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Mobility of Pd and Si in Pd_2Si

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A diffusion-membrane technique is developed and used to determine the mobilities of Pd and Si in Pd_2Si. A multilayer film structure $Pd/Pd_2Si/Ge$, in which Ge serves as a sink for Pd, is used to monitor the flux of Pd through Pd_2Si. The flux of Si through Pd_2Si is determined by using the thin-film structure $Si/Pd_2Si/Cr$ in which Cr serves as a sink for Si. Values for the mobility of Pd and Si are calculated from the measured flux rates. Good agreement is found between the calculated Pd mobility values and the parabolic rate constant for Pd_2Si growth around 200 °C. The mobility of Si at these temperatures is found to be orders of magnitude lower. The activation energy for Si mobility is found to be 2.1 eV in epitaxial Pd_2Si and 1.7 eV in polycrystalline Pd_2Si.

I. INTRODUCTION

The wide applicability of near-noble-metals and their silicides in microelectronic circuitry has stimulated extensive research into the properties of these materials. A good knowledge and thorough understanding of the silicon/metal interaction are essential for successful utilization of silicon/metal and silicon/silicide contacts. Aluminum, which is commonly used for metallization, may penetrate the metal or silicide layer, causing pitting of the contact. Refractory metals such as Cr, Ti, or W have been recognized to serve as sacrificial diffusion barriers to Al when introduced between the near-noble-metal and the Al layer.[1,2] The growth kinetics of the silicon/metal and aluminum/metal compounds during interdiffusion of such multilayer films determine the usefulness of such metallization schemes. The Si (and metal) transport properties of the various layers are therefore important parameters necessary to evaluate the applicability and reliability of these metallization schemes.

Although the silicide formation of Pd, one of the near-noble-metals, has received considerable attention, conclusions regarding the dominant diffusing species during silicide formation are inconsistent.[3-8] The variety of experiments from which conclusions were drawn was carried out under various experimental conditions using different techniques. The results tend to suggest that both Pd and Si diffuse during Pd_2Si growth from Si/Pd couples. The flux ratios of Pd and Si differ from experiment to experiment.

The general (empirical) rule is that when the phase that forms is rich in the metal (or Si) the richer element is also the dominant diffusing species. Both Ni and Pt have been found to be the dominant moving species during Ni_2Si and Pt_2Si formation, respectively.[6] It is therefore surprising that Pd_2Si should deviate from the empirical rule since Pd, Ni, and Pt all belong to the same group of near-noble-metals. Further investigation into the mobility of Pd and Si in Pd_2Si is therefore appropriate.

We have developed a diffusion-membrane technique using multilayer films to determine the mobility of Pd and Si in Pd_2Si. A comparison of these values with the parabolic rate constant for Pd_2Si growth should contribute to a better understanding of the diffusion mechanism during its formation.

II. MEASURING PRINCIPLE

Various techniques have been used to determine the intrinsic diffusion coefficients of metal and silicon in metal silicides. Tu made use of implanted Xe markers to determine the intrinsic diffusivity of Ni and Si in Ni_2Si.[9] Pretorius and co-workers used radioactive Si tracers to determine the Si self-diffusion (intrinsic diffusivity) in PtSi and NiSi.[10,11] Neither of these measuring techniques has been applied successfully to Pd_2Si.

We propose the following membrane technique to determine the mobility of Si and Pd in Pd_2Si. Consider the Pd_2Si layer as a membrane with a source supply of Si or Pd on the one side. The flux of Si or Pd through Pd_2Si can now be monitored with the aid of a suitably chosen layer on the other side which serves as a sink for the diffusing atoms [see Fig. 1(a)]. The thickness of the Pd_2Si layer is Y and it is assumed that A (Pd or Si) diffuses through Pd_2Si whereas B does not. On diffusing through, A and B form a compound A_xB. The flux of A through Pd_2Si, J_A, can be expressed in terms of the chemical potential difference of A across Pd_2Si, $\Delta\mu_A$, and the mobility of A in Pd_2Si, M_A:

$$J_A = -M_A C^A_{Pd_2Si} \Delta\mu_A / Y ,$$

where $C^A_{Pd_2Si}$ is the atomic concentration of A in Pd_2Si

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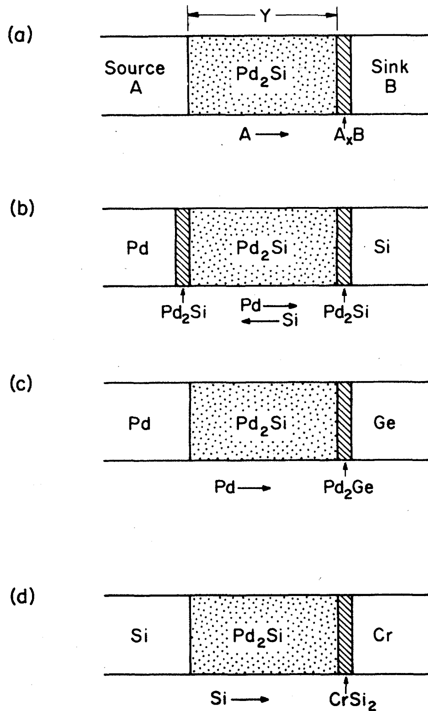

FIG. 1. Schematic of multilayer thin-film structures: (a) general, (b) Pd/Si couple, (c) Pd/Pd_2Si/Ge structure for determining Pd mobility, and (d) Si/Pd_2Si/Cr structure for determining Si mobility.

and Y is the thickness of the Pd_2Si layer. We now define the effective mobility of A in Pd_2Si as

$$\overline{M}_A = -M_A \Delta\mu_A ,$$

so that the flux becomes

$$J_A = \overline{M}_A C^A_{\mathrm{Pd_2Si}} / Y .$$

The rate of A_xB formation R_{A_xB}, in terms of the flux of A through Pd_2Si is given by

$$R_{A_xB} = J_A / C^A_{A_xB}$$

or

$$R_{A_xB} = \overline{M}_A C^A_{\mathrm{Pd_2Si}} / C^A_{A_xB} Y , \qquad (1)$$

where $C^A_{A_xB}$ is the atomic concentration of A in A_xB. The effective mobility of A in Pd_2Si is therefore given by

$$\overline{M}_A = R_{A_xB} Y C^A_{A_xB} / C^A_{\mathrm{Pd_2Si}} . \qquad (2)$$

The effective mobility $\overline{M}_A$ is not equivalent to the intrinsic diffusivity of A in Pd_2Si, since A is subject to a "driving force" $\Delta\mu_A$, but it is comparable to the parabolic rate constant $\widetilde{M}_{\mathrm{Pd,Si}}$, as determined from the growth kinetics of Pd_2Si:

$$\widetilde{M}_{\mathrm{Pd,Si}} = L^2/2t ,$$

where L is the thickness of Pd_2Si formed in time t. The equivalence between $\overline{M}$ and $\widetilde{M}$ can be shown by replacing A by Pd, B by Si, x by 2, Y by y, where y is the thickness of the growing Pd_2Si layer, and R_{A_xB} by dy/dt so that (2) becomes

$$\overline{M}_{\mathrm{Pd,Si}} = \frac{dy}{dt} y = \left. \frac{y^2}{2t} \right|_0^L = \frac{L^2}{2t} = \widetilde{M}_{\mathrm{Pd,Si}} .$$

We have used $\overline{M}_{\mathrm{Pd,Si}}$ rather than $\overline{M}_{\mathrm{Pd}}$ or $\overline{M}_{\mathrm{Si}}$ to indicate that the growth of Pd_2Si could be as a result of both Pd and Si diffusing through it [see Fig. 1(b)].

The following multilayer combinations are suggested for determining Pd and Si mobilities.

(i) Pd mobility. A layer of Ge can serve as a sink for Pd so that Pd_2Ge is formed at the Pd_2Si/Ge interface, as shown in Fig. 1(c).

(ii) Si mobility. The Cr layer which serves as a sink for Si results in $CrSi_2$ formation at the Pd_2Si/Cr interface, as is shown in Fig. 1(d).

The membrane technique as described above allows the determination of the effective mobility of Si or Pd through Pd_2Si, but does not give any other information regarding the diffusion mechanism except the activation energy. It is possible, in the case of the Si/Pd_2Si/Cr structures, that Pd_2Si can dissociate, giving up Si atoms to form $CrSi_2$ and releasing Pd atoms to diffuse through the Pd_2Si layer to the Si/Pd_2Si interface to form new Pd_2Si. In this case the measured effective mobility will be that of Pd and not Si. We believe that this mechanism is unlikely since in a very similar experiment involving Si transport through Pd_2Si, it was found that Pd_2Si does not dissociate during its oxidation at 750 °C.[12] This is in agreement with our assumption that Si and not Pd diffuses through Pd_2Si during $CrSi_2$ growth.

Since we have layers other than Pd or Si in contact with Pd_2Si, the driving force ($\Delta\mu$) across Pd_2Si is influenced, which in turn affects the values obtained for $\overline{M}_{\mathrm{Pd}}$ and $\overline{M}_{\mathrm{Si}}$. The driving forces operative in the various multilayer systems are related to the free energy of formation of the growing compound layers. Since the free energies of formation of the silicides are of the same order of magnitude[13] and since Pd_2Ge is expected to have a free energy of formation of similar magnitude, we believe that the difference in driving forces in the three cases Pd/Pd_2Si/Si, Pd/Pd_2Si/Ge, and Si/Pd_2Si/Cr will not account for large differences in the measured values of $\widetilde{M}_{\mathrm{Pd,Si}}$, $\overline{M}_{\mathrm{Pd}}$, and $\overline{M}_{\mathrm{Si}}$. A comparison of these values is therefore justified.

In order to verify that the growth of A_xB is limited by the diffusion of A through Pd_2Si, the following properties of the A_xB layer grown on Pd_2Si need to be investigated. (i) It should grow linear with time since $t^{1/2}$ kinetics would imply growth controlled by diffusion through a layer which grows with time, that being A_xB. (ii) Equation (1) predicts $R_{A_xB} \propto 1/Y$ so that the rate of A_xB growth should be inversely proportional to the thickness of the interposed Pd_2Si layer. If these two conditions are not satisfied, in which case one of the interfaces or diffusion through A_xB is controlling A_xB growth, then this membrane technique cannot be applied.

III. EXPERIMENTAL PROCEDURE

The various films were sequentially deposited onto chemically cleaned substrates by electron-beam evaporation of the metals at pressures below 10^{-6} Torr. Thin films of Ge, Si, and Pd were deposited on SiO_2 substrates whereas Pd and Cr were deposited on Si(111) and Si(100) substrates. Annealing was carried out in a quartz tube vacuum furnace at pressures below 2×10^{-7} Torr. The Ge/Si/Pd samples were pre-annealed at 220 °C for an hour to obtain the structure SiO_2/Ge/Pd_2Si/Pd. In cases where the Si layer was very thin or very thick, it was necessary to adjust the pre-annealing temperature or time accordingly to ensure complete Pd_2Si formation without any detectable Pd_2Ge. The formation of Pd_2Ge was obtained by further annealing the samples at temperatures between 200 and 240 °C for various periods of time.

By annealing the Si(111)/Pd/Cr samples at 380 °C for 10 min, Si(111)/Pd_2Si/Cr structures were obtained. The annealing time for Si(100)/Pd/Cr samples with approximately 1-μm-thick Pd layers, was increased to 50 min at 380 °C. No $CrSi_2$ formation was detected after such a heat treatment whereas Pd_2Si formation was complete. The samples were further annealed at temperatures between 410 and 500 °C for various periods of time during which $CrSi_2$ was formed on Pd_2Si. In order to investigate Pd_2Ge and $CrSi_2$ growth without an interposed Pd_2Si layer, SiO_2/Ge/Pd and Si(100)/Cr structures were prepared and annealed following the procedure outlined above.

Rutherford backscattering spectrometry employing mega-electron-volt $^4He^{2+}$ ions was used to determine the thickness of the various layers and the composition of the compounds formed. The phases of the various compounds were identified by means of glancing-angle x-ray spectroscopy using the Read and Guinier cameras. It is found that Pd_2Si grows epitaxially on Si(111), which was verified from the absence of the characteristic Pd_2Si lines on the x-ray micrographs.

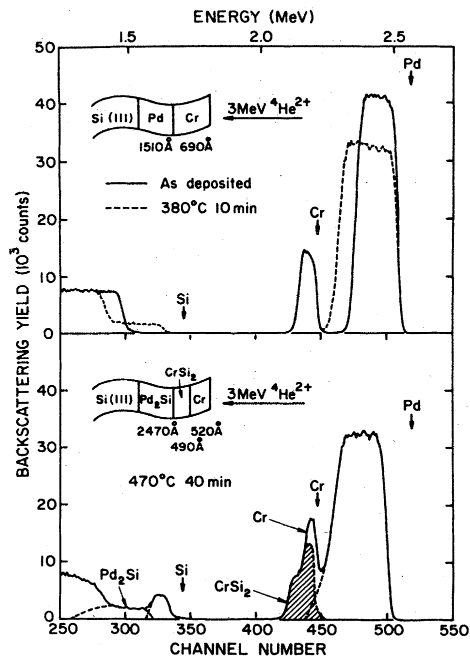

FIG. 2. Rutherford backscattering spectra of Si(111)/Pd/Cr samples: (top) as-deposited and after pre-annealing at 380 °C, and (bottom) after further annealing at 470 °C. The arrows indicate the energies of $^4He^{2+}$ ions scattered from the elements when located on the surface of the sample.

IV. RESULTS AND DISCUSSION

A. Si/Pd_2Si/Cr interdiffusion

Figure 2 shows the Rutherford backscattering spectra of Si(111)/Pd/Cr samples as deposited, after preannealing at 380 °C for 10 min and after further annealing at 470 °C for 20 min. For clarity, the spectrum of the sample annealed at 470 °C is shown separately. When comparing the two spectra in the top part of the figure, it can be seen that the Cr signal has remained unchanged which implies that no $CrSi_2$ was formed during the preannealing process. The broadening of the Pd signal and the appearance of the Si step indicate that palladium silicide has formed. The phase of this silicide could not be identified by means of glancing angle x-ray diffraction since the characteristic lines of Pd_2Si were absent due to the epitaxy of Pd_2Si grown on Si(111). The ratio of the Pd-signal height to the step height of the Si signal was used to establish the composition as Pd_2Si. The step in the Cr (low-energy side) and the second step in the Si signal (high-energy side) in the bottom part of the figure indicate that $CrSi_2$ has formed. The width of both these steps are measures for the thickness of the $CrSi_2$ layer. Both Si and Cr signals of $CrSi_2$ formed on thick Pd_2Si (~ 1 μm) are superimposed on the Pd signal. Since the Cr signal is larger than the Si signal, it was used for determination of the $CrSi_2$ layer thickness.

No Cr diffusion into the Pd or Pd_2Si layers is observed in any of these spectra or spectra of similar samples. The Pd_2Si/$CrSi_2$ interface (high-energy side of the Pd signal in Fig. 2) has remained sharp, although Si has diffused through Pd_2Si to form $CrSi_2$. Degradation of the Si(111)/Pd_2Si interface (long tail of the low-energy edge of Pd signal) is observed when comparing the Pd signals of the spectra of the samples annealed at 380 and 470 °C. We believe that the nonuniform Si(111)/Pd_2Si interface as indicated by the tail in the Pd signal arises from the release of Si from the substrate. The mechanism of Si release is still unknown but could involve Pd_2Si dissociation at the Si(111)/Pd_2Si interface. The Si(100)/Pd_2Si interface of similar samples (same metal-layer thickness and heat treatment) is found to remain uniform during $CrSi_2$ growth and is therefore not affected by the Si release or transport process.

In simple metal/silicon or metal/metal phase-forming interactions, it is relatively easy to conclude from the growth kinetics whether the interaction is controlled by an interface reaction (t kinetics) or by diffusion of the reactants ($t^{1/2}$ kinetics). The interpretation of multilayer growth kinetics is complicated by the fact that a large number of interfaces and layers can control the layer

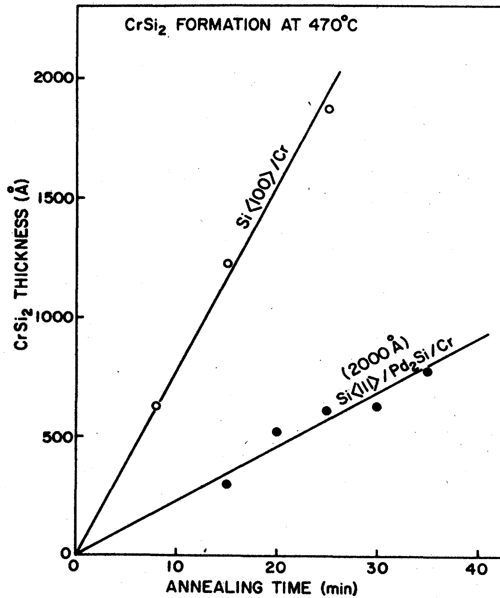

FIG. 3. Growth kinetics of $CrSi_2$ at 470 °C, illustrating the effect of interposed Pd_2Si on the growth rate.

growth. In such cases linear growth kinetics are no longer a sufficient condition for the interaction to be controlled by an interfacial reaction.

Figure 3 shows a plot of $CrSi_2$ thickness formed at 470 °C against annealing time. The growth of $CrSi_2$ on Si(100) using Si/Cr thin-film couples is compared with $CrSi_2$ growth on 2000-Å-thick epitaxial Pd_2Si using Si(111)/Pd/Cr multilayer films. Since the $CrSi_2$ formation on Si is linear with time, we conclude that it is limited by an interface reaction. The linear kinetics for the

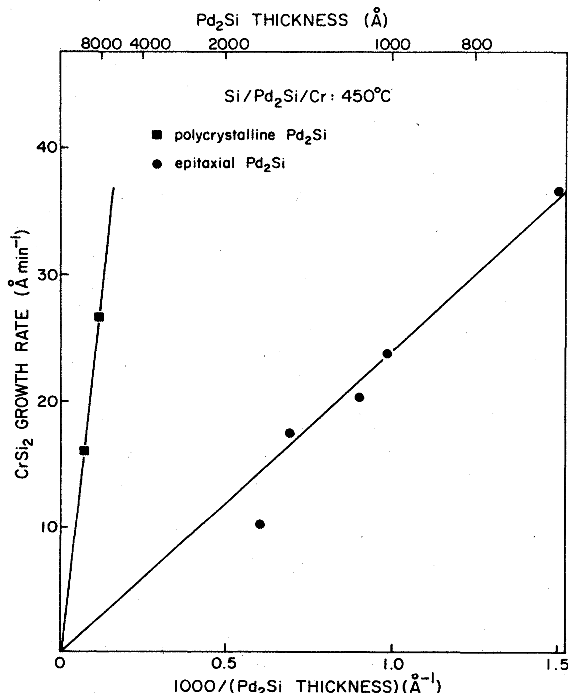

FIG. 4. The rate of $CrSi_2$ growth on Pd_2Si is shown to be inversely proportional to the Pd_2Si layer thickness.

case of interposed Pd_2Si does not necessarily imply that it is also reaction limited. Equation (1) predicts that the $CrSi_2$ formation will be linear with time (constant rate) and inversely proportional to the interposed Pd_2Si layer thickness when it is controlled by diffusion through the Pd_2Si layer. The lower growth rate (gradient) for $CrSi_2$ on Pd_2Si in Fig. 3 is a direct result of the diffusion limiting effect of the Pd_2Si layer.

Figure 4 shows the variation of $CrSi_2$ growth rate with reciprocal Pd_2Si thickness at 450 °C for both epitaxial and nonepitaxial Pd_2Si. It is obvious that the growth rate of $CrSi_2$ is inversely proportional to the interposed Pd_2Si layer, confirming the prediction of (1). The more thickness of nonepitaxial Pd_2Si necessary to limit Si transport for $CrSi_2$ growth is related to the greater mobility of Si in polycrystalline Pd_2Si compared to epitaxial Pd_2Si. The effect of the Pd_2Si layer thickness on $CrSi_2$ growth rate is discussed in greater detail in Ref. 14.

B. Pd/Pd_2Si/Ge interdiffusion

The backscattering spectra of Ge/Si/Pd samples are shown in Fig. 5. The spectrum of the sample annealed at 220 °C for 3 h is shown separately. On comparing the two spectra in the top of the figure, a broadening of the Si signal as well as a step in the Pd signal are observed. This indicates that Pd_2Si, which was confirmed by x-ray diffraction, has formed. The Ge signal around channel 430 has remained unchanged, indicating that no Pd_2Ge has formed on pre-annealing at 220 °C. After further annealing the sample at 220 °C for 3 h, a change in the Ge signal is observed (see bottom of Fig. 5). The decrease of the un-

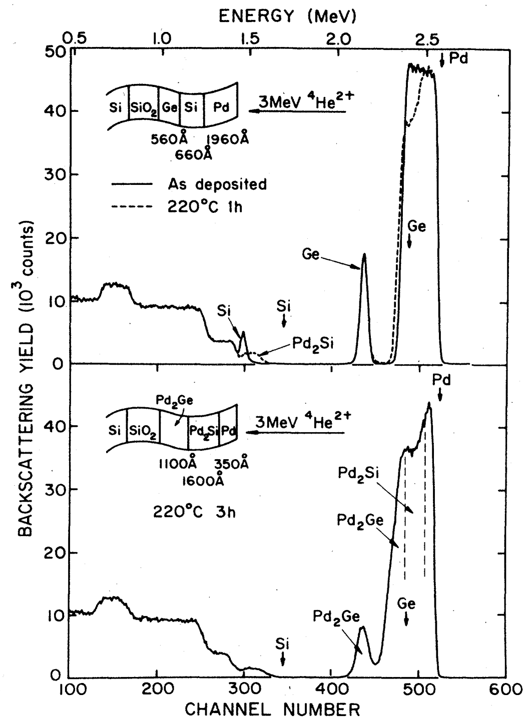

FIG. 5. Backscattering spectra of SiO_2/Ge/Si/Pd structures: (top) as-deposited and after pre-annealing at 220 °C, and (bottom) after further annealing at 220 °C, Pd_2Ge is formed.

reacted Pd was determined and from that the Pd_2Ge thickness was calculated.

It is well documented that impurities in a film can influence the diffusion of atoms through it.[15–17] The extent to which impurities enhance or inhibit diffusion depends on their interaction with the matrix and diffusing atoms and on the diffusion mechanism. Experimentally obtained values for diffusion coefficients could therefore be perturbed by foreign atoms in the diffusing medium. The bottom spectrum in Fig. 5 indicates detectable amount of Ge diffusion into the Pd_2Si layer. Germanium does not differ much from silicon with respect to its chemistry and atomic volume. We therefore believe that the presence of Ge in Pd_2Si will not have an appreciable influence on the diffusion of Pd through the Pd_2Si layer.

The growth of Pd_2Ge at 200 °C is illustrated in Fig. 6. Linear thickness and time scales are used in the figure in order to compare the two cases (with and without interposed Pd_2Si). The parabolic growth (linear when the thickness is plotted against the square root annealing time) of Pd_2Ge from Pd/Ge couples is characteristic of diffusion-limited growth and in agreement with reported studies on Pd_2Ge formation.[18] The interposed Pd_2Si layer is seen to have a dramatic effect on the Pd_2Ge kinetics. It was pointed out in Sec. II that linear kinetics in a multilayer-compound-forming system is not necessarily indicative of interfacial-reaction-controlled growth. Although the growth of Pd_2Ge on Pd_2Si is seen to be linear with annealing time, it is in fact limited by diffusion of Pd through the Pd_2Si layer, the thickness of which does not increase with time.

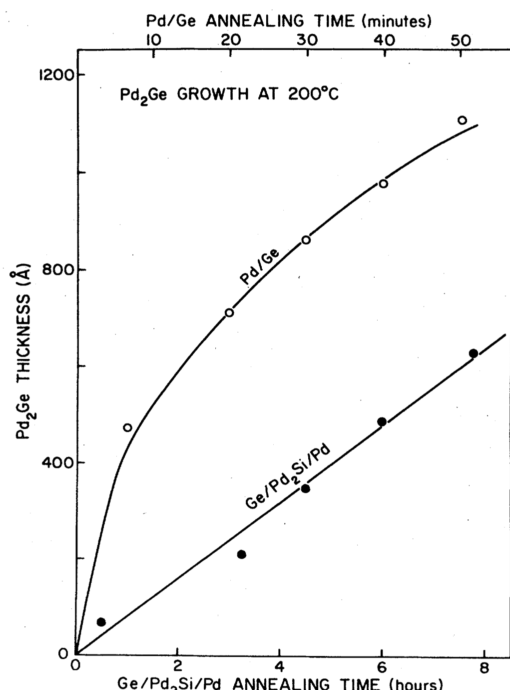

FIG. 6. Growth kinetics of Pd_2Ge at 200 °C, showing parabolic growth for Pd/Ge couples and linear growth for $Ge/Pd_2Si/Pd$ structures. Note that different time scales are used for the two curves.

C. Mobility of Si and Pd

The effective mobilities of Si and Pd in Pd_2Si were calculated from Eq. (2), and the values of the parameters used are tabulated in Table I. These mobility values are plotted against the reciprocal temperature in Fig. 7. Curves representing the parabolic rate constant for Pd_2Si growth, obtained from the literature,[8,19] are also shown in this figure. Good agreement between the effective mobility of Pd and the parabolic rate constant is obvious. This would suggest that the Pd mobility is sufficiently high to account for the Pd_2Si growth.

Calculated values for Si mobility in epitaxial and polycrystalline Pd_2Si are seen to be significantly different. The slopes of the two curves represent activation energies of 2.1 and 1.7 eV associated with Si diffusion through epitaxial and polycrystalline Pd_2Si, respectively. The higher mobility of Si in polycrystalline Pd_2Si could be attributed to Si grain-boundary diffusion. Furthermore, grain boundaries can serve as sources of point defects, thereby enhancing the Si vacancy concentration in polycrystalline Pd_2Si, resulting in a high Si mobility. The high activation energy of 2.1 eV for epitaxial Pd_2Si is characteristic of a lattice diffusion process, e.g., vacancy diffusion. [Similar values were obtained for Si self-diffusion in PtSi (2.1 eV) and NiSi (2.3 eV).[10,11]] The activation energy associated with Pd_2Si growth (epitaxial and polycrystalline) was reported to be approximately 1.0 ± 0.1 eV.[8] It is therefore obvious that Si is not the only diffusing species during Pd_2Si growth. This is supported by the results of Si marker experiments in Ref. 6. A probable diffusion mechanism for Pd_2Si growth is a diffusion process in

TABLE I. Data used in calculating the effective mobility through a Pd_2Si membrane using the relation $\overline{M}=R_{A_xB}YC^A_{A_xB}/C^A_{Pd_2Si}$, where A is the diffusant, B is the sink, the phase formed is A_xB, Y is the membrane thickness, and R the growth rate of A_xB. $C^{Si}_{CrSi_2}/C^{Si}_{CPd_2Si}=2.35$; $C^{Pd}_{Pd_2Ge}/C^{Pd}_{Pd_2Si}=0.96$.

T (°C)	$Y(Pd_2Si)$ (Å)	A_xB rate R (Å/s)	Mobility $\overline{M}$ (10^{-13} cm²/s)
	Si through epitaxial Pd_2Si to from $CrSi_2$		
450	1100	0.3	0.77
470	1000	0.75	1.75
485	1420	1.14	3.8
500	1420	2.9	7.6
	Si through polycrystalline Pd_2Si to from $CrSi_2$		
420	9500	0.12	2.7
425	9500	0.16	3.6
445	9500	0.45	9.9
445	15 500	0.27	9.8
455	9500	0.52	11.6
470	9500	0.85	19.0
470	15 500	0.47	17.2
480	9500	1.1	25.0
	Pd through polycrystalline Pd_2Si to form Pd_2Ge		
200	1600	0.022	0.034
220	1600	0.083	0.132

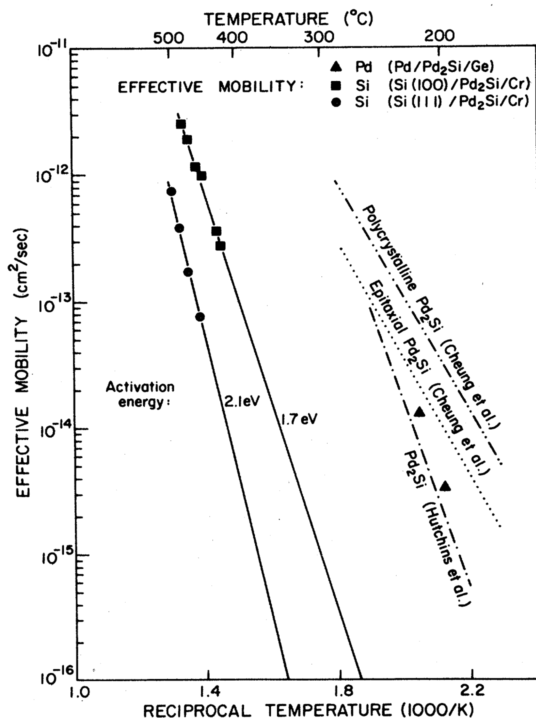

FIG. 7. Arrhenius plot of the effective mobilities of Pd and Si in Pd_2Si. Also shown are the parabolic rate constants for Pd_2Si from Pd/Si couples from Refs. 8 and 17.

which the Pd and Si fluxes are coupled, which results in a significantly lower activation energy than that measured for Si mobility. Although the activation energy for Si mobility in polycrystalline Pd_2Si is lower than that through epitaxial Pd_2Si due to the presence of grain boundaries, it is still significantly higher than the activation energy for Pd_2Si growth.

When the Si mobility curves are extrapolated down to approximately 200 °C, the values are found to be orders of magnitude lower than the parabolic rate constant for both epitaxial and polycrystalline Pd_2Si growth. The mobility of Si is therefore far too low to account for the Pd_2Si growth. This confirms the earlier statement that Si is not the only moving species during Pd_2Si growth.

The validity of the comparison of the effective mobility obtained by means of the membrane technique to the parabolic rate constant as obtained from the growth kinetics of Pd_2Si is based on the assumption that the chemical potential difference $\Delta\mu$ across the Pd_2Si layer is not very different in the three cases Si/Pd_2Si/Pd, Si/Pd_2Si/Cr, and Pd/Pd_2Si/Ge. Based on free-energy-of-formation considerations, we believe the driving forces in the three cases to be of the same order.

V. SUMMARY AND CONCLUSIONS

A diffusion-membrane technique has been developed and successfully applied to determine the mobility of Pd and Si respectively in Pd_2Si. A multilayer film consisting of Ge/Si/Pd was used to determine the mobility of Pd. Initial annealing of this system produced Ge/Pd_2Si/Pd. During subsequent annealing the flux of Pd through Pd_2Si was monitored with the aid of Ge which served as a sink for Pd by forming Pd_2Ge. The mobility of Si in Pd_2Si was determined in a similar manner using Si(111)/Pd/Cr and Si(100)/Pd/Cr structures. In this case Cr served as a sink for Si and formed $CrSi_2$ with the Si which diffused through the Pd_2Si layer in Si/Pd_2Si/Cr.

The mobilities of Pd and Si were calculated by making use of the rate of Pd_2Ge and $CrSi_2$ formation, respectively. The value for Pd mobility determined around 200 °C agrees with the parabolic rate constant for Pd_2Si growth. The mobility of Si in both polycrystalline and epitaxial Pd_2Si at this temperature is found to be orders of magnitude lower. The higher Si mobility in polycrystalline Pd_2Si is attributed to the presence of grain boundaries. An activation energy of 2.1 eV has been determined for the Si mobility in epitaxial Pd_2Si, whereas the value of 1.7 eV has been found for polycrystalline Pd_2Si. Because of the relatively low Si mobility we conclude that Si is not the only diffusing species during Pd_2Si growth. Radioactive marker experiments indicate that both Pd and Si diffuse with comparable fluxes during Pd_2Si formation.[6] This suggests that the atomic transport mechanism in the diffusion membrane (Pd_2Si) is not necessarily identical to the diffusion mechanism during the membrane compound (Pd_2Si) growth. Care should therefore be taken when comparing the mobilities to the parabolic rate constant for drawing conclusions regarding the dominant diffusion species during thin-film compound formation.

The successful application of the diffusion membrane technique to the Pd_2Si system demonstrates its use as a method for determining the mobility of components in a compound. The technique can be applied to other systems provided suitable combinations of multilayer films can be found.

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