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Anisotropic Ni–Fe–B films with varying alloy composition for high frequency magnetics on silicon applications

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1. Introduction

In the last two decades soft magnetic materials with high permeability (μ), high resistivity (ρ), low coercivity (H_c) and high saturation flux density (B_{sat}) have been intensively studied for various silicon integrated applications such as inductors [1–3], transformers [4,5], motors [6] and recording heads [7]. For these applications Ni–Fe alloys have been the most accepted material based on high saturation flux density (<1.5 T), high resistivity (20–45 μΩ-cm) and lower coercivity (<1 Oe) depending on alloy composition. However, Ni–Fe films grown with physical vapour deposition (PVD) techniques result preferentially in columnar growth [8,9] for films greater than 180 nm in thickness. This results in an undesirable tenfold increase in coercivity. Electrochemical processing of Ni–Fe alloys and layer-by-layer deposition eliminates the columnar growth issue. Electrochemical deposition of magnetic films is also a faster process for soft magnetic films such as NiFe (81/19) [10,11], NiFe (45/55) [12] and CoNiFe [13–16].Low stress permalloy NiFe film deposition is well established [17–18].

A significant issue for passive magnetic components such as micro-inductors and micro-transformers on silicon is the reduced efficiency due to losses from both the copper windings and the magnetic core [19,20]. The core suffers from hysteresis and eddy current losses. High coercivity materials increase the overall hysteresis loss whereas high conductivity materials make the core susceptible to eddy current losses. For high frequency operation (up to 100 MHz), the devices require core materials with high anisotropy along the hard axis which prevents the core from saturating during high frequency operation. As a result $Ni_{45}Fe_{55}$ is preferred for its high saturation flux density (1.5 T) and resistivity (45 μOhm-cm) and $Ni_{80}Fe_{20}$ for its lower coercivity (<0.5 Oe). This trade-off between magnetic and electrical properties remains a major challenge in electroplated films. A deposition process wherein the Fe composition can be altered readily depending upon application is therefore highly desirable.

Electroless processing has a number of advantages by comparison with electrolytic deposition. Conformal homogeneous deposits with high yield in high aspect ratio structures and in three-dimensional topologies without full or highly conducting seed layers are achievable [21]. This process is ideal for nanowires growth [22–24], deposition in through silicon vias [25] and can result in less porous films [26] than those achieved with electrolytic deposition. A tertiary current distribution characteristic is another issue leading to non-uniformity in electrolytic films [27] that can require complex and costly anode placement. Electroless deposition gives a significant advantage in this regard [28–30]. For DMAB based electroless deposition the nanocrystalline structure of the resulting deposits leads to a higher saturation flux density (B_{sat}) by comparison with hypophosphite based baths [16]. This is due to the lower codeposit content of boron from DMAB (~1%) than

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Table 1
Borane based nickel–iron baths. Concentrations are in mol-dm^{-3}.

Bath contents	Bath-I	Bath-II	Bath-III	Bath-IV	Bath-V
Di-ammonium citrate	0.027	0.027	0.027	0.027	0.027
Lactic acid	0.22	0.22	0.22	0.22	0.22
$NiSO_4{\cdot}6H_2O$	0.060	0.060	0.060	0.060	0.060
$FeSO_4{\cdot}7H_2O$	0.058	0.058	0.058	0.058	0.058
DMAB	0.017	0.034	0.068	0.102	0.136
pH	6.5	6.5	6.5	6.5	6.5
Temp. (°C)	85	75	65	60	58
Ni and Fe composition	Ni: 81%; Fe: 19%	Ni: 81%; Fe: 19%	Ni: 74%; Fe: 26%	Ni: 55%; Fe: 45%	Ni: 37%; Fe: 63%

phosphorus from hypophosphite (~7%) baths which deteriorates the magnetic properties.

In this work DMAB was used to achieve high saturation flux density and low coercivity. The deposition was performed in the presence of an external magnetic field (>300 Oe) to achieve high anisotropy through preferential orientation of the magnetic dipoles during deposition. Secondly, we report that high DMAB concentration leads to preferable Fe deposition at lower deposition temperatures, when other parameters are kept constant. This contributes to an increase in resistivity with excellent magnetic properties. The target of the work was to achieve high quality magnetic films with the following specific characteristics from a CMOS compatible plating solution to assist with integration of the magnetic devices on silicon:

- Low coercivity (<0.5 Oe)
- High resistivity (>25 μΩ-cm)
- High anisotropy field (>10 Oe along hard axis)
- Saturation flux density (>1 T)

2. Experimental procedure

All the solutions were prepared from de-ionized (DI) water of resistivity 18 MΩ-cm. The chemicals used were purchased from Sigma-Aldrich and used as received. The electroless process was performed in glass beakers. The temperature of the bath was maintained with an Ikamag RCT stirring hotplate and IKA H60 temperature probe. The pH was monitored with an OAKTON pH 6 (Acorn series) and adjusted with ammonium hydroxide solution. Silicon (100) wafers were sputtered with 20 nm Ti for adhesion followed by 100 nm Cu in Nordiko DC magnetron sputtering system. Commercially available positive tone photoresist AZ9260 (Microchemicals GmbH) is used to pattern the wafers. Magnetic films were deposited in the presence of a magnetic field (>300 Oe).

The morphology and composition of the deposits were analysed with a Quanta FEG600 scanning electron microscope (SEM) with energy dispersive X-rays (EDX). Intensity correction for trace elements was performed. Film thickness was confirmed with a Tencor (P-10) surface profilometer. The static magnetic properties were analysed with a BH loop tracer (MESA HF200P, SHB instruments). The coercivity (H_c), saturation flux density (B_{sat}) and anisotropy field (H_k) values were obtained from the BH loops of the samples. Complex permeability characterisation at room temperature were performed with a wide-band (1 MHz to 9 GHz) permeameter (PMM 9G, Ryowa Electronics). The electrical resistivities of the samples were measured with a four-point probe system. The currents (I) and voltages (V) across the sample were measured with an Agilent DC multimeter and the resistivity calculated as: $\rho = K(V^*T/I)$, where K is correction factor (~4.53 in this case) and T is thickness of the plated material. Microstructural X-ray diffraction (XRD) analyses of $Ni_{81}Fe_{19}$ (permalloy) and $Ni_{37}Fe_{63}$ films were performed on a PANalytical X'Pert PRO MPD XRD system equipped with Cu K-alpha radiation (1.5405 Å) source and goniometer resolution of 0.001°. Grain sizes and lattice orientation where determined from the XRD results obtained.

3. Results and discussions

3.1. Deposition rate and alloy composition

$Ni_{81}Fe_{19}$ films were deposited electrolessly in the presence of a magnetic field. The pH of the bath was adjusted to 6.5 with diluted ammonium hydroxide, while the deposition temperature was maintained at 75 °C. All the bath constituents were solubilised before adding DMAB. Table 1 lists the bath compositions to achieve different Ni–Fe alloys where bath-II is the composition for uniform permalloy deposition. Thin films with thickness from 330 nm to ~1.58 μm were deposited to estimate the deposition rate. Fig. 1(a) shows the deposition rate of permalloy films deposited under these conditions. The initial plating rate was ~6.6 μm/h decreasing gradually to 3.1 μm/h after 10 min of plating. The Ni and Fe contents were found to be 78–81% and 22–19%, respectively by EDX, with negligible boron content measured (<1%) in all the deposited films. This deposition process is found to be reproducible with consistent results. Fig. 1(b) depicts the SEM cross-section micrograph of uniform film cross-sections with thickness of 770 nm and 1.58 μm. All the films were deposited in a strong magnetic field, >300 Oe provided by NdFeB magnets. The distance between the magnets and the substrate plays a significant role in determining the anisotropy of films. Hence the distance between the two magnets and substrate was maintained at 15 cm. Deposition in a magnetic field has been reported for CoNiFe films [31–33], but no comprehensive study has been done on electroless Ni–Fe alloy deposition in magnetic field.

Further optimisation of the deposition was investigated with increased metal ion content (Ni and Fe ions) while maintaining the Ni and Fe ratio at 1:1 and the other constituents unchanged. Fig. 2 shows that higher Ni and Fe content in the bath increases the deposition rate. However, as observed from the figure, this increase in deposition rate reaches a plateau as the Ni salt concentration increases above 0.12 mol-dm^{-3}. There is no significant change in composition on increasing the metal ion concentration with deposits in the range Ni (74–81) and Fe (26–19) at. wt.%. This increase in deposition rate can be attributed to higher catalytic activity at the Ni for the oxidation reaction of DMAB. This is consistent with electrolessly deposited Ni–Co alloys reported in literature [14,16,34]. For the remainder of the processing a Ni content of 0.06 mol-dm^{-3} was utilised.

Electroless deposition of Ni–Fe alloys [35–37] from hypophosphite baths has also been reported previously. These baths can provide up to 25% Fe composition [34]. Wang [36] has reported deposition of Ni–Fe–P amorphous films from boric acid and sodium citrate baths to achieve 20% Fe and 10% Ni. The film quality and grain size strongly depends on the complexing additives and reducing agent. However, in this work, diammonium citrate and lactic acid were employed as complexing agents and DMAB was used as reducing agent as similar bath compositions by the authors have shown excellent nanocrystalline CoNiFe-B thin films [16]. For