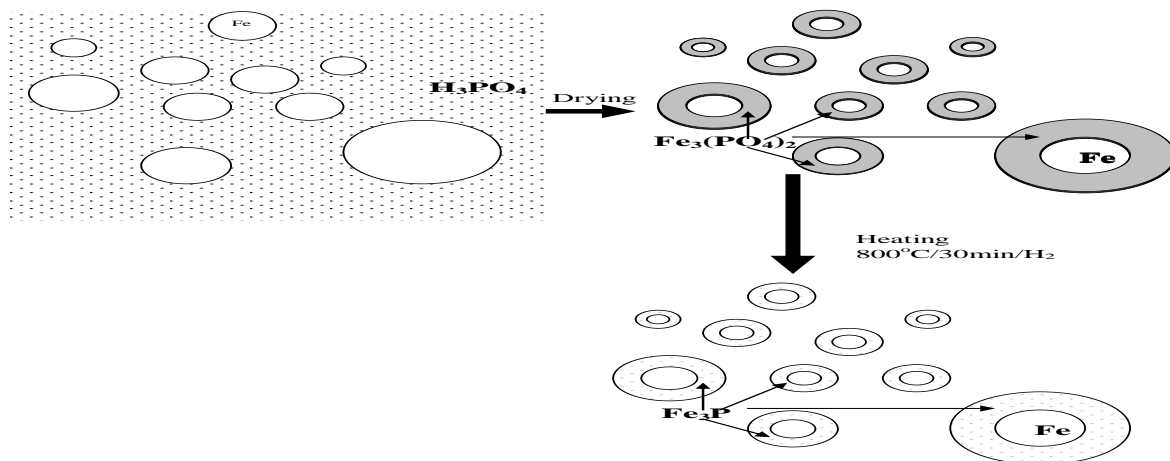
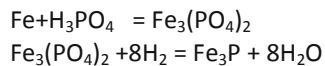


Figure 1: Preparation of Fe₃P powder

The powders are suitably mixed with 0.3wt% of carbon (in the form of graphite; size -100 mesh) to yield the following chemistry:

- (a) Fe-0.3wt%P-0.3wt% Al
- (b) Fe-0.3wt%P-0.5wt% Al
- (c) Fe-0.5wt%P-0.3wt% Al
- (d) Fe-0.5wt%P-0.5wt% Al

The powder mix was filled in a rectangular die (Fig. 2) and the green compacts (preform) with 7mm thickness and 25mm x 50mm size were formed using hydraulic press.

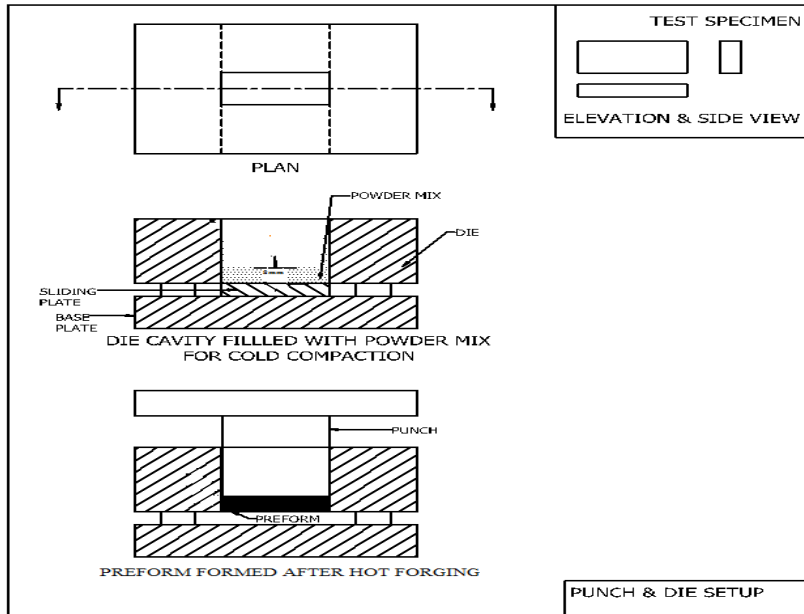


Figure 2: Schematic diagram of die and punch set-up

The oxidation resistant glassy coating was applied on the surface of the preforms like paint with a brush [5]. The coating employed is an alumino-silicate glassy ceramic coating containing carbonaceous constituent also which liquefies at high temperature and wets the metallic surface over which it is applied. The glassy coating is neutral to metallic surface and hence there is no chemical interaction between metallic surface and glassy coating at high temperature. However it acts as a barrier to oxidizing atmosphere, prevailing inside the furnace where samples are heat treated. Coated preforms were baked at 120° C for two hours. These preforms were then transferred to a furnace held at a temperature 1050° C and soaked there for one hour. The iron powder particle is 100% gamma-phase at this temperature. Phosphorous combines with this gamma iron powder particle and dissolves in it. As it dissolves, it gets converted into ferrite (Fig. 3) and as ferrite phase grows out of gamma phase, more and more phosphorous penetrates in it. Hence entire phosphorus is dissolved into iron to yield homogeneous ferrite phase [6,7].

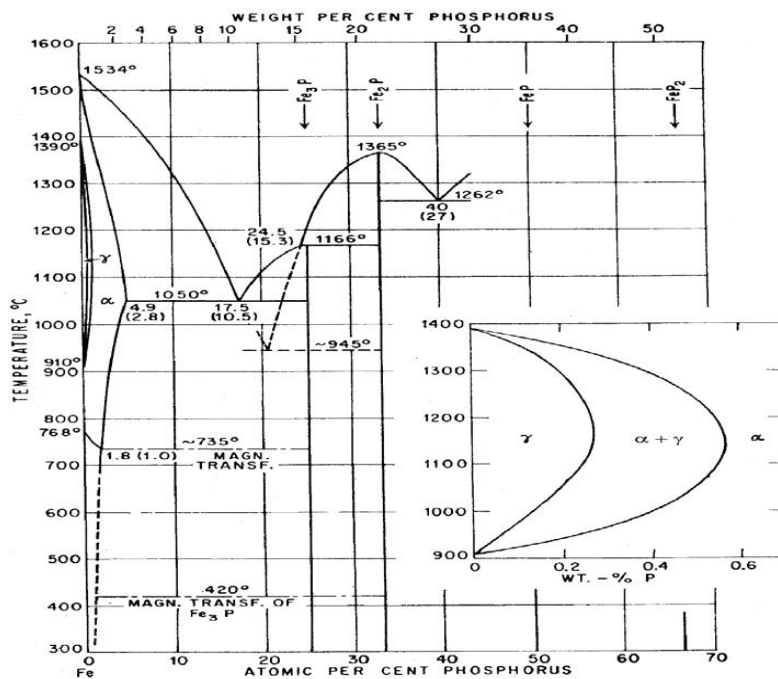


Figure 3: Phase diagram of Fe-P alloy [7]

The hot preforms were immediately transferred to a die fitted in the press and were forged. Then the slabs were hot rolled at temperature of 900°C to form sheets of 0.5mm thickness. Toroids were stamped from the sheet using a die/punch arrangement. Glassy coating was applied on the toroids and these were again annealed at 850°C for three hours to relieve residual stresses. Thereafter the samples were cleaned of residual glassy coating and polished to improve surface finish. A stack of two toroids was used and the dimensions of toroids were 1mm thickness, 50mm outer diameter, 40mm inner diameter with 74 primary and 6 secondary windings using Teflon coated multi strand wire (Fig. 4).

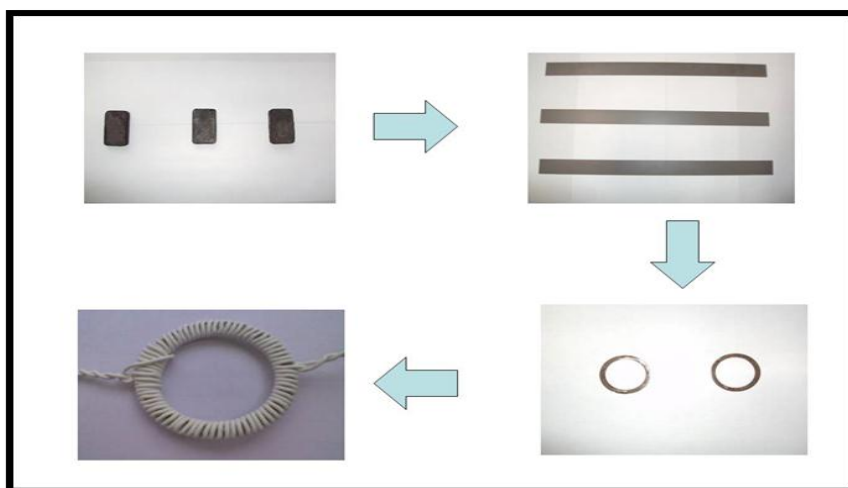


Figure 4: Samples prepared by hot powder perform forging

The samples prepared this way were characterized in terms of their density, microstructure, electrical resistivity and magnetic properties.

RESULTS AND DISCUSSION

Purpose of Graphite Addition

On the basis of literature surveyed it is stated that the presence of carbon is detrimental for soft magnetic performance of devices and the presence of even 0.1% of it is not acceptable. Carbon acts as an interstitial impurity and when its content exceeds the solubility limit, it tends to precipitate out in particle form of carbides at the expense of the host metal. Thus its presence in excessive quantities means a double loss-one represented by the percentage of the impurity itself and the other associated with the host metal lost in the precipitates. Two potential sources of carbon contamination are inadequate lubricant burn-off and improper sintering atmosphere of the finished component. Minimizing it to the lowest possible level will result in improved magnetic performance [8]. In the present investigation carbon is not treated as an impurity. It is purposely added to Fe-P alloys and it helps in a number of ways. Firstly, as a solid lubricant during cold compacting; secondly as solid state reducing agent to take care of oxygen situated at iron powder particles surfaces during high temperature processing. It has been possible to eliminate the use of costly hydrogen (employed as protective atmosphere during sintering of such alloys) by the use of addition of carbon (in the form of graphite). Carbon combines with oxygen and forms protective reducing atmosphere of CO within the compact. The CO gas is not allowed to escape from the compact by the use of tenacious and adherent glass-based ceramic coating [5]. This coating also serves as a protective coating to avoid atmospheric oxygen attack over the compact held at a high temperature. Thirdly it pushes phosphorus into ferrite grains as a solute and thereby discourages it to precipitate as phosphide along the ferrite grain boundaries. Hydrogen atmosphere created during sintering reduces the outer layers of the compact only and the compact remains unreduced from the core. While carbon cleans the entire powder and minimizes the impurities to lowest possible level.

Physical Measurements

Measured values of density of forged and rolled and annealed sheets are given in Table 1 along with the spectroscopic measurement of carbon and oxygen content. These values are comparable with their

wrought counterparts [1]. The electrical resistivity values of the alloy sheets made in the present investigation are recorded and are given in Table 1. It is observed that the resistance of iron increases greatly due to alloying additions like P and Al. The higher the alloying addition, the higher is the alloy's resistivity which corroborates the earlier findings [1, 5]. These alloying elements formed solid solution with ferrite and distorted the ferrite lattice and thereby hindered the passage of electrons which has resulted in increased electrical resistivity of alloys.

Table 1: Density and electrical resistivity of samples after spectroscopic analysis

Material (final chemistry)	Density of compacts (as hot forged) (g/cm ³)	Density of toroids (rolled and annealed) (g/cm ³)	Electrical Resistivity (μΩcm)
Fe-0.29wt%O-0.08wt%C-0.3wt%P-0.3wt%Al	7.62	7.71	27.6
Fe-0.26wt%O-0.07wt%C-0.3wt%P-0.5wt%Al	7.03	7.43	29.7
Fe-0.27wt%O-0.09wt%C-0.5wt%P-0.3wt%Al	7.34	7.53	27.1
Fe-0.28wt%O-0.07wt%C-0.5wt%P-0.5wt%Al	7.47	7.58	28.4

Metallurgical Measurements

Microstructures etched in 2% Nital of rolled and annealed sheets of (a) Fe-0.3wt%P-0.3wt% Al, (b) Fe-0.3wt%P-0.5wt% Al, (c) Fe-0.5wt%P-0.3wt% Al and (d) Fe-0.5wt%P-0.5wt% Al alloy are given in Fig. 5(a)–(d). The microstructures consist of single phase ferrite only.

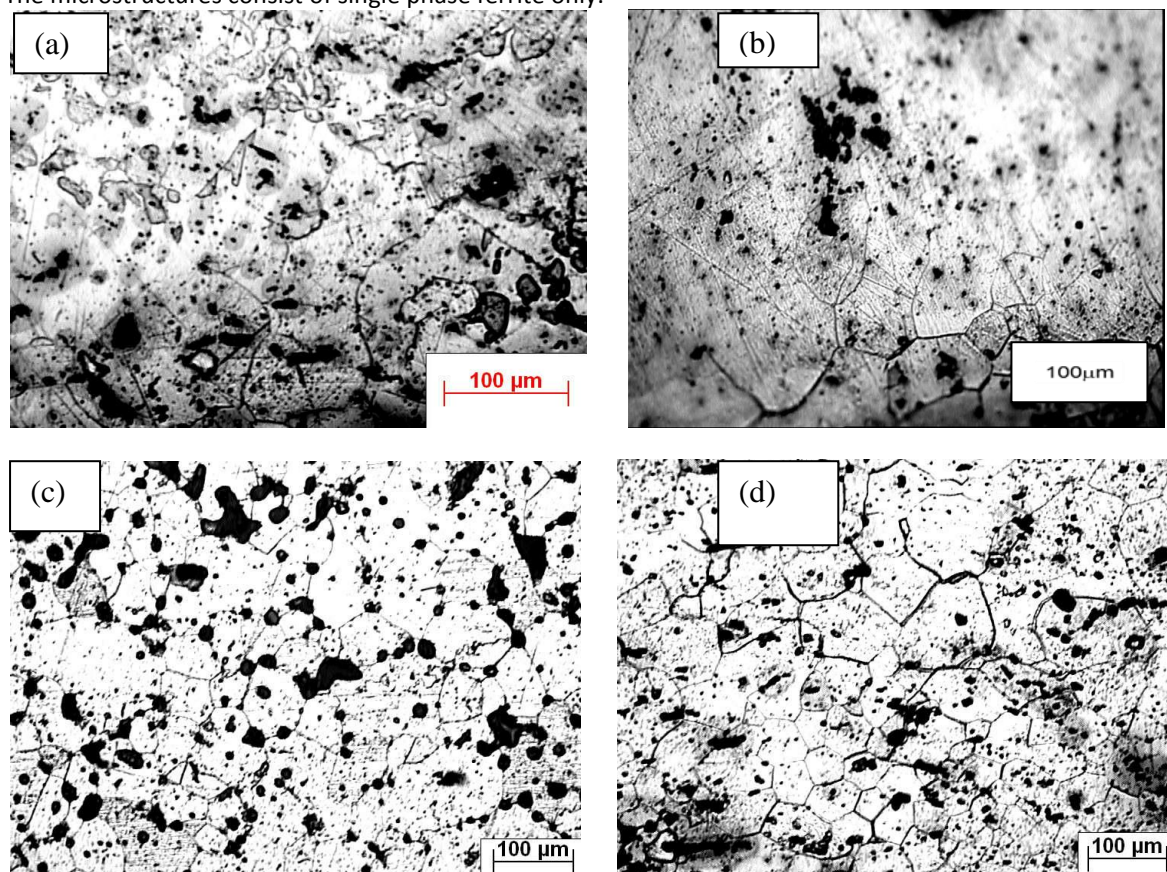


Figure 5: Microstructures of the samples prepared after etching

All the above-mentioned alloys are polycrystalline. X-ray diffraction plot of Fe-0.5wt%P-0.5wt% Al alloy showed (Fig. 6) the presence of BCC alpha-ferrite only, at room temperature (as expected).

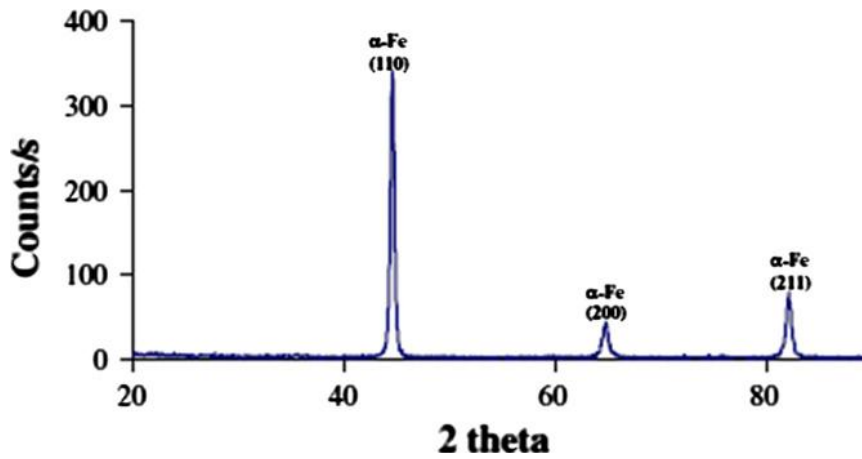


Figure 6: X-ray diffraction plot of Fe-0.5wt%P-0.5wt% Al alloy showing the presence of only α-Fe (BCC) phase.

Lattice parameter for the BCC alpha-ferrite phase had also been calculated and recorded in Table 2. All the alloying elements are BCC ferrite stabilizers and they are added to iron below their solubility limit in iron, therefore it is expected that only alpha phase is present as the product phase.

Table 2: X-ray diffraction data of Fe-0.5wt%P-0.5wt% Al alloy

2θ	Sin ² θ	(hkl)	h ² +k ² +l ²	C = {Sin ² θ / (h ² +k ² +l ²)}	Average C	A = {1.54056 / 2 x √C, Å}
44.51	0.143	110	2	0.07174	0.07180	2.8745
64.76	0.286	200	4	0.07170		
82.16	0.431	211	6	0.07197		

Microstructure test report from optical microscope for the alloys named 5(a)–5(d) shows porosity percentage to be 1.32, 1.29, 1.13 and 1.11 and the average grain size to be approximately 96, 99, 101 and 104 μm, respectively. The microstructures show residual alignment of porosity due to rolling. Pore rounding and coagulation of smaller pores into bigger pores are observed in the microstructures. This may be due to the presence of phosphorous content in the alloys [3]. Large elongated pores as well as small round pores are observed in the microstructures. Pores are elongated due to unidirectional compressive force as schematically shown in Fig. 7.

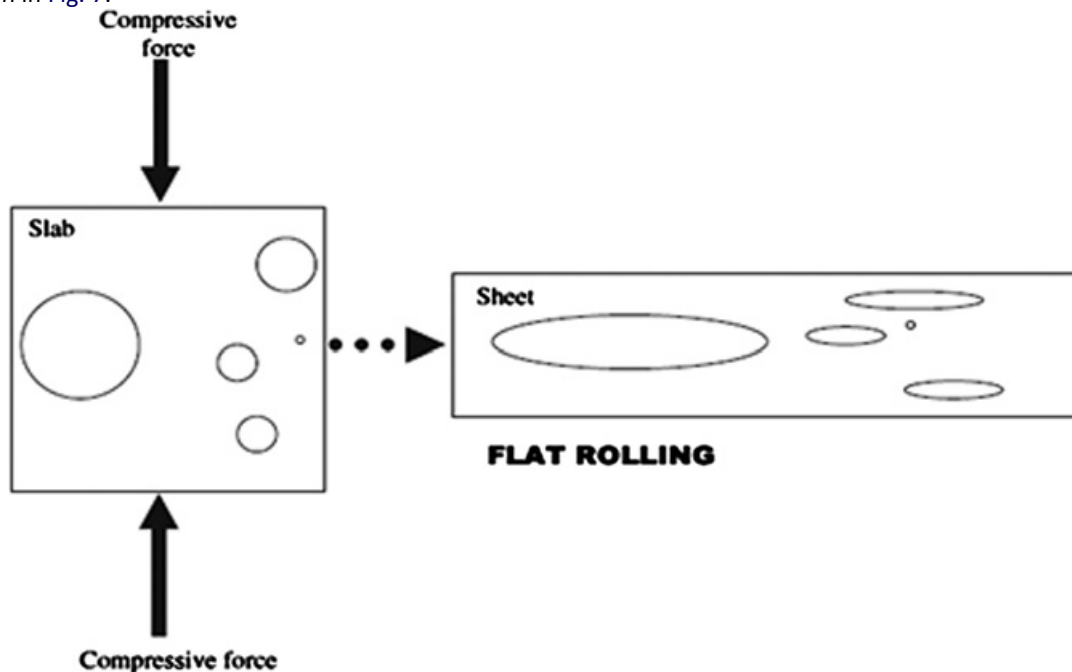
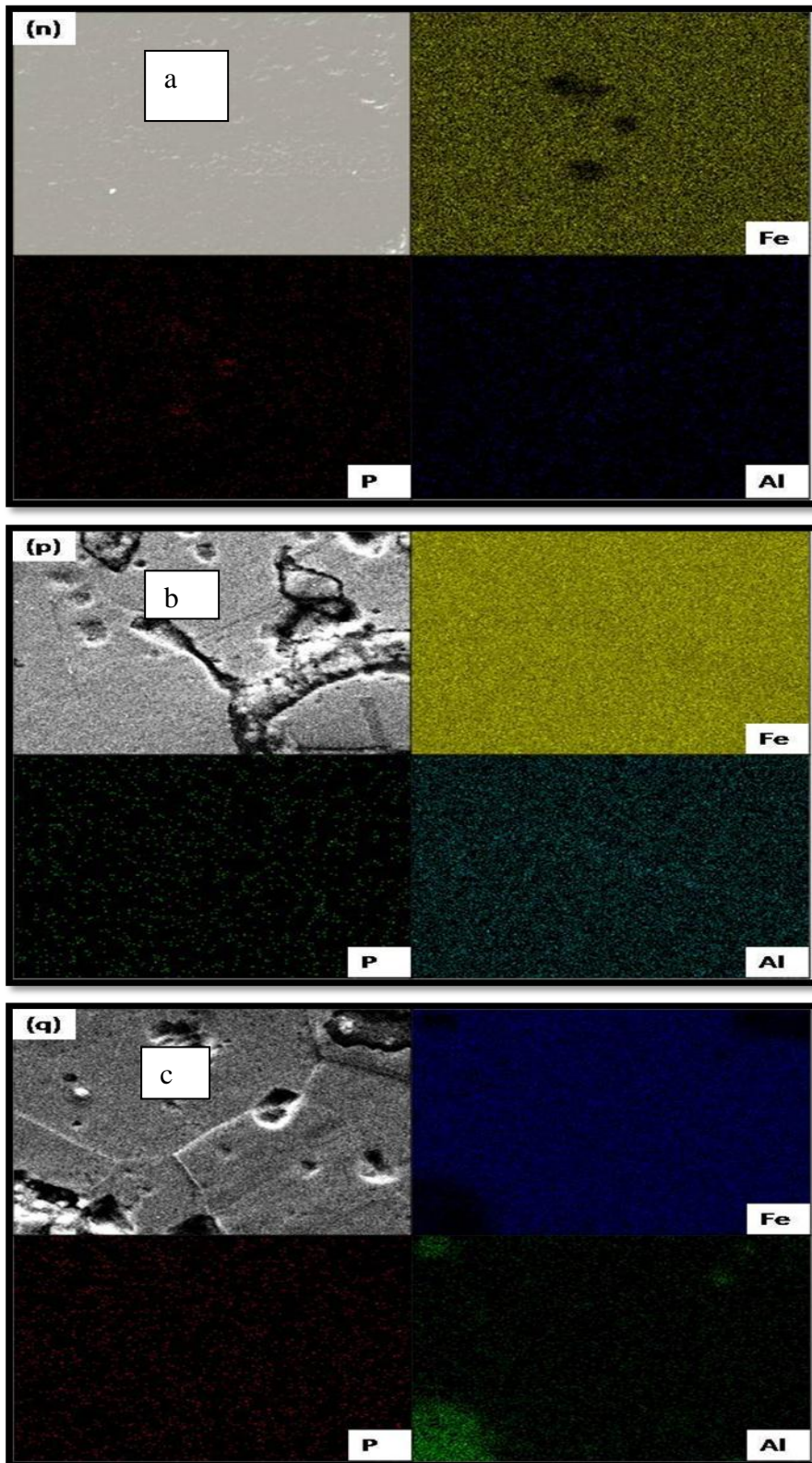


Figure 7: Schematic diagram showing the shape of pores before and after flat rolling

Relatively larger grains are observed in Al containing alloy sheets (Fig. 5(c) and (d)). It may be due to the fact that the alloys were subjected to a longer time of homogenization (at 1250^o C for 3 h). Low porosity may be partly caused by phosphorus which activates the sintering of Fe–Al-based alloy and partly caused by improved processing technique (hot powder preform forging). The alloys developed in the present investigation are free of any segregation of the alloying elements along the grain boundaries. They get distributed uniformly in the entire structure (Fig. 8).



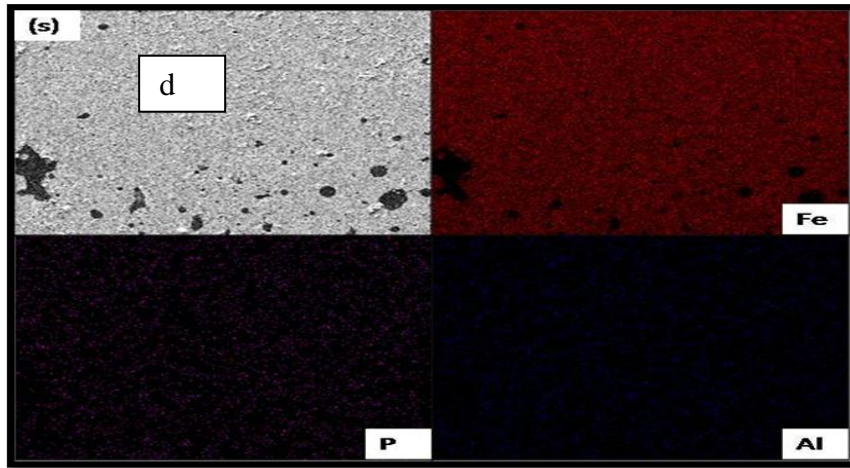


Figure 8: Compositional image (secondary image) and X-ray mapping showing uniform distribution of alloying elements Fe, P and Al in the alloys formed in the present investigation at 3000X.

Magnetic Measurements

The coercivity of the alloys developed in the present investigation is recorded in Table 3. Coercivity of Fe-based alloys falls as we add alloying elements such as P and Al to Fe. The higher the Al and P content, the lower were the coercivity values observed which corroborates the earlier findings [1, 9]. In spite of high percentage of porosity (1.32), Fe-0.3 wt%P-0.3 wt% Al alloy showed low coercivity of 0.53 Oe. It may be due to the fact that the high volume percent porosity is offset by the coagulation of pores (which lowers the coercivity level by reducing specific surface area of pores) and relatively larger grain size (96 μm) caused by the presence of phosphorous [3]. Furthermore, P addition also reduces the coercivity of Fe significantly [9]. Fe-0.5 wt%P-0.5 wt% Al alloy showed lowest coercivity amongst all the alloys developed. This was due to fact that this alloy had a very low level of porosity (1.11), a large grain size (104 μm), and a maximum Al and P alloying addition. In this investigation, it was found that the combined influence of P and Al was much better than the influence of P alone on lowering of coercivity value of iron. In the present investigation, alloying minute quantities of P and Al to Fe gives a high value of saturation magnetization as given in Table 3. It may be due to the fact that P addition activates sintering by uniform distribution of eutectic liquid phase around the grains of iron powder. Hence, a highly densified structure with large grains can be achieved. In addition, as the density of the material exerts the greatest controlling influence on magnetic properties, the loss of saturation magnetization due to porosity can be minimized. Saturation magnetization of iron increases from 1.6 to 1.78 T when phosphorous is added to iron up to 0.8 wt% and decreases thereafter [3,10]. The permeability of the toroidal samples made in the present investigation is recorded in Table 3. It is observed in this investigation that the permeability of iron increases greatly due to alloying additions like P and Al. The higher the alloying addition, the higher is the alloy's permeability which corroborates the earlier findings [1,3,4,11]. The hysteresis loop of Fe-0.28wt%O-0.07wt%C-0.5wt%P-0.5wt%Al alloy is shown in Fig. 9.

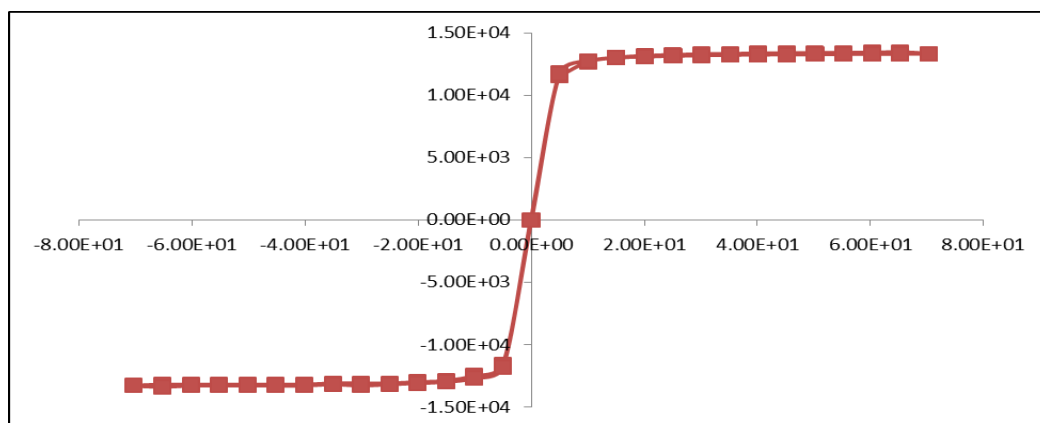


Figure 9: Hysteresis Loop of Fe-0.28wt%O-0.07wt%C-0.5wt%P-0.5wt%Al alloy

The effect of alloying on total magnetic loss for all the samples developed in the present investigation is also given in Table 3. It has been found that alloying reduces the losses to a great extent. Combined addition of P and Al decreases the total magnetic loss of Fe–P–Al alloys significantly [4]. The total magnetic loss is the sum of eddy current loss and hysteresis loss [10]. This may be due to the fact that Al-containing alloys have simultaneously high resistivity and low coercivity values. These contribute toward achieving low magnetic losses. Such a combination of properties is achieved by the use of hot powder preform forging technique employed in the present investigation.

Table 3: Magnetic properties of the alloys developed in present investigation

S. No	Alloy	Coercivity (Oe)	Saturation Magnetization (G)	Permeability	Loss (W/Kg)
1	Fe-0.3wt%P-0.3wt%Al	0.53	13780	11211	0.143
2	Fe-0.3wt%P-0.5wt%Al	0.35	13030	11109	0.105
3	Fe-0.5wt%P-0.3wt% Al	0.46	13900	11780	0.153
4	Fe-0.5wt%P-0.5wt%Al	0.33	13567	12005	0.142

CONCLUSIONS

- Alloys developed in the present investigation show very low coercivity values which range from 0.33 to 0.53 Oe. Alloying additions such as P and Al lower the coercivity of the alloys. The higher the Al and P content (within solubility limit), the lower were the coercivity values observed.
- Permeability and coercivity of the alloys made in the present investigation are comparable to those of commercial Fe–3% Si wrought steel.
- The technology developed in the present investigation showed very low total loss values such as for Fe-0.28wt%O-0.07wt%C-0.5wt%P-0.5wt%Al alloy is 0.142 W/kg.
- Alloying elements such as P and Al increase the resistivity of iron from 27.1 to 29.7 $\mu\Omega\text{cm}$. The higher the alloying addition, the higher is the alloy's resistivity.
- The use of ceramic protective coating eliminates the need of hydrogen protective atmosphere during heating.

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