

A study of Pd-Ta on Si(100) using AES, RBS and variable energy positron annihilation.

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ABSTRACT

The applicability of $\text{Pd}_x\text{Ta}_{1-x}$ as a diffusion barrier on Si has been investigated. For this purpose $\text{Pd}_x\text{Ta}_{1-x}$ films of 200 nm thickness (x ranges from 0 to 1) were deposited on Si(100), and the reaction between over-layer and substrate was studied as a function of temperature. Interaction was found to occur at temperatures increasing with the Ta content. The as-deposited $\text{Pd}_x\text{Ta}_{1-x}$ films with $0.2 \leq x \leq 0.6$ were found to be amorphous. The amorphous phase had a higher reaction temperature than the crystalline one, causing a discontinuous step in the reaction temperature. RBS spectra revealed that for the Pd-rich compositions first a stoichiometric Pd_2Si layer formed underneath a pure Ta layer. At higher temperatures TaSi_2 formed at the surface. For Ta-rich compositions Pd_2Si formed first as well, however, the reaction temperature was so high that Pd_2Si grains formed in a Si matrix. The defect density of the Ta layer, which remained after outdiffusion of Pd, was investigated using variable energy positron annihilation. The defect concentration is very high, as deduced from the trapped positron fraction. A model is presented that describes the composition dependence of the reaction temperature.

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MASTER

I. INTRODUCTION

The potential application of binary amorphous alloys as diffusion barriers in micro-electronic devices has been recognized by various groups after the suggestion by Wiley et al. (1). Nicolet and co-workers demonstrated that 200 nm thick amorphous films, consisting of an early and a late transition metal, effectively suppress the reaction between Al and Si up to a temperature of 500 °C, (see e.g. 2,3,4). It is believed that the absence of grain boundaries hampers the transport of Si through the film (5). A good barrier action can also be obtained in poly-crystalline films, provided that the grain boundaries are blocked. This can be achieved by a combination of compressive stress and filling of the grain boundaries with small atoms like N. For systems with a large tensile stress, like most e-gun deposited binary alloys, amorphous phases will certainly display a better barrier function than crystalline ones, since the grain boundaries in the latter will allow diffusion.

Good diffusion barriers have no easy diffusion paths, so transport of Si (or Al) takes place predominantly through the bulk of the barrier. This would result in silicide (or aluminide) formation, since most silicides (and to a lesser extent Al-alloys) have a large negative heat of formation. Consequently, the failure mechanism of good amorphous diffusion barriers is the reaction with Si (or Al). This reaction may also be applied beneficially. In early work by Tu, Mayer and others (6,7,8) it was demonstrated that the reaction of solid solutions of Pd-V, Pd-W and Pt-Cr with Si proceeds as follows: first, the near noble metal reacts with Si, forming a silicide on top of the substrate but underneath the remaining film. Upon further annealing the top-layer is silicidized as well. Hung et al. (9) demonstrated that the reaction of binary amorphous alloys composed of an early transition metal and a near noble metal with Si generally proceeds in the same way as that of the solid solutions mentioned above. The reaction is thought to be useful to form shallow silicides that are free of pinholes and pores (10).

In this study this double interest is reflected. The onset of the reaction of Pd with Si was studied for various compositions using Rutherford Backscattering Spectrometry (RBS) after in-situ annealing, and using X-ray Diffraction (XRD). Furthermore, to investigate the potential application of PdTa films as a diffusion barrier, we monitored the appearance of Si at the surface of Pd_xTa_{1-x} films (with x ranging from 0 to 1) using Auger Electron Spectroscopy (AES) after in-situ annealing. For one composition variable energy positron annihilation was employed to observe the silicide formation, and to obtain information on the defect density in the top-layer. Transmission electron microscopy (TEM) was applied to study the microstructure of the films.

II. EXPERIMENT

The $\text{Pd}_x\text{Ta}_{1-x}$ films were deposited by co-evaporation on Si(100) substrates using e-beam evaporators. The TEM specimens were deposited on NaCl crystals. The deposition rate was controlled with quartz crystal monitors. Films were grown of different composition, with x varying in steps of 0.1; the indicated compositions are as derived from the quartz crystal monitors, the actual composition may be off with at maximum 0.02 as confirmed by RBS results. The base pressure in the deposition system was 10^{-6} Pa. During deposition the pressure was kept below 2×10^{-5} Pa; the main contribution to this pressure was from hydrogen. The film thickness was approximately 200 nm, except for the TEM specimens, which were 30 nm thick, and for the specimen used for the variable energy positron annihilation experiment, which was 73 nm thick. The Si(100) substrates were dipped in a 1% HF solution to remove the oxide layer and rinsed in de-ionized water. The substrates were checked to be hydrophobic, subsequently placed in the deposition system which was immediately pumped. The TEM specimens were prepared by deposition on NaCl crystals, which were dissolved in de-ionized water. Subsequently the thin films were placed on Pt grids, suitable for use in the TEM.

The specimens were mounted in either an AES or an RBS analysis chamber. For the AES measurements the specimens were sputter-cleaned and analyzed after in-situ annealing. The annealing was performed using electron bombardment of a carrier plate, the temperature was determined with a thermocouple mounted onto the specimen-clamps.

The 2 MeV RBS analysis was also carried out after in-situ annealing. In this case the annealing was performed by passing a current through the Si substrate. The temperature was monitored using a pyrometer. For both analysis methods the temperature was stepwise increased with 50 °C; the annealing time was always 15 min, the pressure during annealing was below 10^{-6} Pa, and the analysis was performed with the substrate at room temperature.

The apparatus used for the variable energy positron annihilation is described in detail in refs. 11,12. Essentially a specimen is subjected to bombardment with positrons with a well defined incident energy E_i , ranging from 200 eV up to several tens of keVs. The implantation profile is analogous to that of electrons (13). Those positrons that are not re-emitted from or reflected at the specimen surface annihilate with electrons in the specimen after being thermalized. The width of the annihilation gamma peak at 511 keV is a measure for the momentum distribution of the electrons involved in the annihilation process, and is therefore sensitive to the composition of a specific layer. The shape of the annihilation gamma peak is

characterized by a single line-shape parameter S , for a definition of S see ref. (12). The dependence of S on the incident positron energy E_i reflects penetration and subsequent annihilation in a specific layer. The straggling during implantation as well as positron diffusion after moderation affect the depth resolution: interfaces are not sharply reproduced in the S versus E_i spectra.

A feature which is rather unique to the technique is its sensitivity to defects. Positrons that have diffused back to the surface can pick up an electron while being emitted into the vacuum, thereby forming Positronium (Ps). The fraction of formed Ps can be measured, since the decay of Positronium in the triplet state does not contribute to the 511 keV annihilation peak. By measuring the relative amount of Positronium formed as a function of the primary positron energy E_i , the fraction of positrons which have diffused back to the surface is determined. From the latter fraction an effective diffusion length can be obtained, which in turn is directly correlated to open volume defect density. For more details about the method see refs. 12,13,14.

A 40 mCi ^{22}Na positron source was used, moderation of the positrons was accomplished by placing a W(100) crystal with a thickness of 200 nm in front of the source. Acceleration was done by an electrostatic field. The beam diameter was about 8 mm, a Ge detector was placed immediately behind the specimen, which was mounted on a Ta foil. Annealing of the specimen was performed by passing a current through the Ta foil; the specimen-temperature was estimated from the temperature indication of a thermocouple, which was clamped between the Ta-foil and the specimen. The pressure during annealing remained below 10^{-5} Pa.

III. RESULTS

A. TEM observations

It was found that as-deposited films of $\text{Pd}_x\text{Ta}_{1-x}$ with $0.2 \leq x \leq 0.6$ were amorphous. The crystalline alloys at the Pd-rich side were single phase fcc solid solutions; at the Ta-rich side the β -Ta phase was found. In all crystalline films small cracks were observed with TEM, whereas the amorphous films appeared to be continuous. The as-deposited amorphous films were stepwise annealed (stepsize 50 °C) in the hot stage of the TEM; the temperature was kept constant for 15 minutes with the electron beam switched off. After cooling down of the specimen a bright field image, a dark field image and the electron diffraction pattern were obtained. In fig. 1 the crystallization temperatures thus established are shown. The indicated crystallization

temperature is taken 25 °C below the temperature of the annealing step at which the first crystallites are observed. The crystallization process itself turned out to be complex, with a primary crystallization step leading to small fcc crystallites embedded in an amorphous matrix, followed by a secondary crystallization step leading to a β -Ta phase. For more details see ref. (15).

B. Interface reaction between Si and $\text{Pd}_x\text{Ta}_{1-x}$

We studied the reaction of the metal overlayer with RBS for the following compositions: $\text{Pd}_{0.9}\text{Ta}_{0.1}$, $\text{Pd}_{0.7}\text{Ta}_{0.3}$, $\text{Pd}_{0.5}\text{Ta}_{0.5}$ and $\text{Pd}_{0.3}\text{Ta}_{0.7}$. Films with the first two compositions are crystalline while the latter two are amorphous.

In fig. 2 we show some RBS results for the most Pd-rich composition. Quite evidently the reaction starts at the Si/PdTa interface, leading to a layer containing Pd and Si. At 300 °C the reaction proceeds already at a noticeable rate. In the XRD spectra peaks were observed after annealing at 300 °C, indicating the presence of substantial amounts of Pd_2Si . RBS indicates that a concentration gradient of Ta is present after annealing at 350 °C: the deeper layers are slightly enriched in Ta due to the depletion of Pd starting at the Si/PdTa interface. After annealing at 400 °C the Pd_2Si layer has increased in thickness, accompanied by a shift of the Ta-rich layer to the surface and by a considerable take-up of Si in this layer, suggesting the formation of TaSi_2 . According to XRD the fcc Pd(Ta) phase is no longer present. After annealing at 450 °C the formation of TaSi_2 is observed with XRD. A simulation of the RBS spectra (16) indicated that all Ta had reacted with Si after annealing at 600 °C forming stoichiometric TaSi_2 .

For the other compositions the reaction proceeds similarly, the reaction temperatures differ slightly. A RBS spectrum for an amorphous composition is given in fig. 3. Table 1 gives the temperatures at which the diffusion of Pd out of the Ta layer starts as observed by means of RBS, and the temperatures at which the two silicides were observed with XRD. For the two amorphous films amorphous characteristics persisted in the diffraction patterns up to an annealing temperature of 600 °C. Furthermore, for these films the outer layer still contained Pd, according to RBS. To investigate this further, a specimen with composition $\text{Pd}_{0.4}\text{Ta}_{0.6}$ was annealed at 750 °C and analyzed with Scanning Auger Microscopy (SAM). The film surface was found to be inhomogeneous in composition, showing islands with a size of a few 100 nm, composed of Pd and Si, whereas the regions in between the islands displayed Ta and Si. Apparently, Pd_2Si islands have formed on a TaSi_2 surface, explaining the RBS results.

For a film with a thickness of 73 nm and a composition of $\text{Pd}_{0.51}\text{Ta}_{0.49}$ (both determined with RBS) the reaction with Si was followed utilizing the variable energy positron annihilation technique. In fig. 4 the dependence is shown of the line-shape parameter S on the initial incident energy E_i of the positrons. Since the average penetration depth of the positrons is proportional to $1/\rho \cdot E_i^{1.6}$; with ρ the density, the depth scale is more expanded towards the surface (13). A tentative depth scale, which only holds for the unreacted metal overlayer, is given at the upper horizontal axis. The initial steep decrease of the curve indicates the influence of the surface. The minimum in the curve represents the value of S in the PdTa layer, whereas the value of S of 0.48 at incident positron energies higher than 14 keV approaches the value for pure Si in this specific set-up. The transition region between 4 and 12 keV reflects the fact that at a given energy the positrons partly annihilate in the PdTa film and partly in the Si layer. This ambiguity is due to the spread in penetration depth and possibly to diffusion after thermalization. After annealing the surface has changed and the S-value has increased. For this specific experiment insufficient information was available about the surface to base firm conclusions on this surface-related S-value. Part of the increase may be caused by the larger fraction of Positronium as discussed in the next section. Another observation is that the value of S for incident positron energies between 4 and 8 keV increases drastically. This is even more clear from the plot of the ΔS value, being the difference between the S-value after annealing at 600 °C and the S-value of the as-deposited film (fig. 4b). The interpretation is straightforward: from earlier experiments it is known that the S-value of Pd_2Si is in between that of pure Pd and of pure Si. For this specific apparatus the value is approximately 0.45 (17); for TaSi_2 a similar S value was measured. Consequently, the upward shift of S indicates the take-up of Si by the overlayer. Another feature is that if the take-up would have been homogeneous throughout the layer up to the surface, the ΔS value would have shown a more gradual dependence on E_i . Since the dependence is rather steep at an energy of 4 keV, the Si take-up must be limited to a certain layer thickness. After annealing at 700 °C the S-value at lower energies has increased as well, indicating Si-take up throughout the Pd-Ta film.

C. Defect density in the outer layers.

The Positronium fraction as a function of the incident energy was determined for the same specimen used for the S/E measurements described in the previous section. The decrease of the Ps fraction as a function of incident positron energy is a direct indication for the positron diffusion length in the outer layers. The diffusion length for the as-deposited PdTa film was estimated to be of the order of 1 nm. The accuracy is

limited since the total fraction of Positronium was of the order of 0.02, so that there is a large contribution from the background signal. This short diffusion length is typical for amorphous materials; generally, defect free metal single-crystals yield values of the order of 100 nm, and crystalline alloys with many defects yield values in between the value for amorphous phases and that for a single crystal (18). It should be realized here, that the information obtained about the defect density is only coming from a surface layer with a thickness of a few times the diffusion length, so that in this specific case the defect density is probed in approximately the outer 4 nm. Up to an annealing temperature of 500 °C hardly anything changes, between 600 and 700 °C the total fraction of Ps is observed to increase strongly. During earlier experiments concerning the reaction of a Pd overlayer with a Si(100) substrate, performed under exactly the same conditions, the increase of the Positronium fraction could be correlated with the arrival of Si at the surface (17). The presence of Si at the surface after annealing at 600 °C was confirmed by RBS. The positron diffusion length, however, hardly has changed after annealing at 600 °C. Although Pd has diffused out partly and Si has diffused in the overlayer, the defect density remains very high, indicating an amorphous or microcrystalline structure.

D. Si arrival at the surface of $\text{Pd}_x\text{Ta}_{1-x}$

The temperature at which Si was observed with AES (meaning a few at. %) at the surface of $\text{Pd}_x\text{Ta}_{1-x}$ (called the failure temperature) is given in fig. 5. The failure temperature increases with the Ta content up to 625 °C for $\text{Pd}_{0.5}\text{Ta}_{0.5}$ and remains further constant. To investigate the failure mechanism (reaction of Si with the overlayer or diffusion of Si through the overlayer), the very same specimens used for AES were analyzed by RBS after the detection of Si at the surface. Only for the Ta-rich crystalline compositions Si was found at the surface and not in the metal film, indicating the latter failure mechanism. For these compositions several specimens were tested, some of which displayed a higher failure temperature, in which case RBS showed the presence of Si throughout the overlayer, indicating failure due to reaction. The latter measurements suggest that crystalline alloys not necessarily have to lead to relatively low failure temperatures, and the higher temperature is therefore taken as characteristic for these compositions. The relatively low failure temperature of the specimen with composition $\text{Pd}_{0.7}\text{Ta}_{0.3}$ was reproducible and RBS showed in that case Si throughout the overlayer after annealing at 450 °C.

4. DISCUSSION

(i) Crystalline Pd(Ta) solid solutions. Generally, the reaction proceeds as follows: Pd diffuses out of the Pd-Ta layer forming Pd₂Si at the Si/PdTa interface. This is in agreement with earlier measurements by Tu et al. on the system Pd-V (6). The temperature at which Pd starts to diffuse out depends on the composition. The RBS spectra also reveal, that the remaining metallic film is enriched in Ta at the Pd₂Si/PdTa interface. Therefore at the Si/PdTa interface Pd must be consumed to form silicide. Consequently, Pd depletion takes place close to the interface, and Pd has to be supplied from the outer PdTa layer. This requires diffusion of Pd through the PdTa layer. The RBS spectra indicate that this is not accompanied by Si diffusion into the PdTa layer. At higher annealing temperatures the remaining layer, which finally is composed of nearly pure Ta, reacts as well. Both AES and RBS indicate a reaction temperature of 400 to 500 °C for this Ta layer. XRD indicated the formation of TaSi₂. This reaction temperature is well below the temperature at which silicide formation is observed for pure Ta; the out-diffusion of Pd must have left behind a Ta layer with a high defect density in order to explain this low reaction temperature.

(ii) amorphous PdTa alloys. The reaction of amorphous PdTa alloys proceeds similarly to that of Pd(Ta) solid solutions; Pd diffuses out to form Pd₂Si. The temperature at which this occurs, is high so that Si diffuses simultaneously into the PdTa layer. The variable energy positron annihilation measurements indicate, that the out-diffusion of Pd is not accompanied by a massive out-annealing of defects. After silicidation the surface layers still contained a very high defect concentration.

According to Hung et al. and Saris et al. (9,19) the reaction temperature of amorphous alloys composed of a refractory metal and a near noble metal is the same as that of the refractory metal with Si. Our observations corroborate this rule; for all amorphous compositions the reaction was far advanced, resulting in Si at the specimen-surface, already after annealing at 600 to 650 °C. However, we also observed a dependence of the temperature at which Pd diffused out on the composition, regardless whether a solid solution or an amorphous alloy was involved. Therefore an explanation is proposed analogous to the rule concerning the crystallization temperature, as originally proposed by Buschow (20) and modified by Barbour et al. (21). For relatively stable amorphous binary alloys crystallization proceeds by diffusion, the diffusion is possible only if sufficient holes of atomic size are available. The crystallization temperature is therefore related to the hole enthalpy. Buschow originally proposed to make a correlation with the enthalpy of a hole with the size of the smaller atom. Barbour et al. made it clear that it is more correct to take a hole with the size of the larger atom. In this specific study the rate-limiting factor for the formation of Pd₂Si is the transport of Pd

through the Pd-Ta layer, and not the interdiffusion of Pd and Si; the latter takes place already at 200 °C (22). Moreover, the results for the Pd-rich compositions indicate that the formation of TaSi₂ is not a necessary condition for the reaction to proceed. Consequently, the temperature at which a substantial amount of Pd₂Si is formed will depend on the diffusion of Pd through the amorphous Pd-Ta layer and therefore has to be correlated with the enthalpy of a hole with the size of a Pd atom. Taking the proportionality constant of 4.5 K.mole/kJ proposed by Barbour et al. and using Miedema's approach (23,24) to estimate the hole enthalpies in a disordered alloy, we calculated the reaction temperatures as given in the fourth column of table 1. The trend is reproduced, but the quantitative accuracy is rather poor. Since the Miedema model is best in predicting general trends, but may yield less accurate data in specific cases, it may be better to use a slightly different approach. According to Miedema (23,24) the enthalpy of a hole in a specific alloy is proportional to $V^{5/6}$, where V is the volume of the hole. The crystallization temperature is associated with the larger hole, and Pd diffusion with the smaller hole. Therefore, we propose that the initial temperature at which Pd reacts with Si is related to the crystallization temperature as follows:

$$T_{\text{reac}}/T_{\text{cryn}} = (V_{\text{small}}/V_{\text{large}})^{5/6}$$

In the last column of table 1 reaction temperatures are given as calculated with this rule. Its validity is certainly very limited. For reactions of amorphous alloys with other materials, like Al, the refractory metal may react already at such a low temperature that diffusion of the smaller atom is not the rate-limiting factor. Furthermore a necessary condition is that crystallization takes place by diffusion. For this specific case both conditions are fulfilled, and the agreement with the experimentally obtained results is very good, although more experiments are needed to test its validity more widely.

Shallow silicide formation

The formation of a shallow silicide by outdiffusion of one element from a binary alloy is only feasible, when the final layered structure is not very rough.

For all compositions we observed phase separation with XRD into the Pd₂Si and the TaSi₂ phase. The RBS measurements indicate that for the Pd-rich compositions a layered structure is formed, with a Pd₂Si layer growing first at the Si/Pd-Ta interface; upon further annealing the remaining Ta layer is silicidized as well, see figs. 2 and 3. For the amorphous films Pd out-diffusion is also observed, however, the final structure shows considerable mixing or roughening of the layered structure. The tentative explanation is as follows: pure Pd on Si reacts at 200 °C, forming Pd₂Si in the process (22). Pd₂Si can recrystallize at temperatures of

about 600 °C, but Pd₂Si on Si reacts to form PdSi only when heated at 800 °C according to Tu (25). If the temperature, at which Pd is released from the metal overlayer, is higher than 500 °C, as is the case for the amorphous films, interface roughening takes place already during silicide formation. The formation of PdSi is not very likely at this temperature, so the roughening will be caused by the growth process of the Pd₂Si layer. The observed roughness is more pronounced than for initially smooth Pd₂Si layers, that are post-annealed at 600 °C. In fig. 6 schematical cross sections are shown of the layer structure before and after annealing.

Barrier function of Pd_xTa_{1-x}

The diffusion of Si through a PdTa overlayer was investigated. Interesting is, that for compositions between Pd_{0.6}Ta_{0.4} and Pd_{0.2}Ta_{0.8} as-deposited films are amorphous so that at two different compositions the influence of the microstructure on the barrier function is observed. Furthermore, the overall effect of the composition on the failure temperature is studied. The following points are worth noting.

(i) The failure temperature depends on the composition: it increases with Ta-content. The failure temperature is not correlated with the temperature at which Pd starts to diffuse out, but with the temperature at which the remaining Ta layer takes up Si. There are two reasons for this composition dependence: the thickness of the Ta-enriched layer increases and the defect density decreases with increasing initial Ta concentration, both leading to a higher failure temperature.

(ii) At the Ta-rich side there is no difference in the ultimate failure temperature between crystalline and amorphous alloys, clearly demonstrating that the barrier function is performed by the Ta layer remaining after out-diffusion of Pd.

(iii) For the crystalline Pd_{0.1}Ta_{0.9} and Ta overlayer the failure temperature is sometimes rather low, indicative for diffusion through the crystalline alloy. Since it is not reproducible and since the TEM images showed large cracks, we do not believe that this is a property intrinsically associated with the crystalline state. Instead, it reflects the limited ability of e-gun deposited crystalline alloy films to remain continuous.

(iv) At the Pd-rich side there is a distinct difference between the failure temperature of crystalline and of amorphous alloys; the latter is 150 °C higher. Both RBS and XRD indicate that Pd₂Si formation is delayed for the amorphous alloys, proving that Pd diffusion in these specific Pd(Ta) solid solutions takes place at a lower temperature than in the amorphous alloys.

Generally this study shows that the composition dependence of barrier layers can be quite substantial. An example of this is found in a recent study of Hornstrom et al. (26). They compared the barrier performance of crystalline $W_{0.7}Re_{0.3}$ with that of amorphous $W_{0.30}Re_{0.61}$ and $W_{0.28}Re_{0.72}$. Although being amorphous, the alloy with the latter composition showed the most pronounced reaction with Al. This is a clear demonstration of the compositional effect; the heat of formation of Re-Al alloys is considerably more negative than that of W-Al alloys.

5. CONCLUSIONS

- Growth of a shallow silicide by diffusion of Pd out of a PdTa alloy results in a homogeneous layer of Pd_2Si only for the more Pd-rich alloys. For alloys with more than 40 at.% Ta the out-diffusion of Pd occurs at such high temperatures that a rough Pd_2Si layer is formed, furthermore, Pd_2Si is found on top of the film as well.
- The temperature at which Pd diffuses out is composition dependent, and is approximately proportional to the enthalpy of a hole with the size of the Pd atom in the PdTa alloy.
- The barrier function of amorphous PdTa alloys is considerably better than that of Pd-rich solid solutions. Si is observed at the surface only after annealing at 600 to 650 °C. However, the failure temperature of the amorphous alloy does not exceed the ultimate failure temperature of the Ta-rich crystalline alloys, indicating that the crystalline state in itself not necessarily has to lead to an inferior barrier performance.
- The reproducibility of the failure temperature for the amorphous alloys was very good. Ta-rich crystalline specimens occasionally were observed to have a relatively low failure temperature. This is probably caused by deterioration of the film, as pores and cracks were observed with TEM.
- The reaction of a metal overlayer with Si is detectable with variable energy positron annihilation. Information about the defect density in the outer layers can be obtained, that is complementary to that obtained by other techniques.

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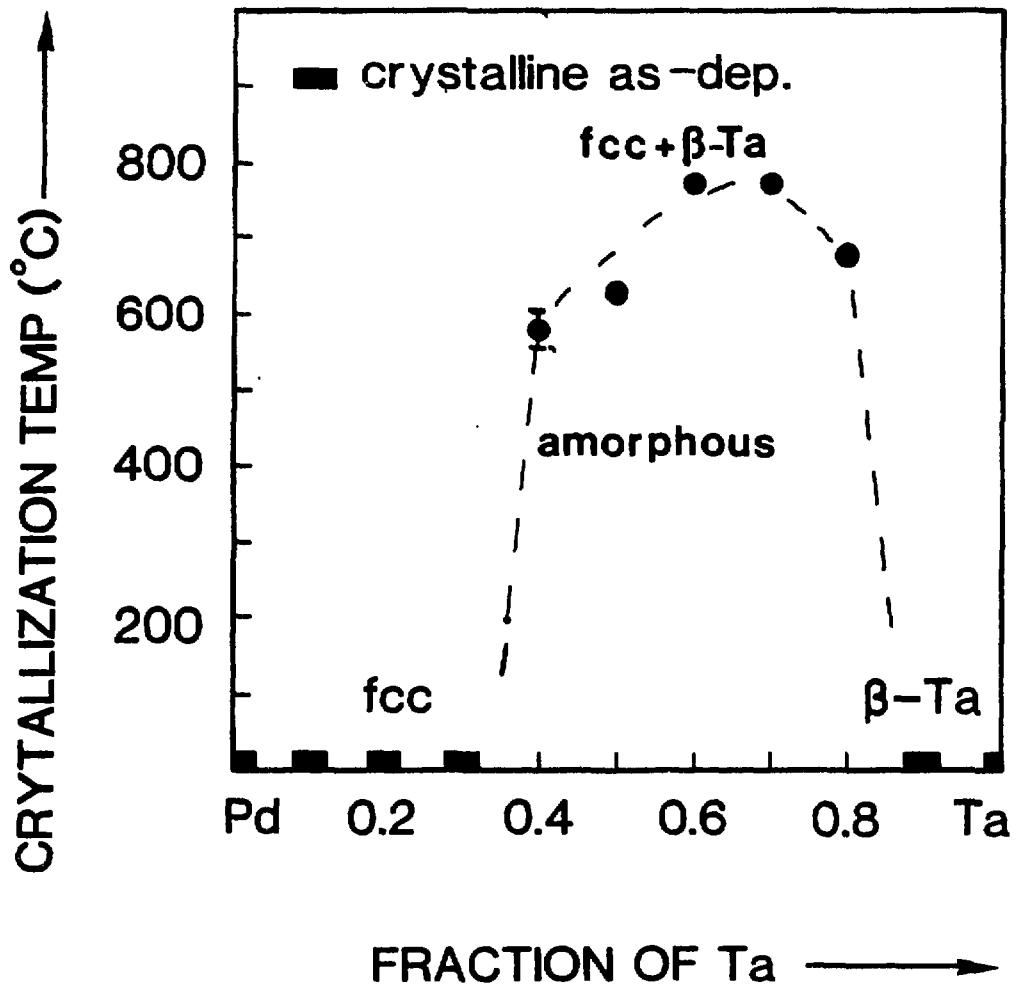
TABLE 1

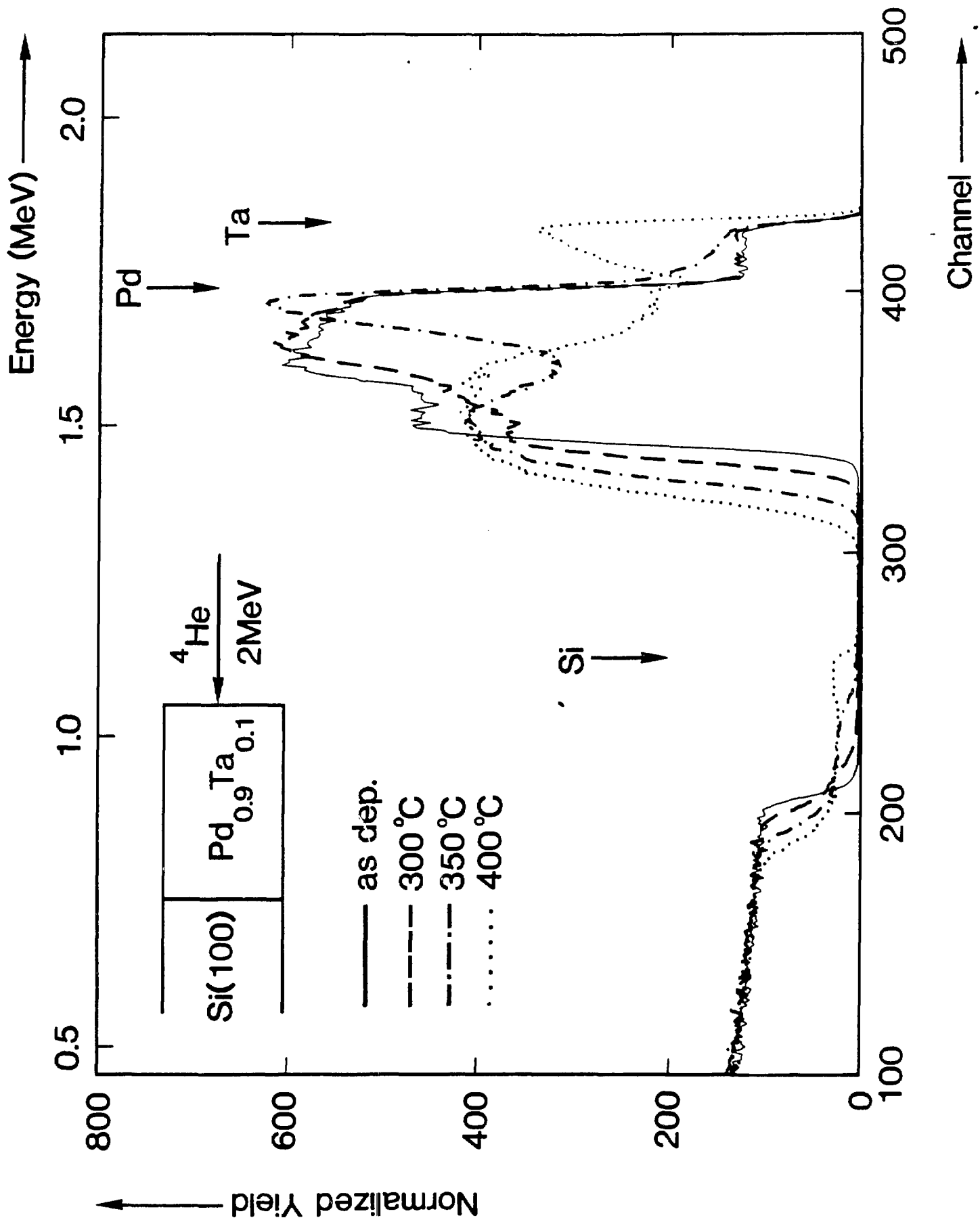
Annealing temperatures at which TaSi₂ and Pd₂Si phases were observed with X-ray diffraction, at which Pd diffusion was observed with RBS, and predictions for Pd diffusion using Barbour's constant and using the crystallization temperature.

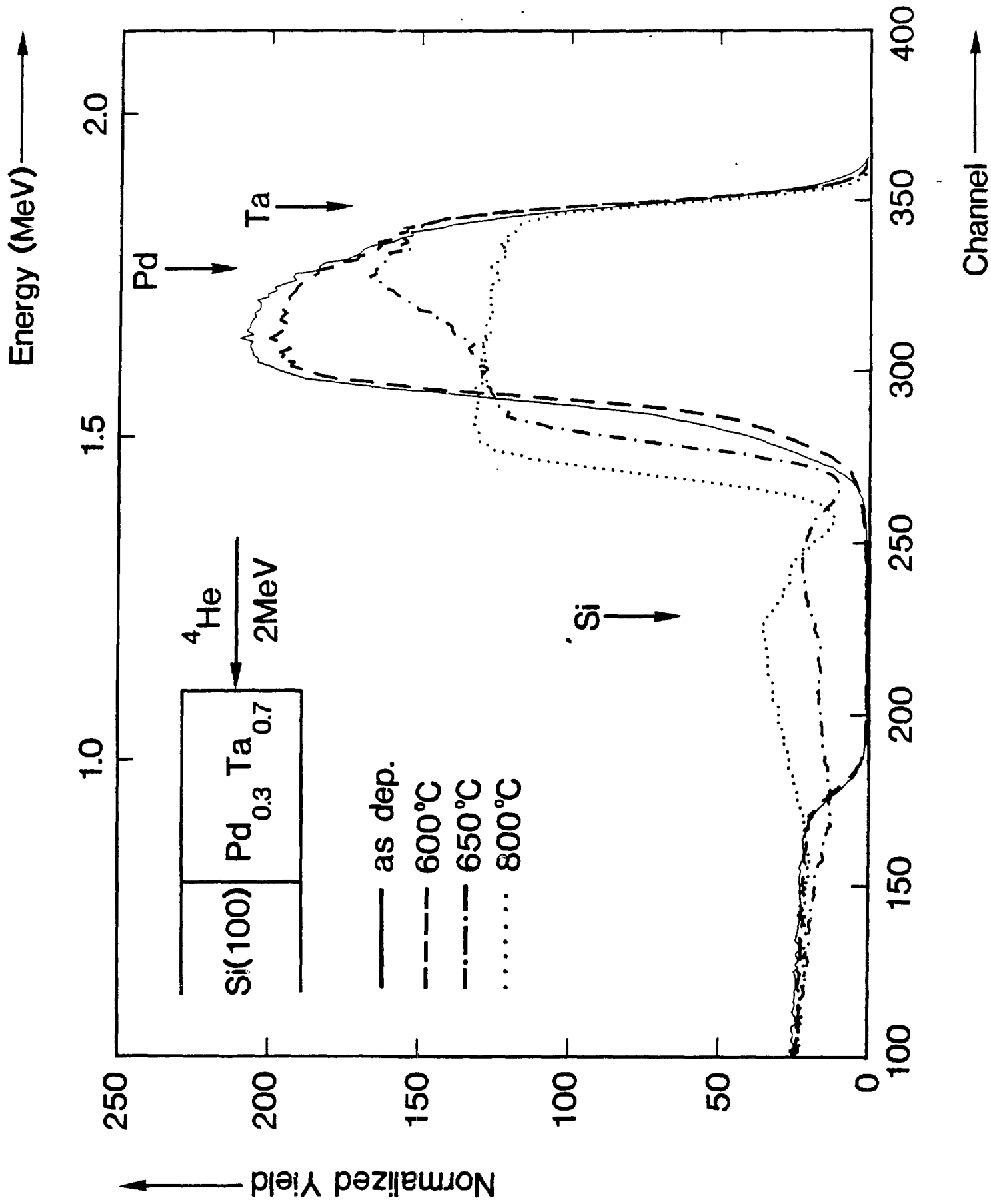
	TaSi ₂ (XRD)	Pd ₂ Si(XRD)	Pd diff. (RBS)	Pd diff. (Barbour)	Pd diff. (cryst.)
Pd _{0,9} Ta _{0,1}	450°C.	300°C.	300°C.	320°C.	
Pd _{0,7} Ta _{0,3}	500°C.	350°C.	350°C.	370°C.	
Pd _{0,5} Ta _{0,5}	650°C.	500°C.	500°C.	420°C.	490°C.
Pd _{0,3} Ta _{0,7}	650°C.	600°C.	600°C.	470°C.	620°C.

FIGURE CAPTIONS

- (1) Phases of as-deposited crystalline PdTa films, and crystallization temperature and crystallization products of amorphous PdTa films. The film thickness was 30 nm, observations were by TEM.
- (2) Rutherford Backscattering spectra for Si(100)/Pd_{0.9}Ta_{0.1} samples before and after annealing at the temperatures indicated.
- (3) Rutherford Backscattering spectra for Si(100)/Pd_{0.3}Ta_{0.7} samples before and after annealing at the temperatures indicated.
- (4a) Line-shape parameter S of the 511 keV annihilation gamma peak as a function of the energy of a positron beam, incident on a 73 nm thick Pd_{0.51}Ta_{0.49} overlayer on Si(100); as-deposited and after annealing at 600 °C and 700 °C. Lines are drawn to guide the eye.
(4b) $\Delta S = S(600\text{ °C})$ minus $S(\text{as-deposited})$.
- (5) Annealing temperatures at which Si was observed at the surface with AES for 200 nm thick Pd_xTa_{1-x} overlayers on Si(100). The amorphous phase formation range is also indicated.
- (6) Schematical cross-section of specimens before and after annealing for a Pd-rich and a Ta-rich composition.



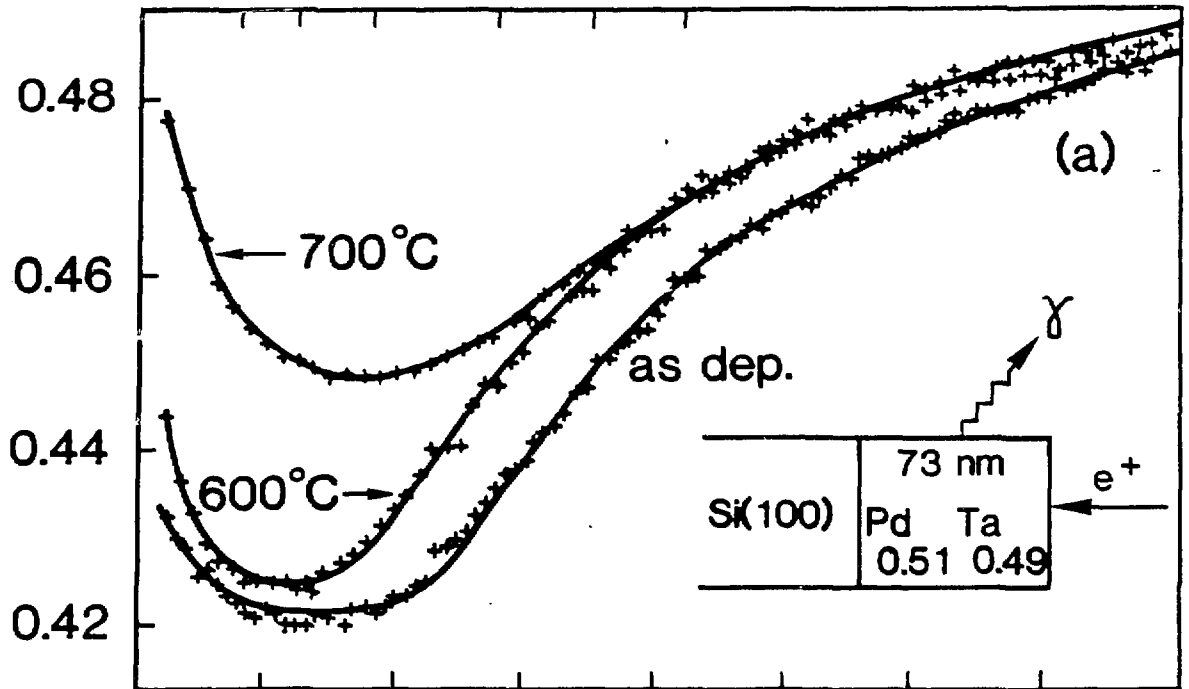




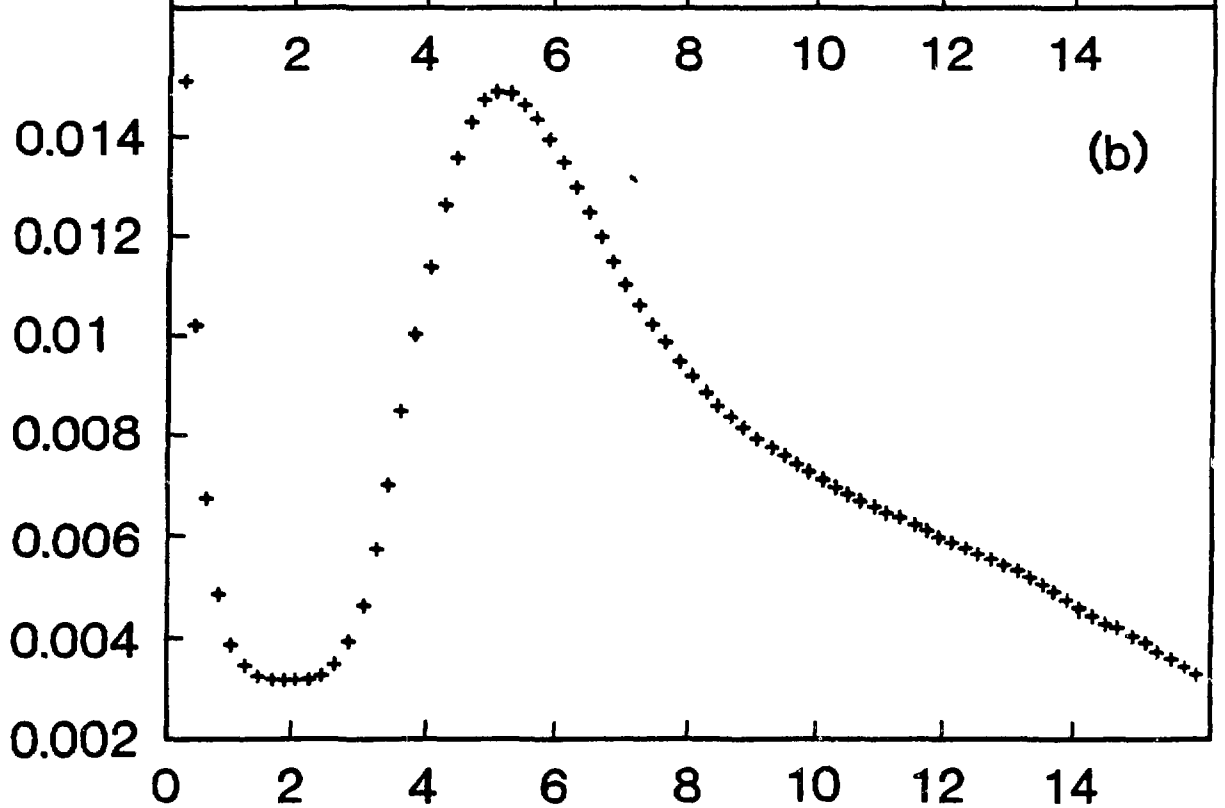
ESTIMATED DEPTH SCALE (nm) \longrightarrow

5 10 20 40 60 80

LINE SHAPE
PARAMETER S



$\Delta S = S(600^\circ\text{C}) - S(\text{as dep.})$



INCIDENT POSITRON
ENERGY E_i (keV) \longrightarrow

