Defect and Microstructural Characterisation of a-Si:H deposited by low temperature HW-CVD on paper substrates

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Abstract

a-Si:H has been deposited on 80 g m^{-2} wood-free paper, with and without an intermediate metallic interlayer, using low temperature hot wire chemical vapor deposition HW-CVD. In this paper we compare the differences in microstructural properties of the two types of layer, concentrating on the influence of the substrates, including their effect on the deposition rate of the material and substrate temperature. Techniques employed in the characterisation include, X-ray diffraction to study composition, and crystallinity, positron annihilation for open-volume defects, and scanning-electron and optical microscopy.

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Introduction

Amorphous hydrogenated silicon (a-Si:H) has a broad field of application, e.g. TFT's mainly as pixel switches [1] and solar cells [2]. Its production, based on CVD processes, enables large area applications under the use of a wide variety of substrates. For flexible devices the most common substrate is steel foil, with the advantage of deposition and processing temperatures as high as 1000 ^{o}C [1]. Other flexible substrates discussed so far are plastic foils, where the device production temperatures must be lower than the working temperature of the specific polymer. For example a-Si:H has been successfully deposited on polyethylene tetraphthalate (PET) by PE-CVD at 110 ^{o}C [3] and by HW-CVD at 100 ^{o}C [4]. Besides the restriction in deposition temperature, to use flexible substrate materials such as thin plastics, the control of mechanical stress due to the differences in thermal expansion coefficients of the components and the deposition process itself [5] must be taken into account [6]. Other factors are chemical stability, adhesion of the layer, and surface roughness [1].

In contrast to flexible plastic substrates, we present the results of depositing a-Si:H on ordinary paper, using HW-CVD at low temperature. Differences in microstructural properties of two types of layer, with and without an intermediate metallic interlayer, are shown. Techniques employed in the characterisation include, X-ray diffraction to study composition, and crystallinity, positron annihilation for open-volume defects, and scanning-electron and optical microscopy. Emphasis is given on investigating the influence of the substrate on the microstructure, taking into account local changes in the deposition rate and substrate temperature.

Sample Preparation

As substrate, simple 80 g m^{-2} wood-free paper, without any subsequent preparation was used for layer deposition. To study both, a-Si:H on paper as well as a-Si:H with an intermediate metallization layer, under the same deposition conditions, we used a 10×10 cm chess board pattern with 4×4 squares, where half of the squares were covered with the metallization, and the other half were left free. The whole structure was coated with an a-Si:H layer by HW-CVD using pure silane gas at a pressure of 80 μbar and a flow rate of 60 sccm. The nominal substrate temperature was 100 ^{o}C and the temperature of the Ta filament was 1600 ^{o}C. The deposition time was 15 minutes.

Only two of the central squares were used for further investigation because of the inhomogeneous coating thickness in the outer region of the sheet, which could be seen by a series of beautiful concentric interference fringes. The actual substrate temperature was estimated by comparison of the color change of the same sort of paper due to heat treatment for the same period of time. For sample B, paper with metallization and an a-Si:H coating, the substrate temperature of 200 – 220 ^{o}C was slightly higher than the 150 – 200 ^{o}C for sample A, the sample without the metallization.

Experimental details

The morphology of the deposited layers was investigated by scanning electron microscopy, using secondary electron imaging.

X-ray diffraction experiments were performed at Huber Diffraktionstechnik GmbH & Co. KG using a Guinier system, equipped with an imaging plate Guinier Camera 670. The measurements were carried out in transmission geometry, with the deposited layer facing the detector, using monochromatic Cu $K\alpha_1$ radiation. Each spectrum was recorded for 10 minutes, with the imaging plate being read 10 times. The camera recorded a 2Θ range of zero to 100 ^{o}, in intervals of 0.005^o.

Positron lifetime spectroscopy was performed on the deposited material using the pulsed positron beam at the Universität der Bundeswehr München [7]. In this system a combination of rf pulsing elements superimposes a time structure onto a moderated positron beam, allowing timing spectroscopy to be performed, with a resolution of 250 ps, at various depths in the sample (determined by the incident positron energy). Lifetime spectra were recorded over a complete range of beam energies, up to 18 keV, and analysed both in terms of the mean positron lifetime and decomposition into different components corresponding to the different states from which the positrons ultimately annihilate.

Results and Discussion

The X-ray diffraction patterns of both samples are similar (Fig. 1), with the exception of the additional peaks due to the metallic interlayer. Both layers show many crystalline peaks, which have been identified as calcite, which is added to the paper in the form of crushed limestone as a filler. In the transmission geometry, the intensity of these peaks is reduced uniformly in the metallized sample because of absorption of X-rays in the metallic interlayer. In neither sample is there evidence of any crystalline silicon contribution, but there are weak broad amorphous peaks underlying the diffraction patterns. This is most easily seen in the pattern for the layer deposited without prior metallization as a convex background under the first calcite peak and in the range 40 – 55^{o}.

Figure 2 shows the morphology of the a-Si:H deposited on the paper substrate, with and without metallization. As can be seen in the figure, the two layers appear vastly different, with the layer deposited over the metal forming a fish-scale like structure (fig 2a). In contrast, the layer deposited on paper appears to be completely absent, and particles of the limestone filler are clearly visible (fig 2b). Nevertheless, under optical imaging the deposited silicon is clearly visible, and the individual fibres of the paper have been uniformly coated, with only little filling in between the fibres.

Despite the radical difference in morphology, the defect structure, as seen by positron annihilation is very similar in both layers. The main differences in the mean lifetime (Fig 3a) of the samples occur for deeper positron implantation and can be attributed to differences in the substrate – open-pored cellulose fibres as opposed to a metallic layer. A component analysis yields a single component in the layers, as illustrated by the intensities shown in (fig 3b), with the same value of 390 ± 1 ps. This is longer than that observed for high temperature growth on glass substrates [8,9], indicating a larger free-volume at the dangling-bond complexes, but there is no long-lived component indicating the presence of voids.

From the energy at which 50% of the positrons reach the substrate, it is possible to estimate the thickness of the silicon layer. Assuming a typical density of 2.3 g cm^{-3}, this corresponds to layer thicknesses of 650 and 550 nm on the paper and the metallization respectively. This suggests a slightly lower growth rate on the metal, in agreement with the observed difference in X-ray diffraction intensities for the a-Si:H, which could possibly be explained by the difference in substrate temperature.

Conclusions

We have shown that a-Si:H of reasonable quality, without large structural defects, can be grown on normal paper substrates by HW-CVD. During the deposition process, the metallized substrates reach a higher temperature than plain paper. Both X-diffraction and positron beam profiling indicate that the growth rate on the uncoated substrate is slightly higher than with prior metallization. There is no evidence of a crystalline phase or voids in the a-Si:H layers. Although both layers have a radically different microstructure and morphology, the internal defect structure is similar, with a dominant dangling-bond complex of similar size.

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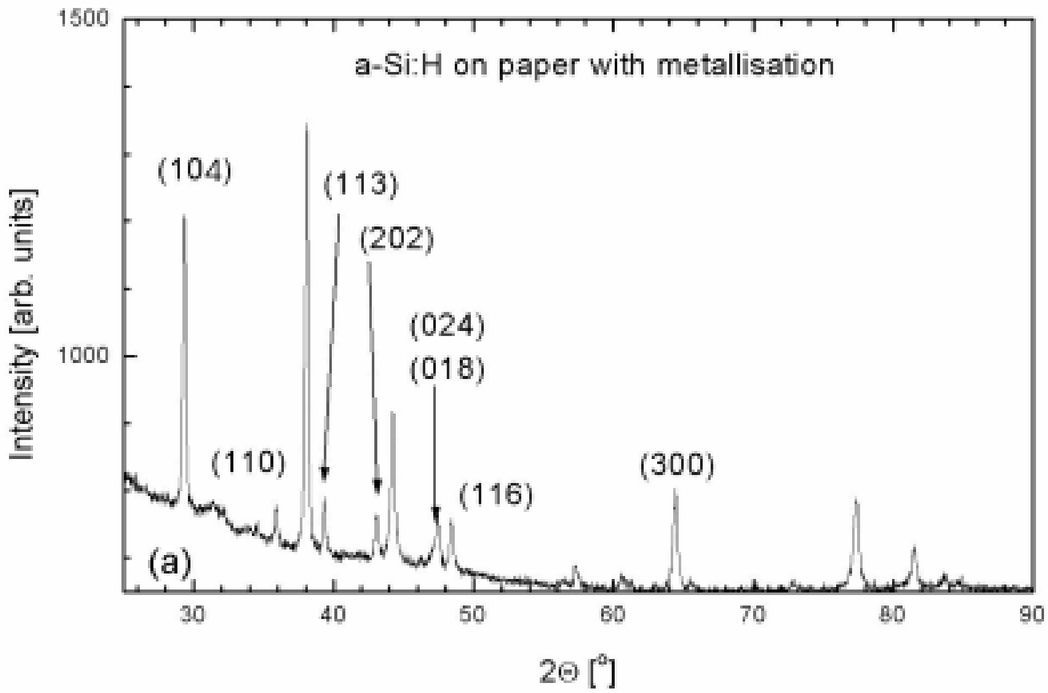
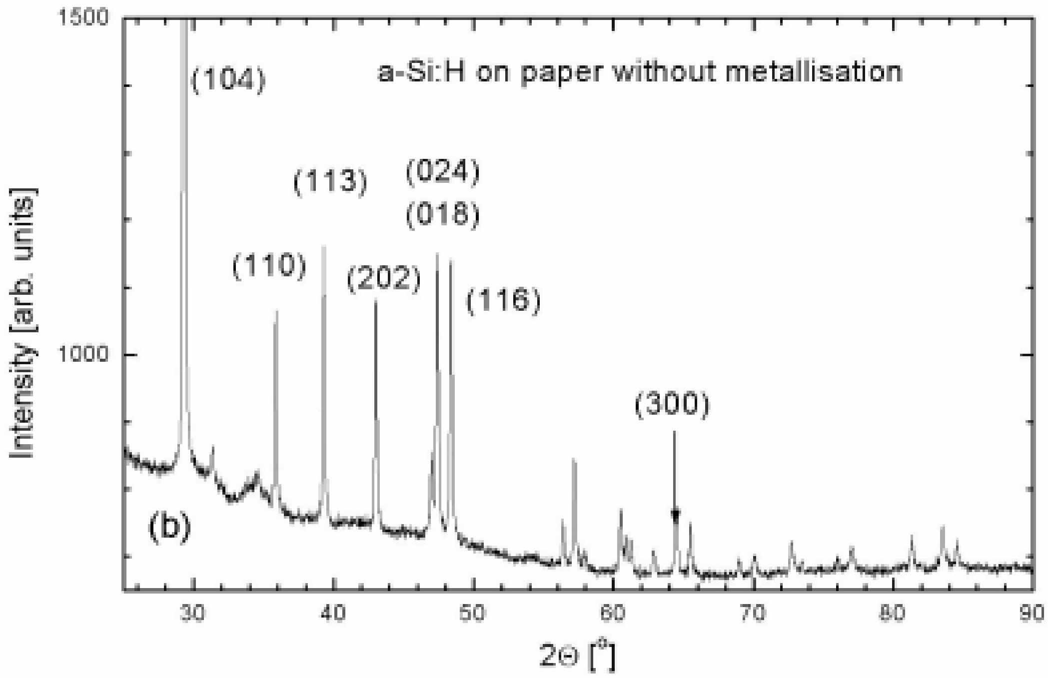
Figure Captions

Fig1.: X-ray diffraction patterns for a-Si:H deposited by HW-CVD on paper substrates: (a) with metallic interlayer, and (b) without prior metallization. The diffraction peaks for the limestone filler in the paper are indicated.

Fig.2.: Scanning electron micrographs of the surface a-Si:H deposited by HW-CVD on paper substrates: (a) with metallic interlayer, and (b) without prior metallization.

Fig. 3: Positron lifetime characteristics as a function of incident positron energy for a-Si:H deposited on paper with and without prior metallization: (a) mean positron lifetime, and (b) intensity of the main component.


a-Si:H on paper with metallisation
(104)
(113)
(202)
(024)
(018)
(110)
(116)
(300)
(a)
Intensity [arb. units]
1500
1000
30
40
50
60
70
80
90
2Θ [°]


a-Si:H on paper without metallisation
(104)
(113)
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(018)
(110)
(202)
(116)
(300)
(b)
Intensity [arb. units]
1500
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40
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2Θ [°]

Figure 1

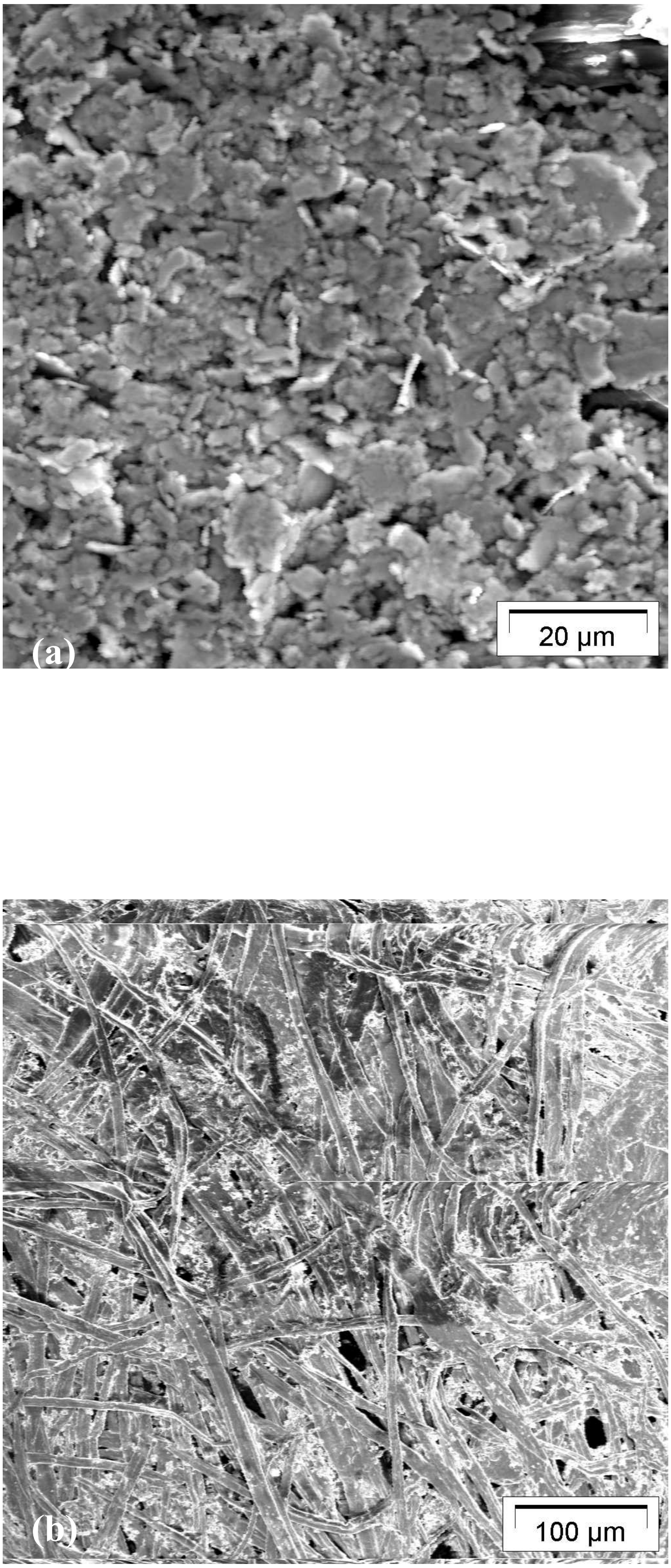

Fig 2

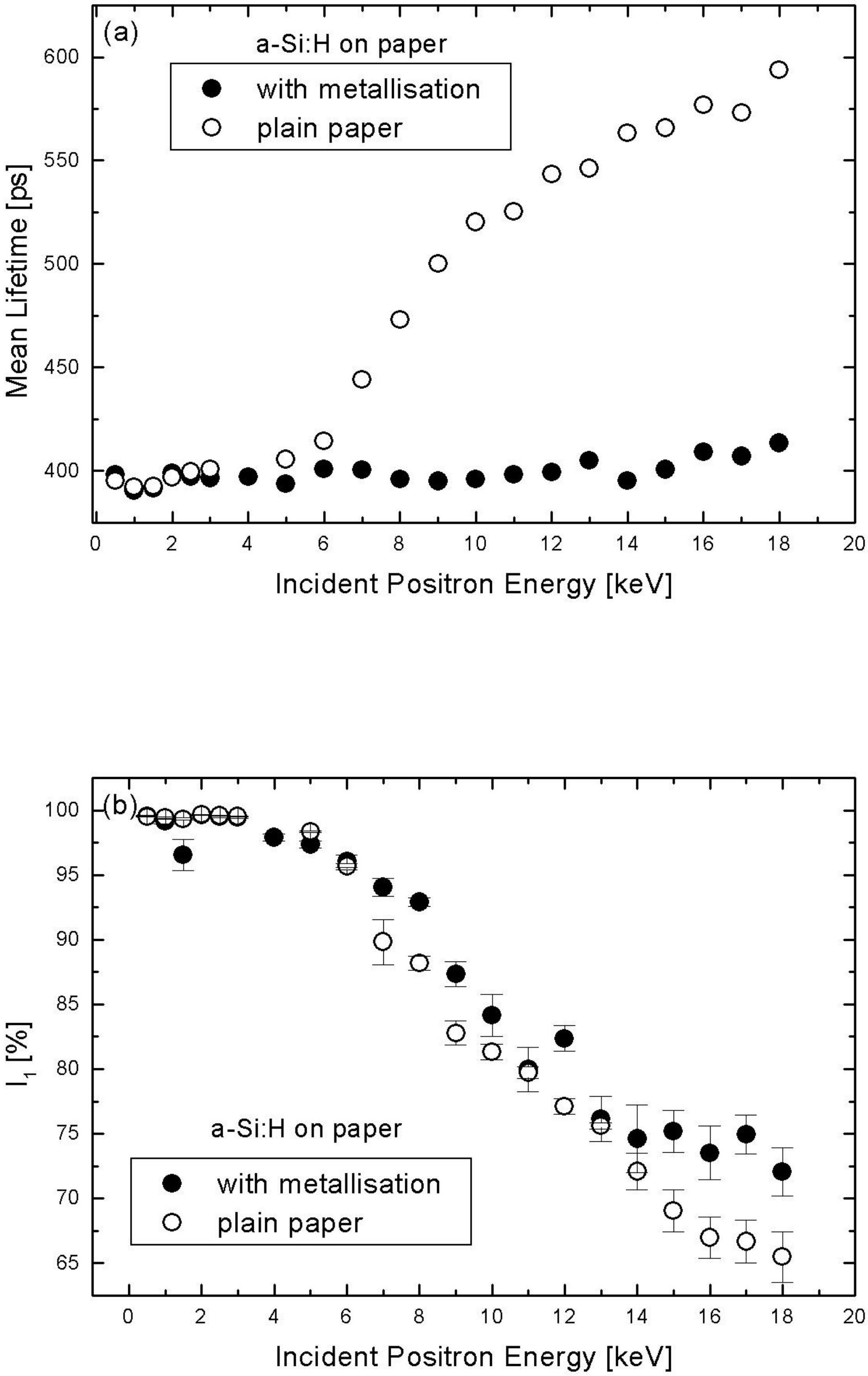
(a)
a-Si:H on paper
with metallisation
plain paper
Mean Lifetime [ps]
Incident Positron Energy [keV]
(b)
a-Si:H on paper
with metallisation
plain paper
I_1 [%]
Incident Positron Energy [keV]

Figure 3