

Modeling Metallic Precipitate Dissolution in Silicon Under Point Defect Injection

Subcontract Report
NREL/SR-520-37991
May 2005

Final Subcontract Report
20 January 2004 – 19 January 2005

T.Y. Tan
Duke University
Durham, North Carolina

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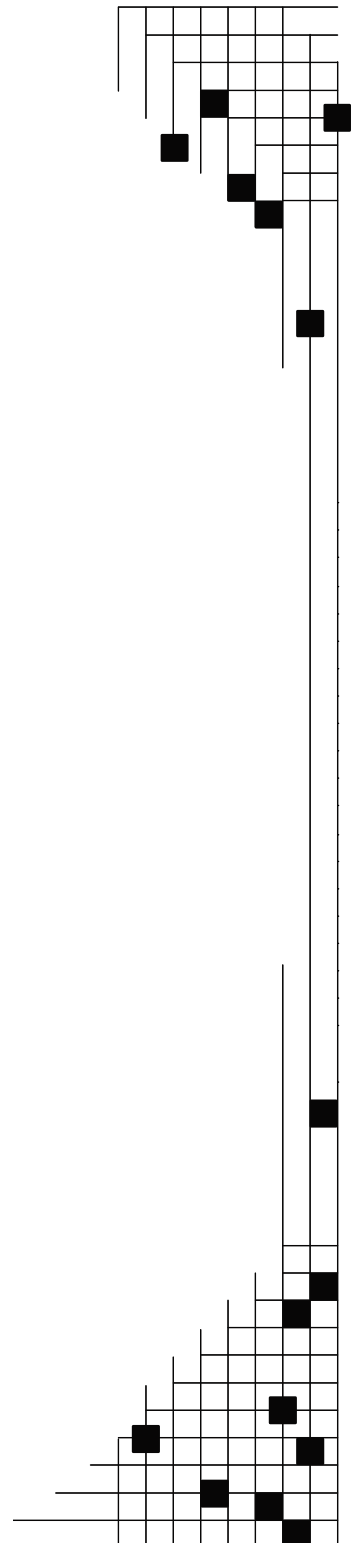
T.Y. Tan
Duke University
Durham, North Carolina

NREL Technical Monitor: Richard Matson
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National Renewable Energy Laboratory
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INTRODUCTION

Gettering of dissolved and precipitated metallic contaminants is important in Si solar cell fabrication for improving the cell efficiency. Gettering is needed because Si is an indirect band-gap semiconductor, with its carrier lifetime controlled by electrically active impurities (metals) and defects. Such impurity and defect electrical activities are responsible for the desirable fast action of switching devices, but also for the detrimental result of device/circuit failures due to excessively large pn junction leakage current and extremely short storage capacitor charge holding times. In the case of solar cells, these impurities and defects lead to low cell efficiencies. Point defects must be present to their thermal equilibrium concentrations. The presence of impurities is, however, an extraneous factor that can be controlled by cleaning and by gettering.

We have recently devoted an effort to identify the physics and to model the metal electrical activity,¹⁻³ and the metal gettering processes.⁴⁻⁷ The latter includes metal dissolution from the precipitates, diffusion of metal atoms to, and their stabilization at the gettering sites. In the present study, we further address the role of point defect in affecting the precipitate dissolution process, which originates from the fact that almost all metal silicide precipitate species are associated with a volume misfit with the Si matrix. We have formulated the problem using an Al-Si liquid layer as the gettering substance and interstitial Fe and FeSi₂ as the metal species and precipitates that are gettered. In this problem, the misfit of the precipitate is assumed to be accommodated by the point defect species vacancies (V) and Si self-interstitials (I). The point defects are either generated or absorbed during gettering and are hence rendered non-equilibrium for which equilibrium restoring process is assumed to be in- or out-diffusion from or to the Si exterior surfaces. Based on the physical modeling, numerical simulation results showed that injection of the appropriate point defect species from the Si exterior surfaces can be an effective means of speeding up the gettering process.

FORMULATION OF THE PROBLEM

Gettering of M by the Al-Si liquid involves the segregation processes taking place at the interface of Si and the liquid layer, which occurs simultaneously with impurity diffusion. Accounting for M diffusion, segregation, and dissolution of the precipitates, the time change rate of M is given by

$$\frac{\partial C_M}{\partial t} = \frac{\partial}{\partial x} \left[D_M \left(\frac{\partial C_M}{\partial x} - \frac{C_M}{m_M} \frac{\partial m_M}{\partial x} \right) \right] - \rho \frac{4\pi r^2}{\Omega_{MSi_x}} \frac{dr}{dt}, \quad (1)$$

where D_M is the M diffusivity, C_M is the M concentration in Si, m_M is the M segregation coefficient, ρ is the precipitate density, r is the radius of the precipitate, Ω_{MSi_x} is the volume of one MSi_x molecule, and dr/dt is the precipitate radius change rate given by

$$\frac{dr}{dt} = \Omega_{MSi_x} \frac{C_M - C_M^*}{r}, \quad (2)$$

where C_M^* is the dynamical equilibrium concentration of M at the precipitate-Si interface which maintains the precipitate to neither grow nor shrink.

Irrespective of whether the impurity is a substitutional or an interstitial species, upon precipitation, there is usually a volume change between the precipitate and the involved metal and Si atoms, i.e., the misfit. For the silicide MSi_x with its molecules formed by one M atom and x Si atoms, mass conservation condition requires the reaction

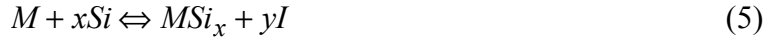


to hold. On the other hand, if the precipitate is strain-free, volume conservation condition requires the relation

$$\Omega_{MSi_x} = \Omega_M + (x + y)\Omega_{Si} \quad (4)$$

to hold. In Eq. (4) Ω_M and Ω_{Si} are respectively the atomic volumes of the M and Si atoms, and y is the misfit, with its value being either positive or negative. For M being a substitutional species $\Omega_M = \Omega_{Si}$, and for M being an interstitial species $\Omega_M = 0$. Upon continued growth or shrinkage of the precipitate, the misfit cannot be cumulative, since then the strain energy will be too large. In the following discussion, we let such misfits be relieved by point defects.

For the case assuming that the dominant point defect species is I, the combined mass and volume conservation condition requires



to hold. A consideration of the Gibbs free energy of the system in accordance with reaction (5) yields that, at the precipitate-matrix interface

$$C_M^* = C_M^{eq} \left(\frac{C_I}{C_I^{eq}} \right)^y \exp \left(\frac{2\Omega_{MSi_x}\sigma}{rk_B T} \right) \quad (6)$$

holds. In Eq. (6) C_M^{eq} is the thermal equilibrium concentration of M, C_I and C_I^{eq} are the actual and thermal equilibrium concentrations of I in the Si matrix, σ is the surface energy density of the precipitate-Si interface, r is the radius of the precipitate, k_B is Boltzmann's constant, and T is the absolute temperature. Now, with the C_I change rate given by

$$\frac{\partial C_I}{\partial t} = \frac{\partial}{\partial x} \left(D_I \frac{\partial C_I}{\partial x} \right) + y\rho \frac{4\pi r^2}{\Omega_{MSi_x}} \frac{dr}{dt}, \quad (7)$$

where D_I is the I diffusivity in Si, the I dominated case is modeled by Eqs. (1), (2), (6), and (7).

For the case assuming that the dominant point defect species is V, the combined mass and volume conservation condition requires



to hold. A consideration of the Gibbs free energy of the system in accordance with reaction (8) yields

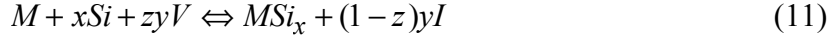
$$C_M^* = C_M^{eq} \left(\frac{C_V^{eq}}{C_V} \right)^y \exp \left(\frac{2\Omega_{MSi_x}\sigma}{rk_B T} \right), \quad (9)$$

where C_V and C_V^{eq} are the actual and thermal equilibrium concentrations of V in the Si matrix. With the C_V change rate given by

$$\frac{\partial C_V}{\partial t} = \frac{\partial}{\partial x} \left(D_V \frac{\partial C_V}{\partial x} \right) - y\rho \frac{4\pi r^2}{\Omega_{MSi_x}} \frac{dr}{dt}, \quad (10)$$

where D_V is the V diffusivity in Si, the V dominated case is modeled by Eqs. (1), (2), (9), and (10).

The most accurate approach is to assume that both I and V contribute to the relieve of the misfit, for with the combined mass and volume conservation condition requires the reaction



to hold. In reaction (11) z (with a value less than 1) is the fractional contribution of V to the relief of the misfit and $(1-z)$ is that of the I contribution. Using reaction (11) in the Gibbs free energy consideration of the system, we obtain

$$C_M^* = C_M^{eq} \left(\frac{C_I}{C_I^{eq}} \right)^{(1-z)y} \left(\frac{C_V}{C_V^{eq}} \right)^{zy} \exp \left(\frac{2\Omega_{MSi_x} \sigma}{rk_B T} \right). \quad (12)$$

For this case, the change rate of both C_I and C_V need to be considered, and they are given by

$$\frac{\partial C_I}{\partial t} = \frac{\partial}{\partial x} \left(D_I \frac{\partial C_I}{\partial x} \right) + (1-z)y\rho \frac{4\pi r^2}{\Omega_{MSi_x}} \frac{dr}{dt} + 4\pi r_{IV} (D_I + D_V) (C_I^{eq} C_V^{eq} - C_I C_V), \quad (13)$$

$$\frac{\partial C_V}{\partial t} = \frac{\partial}{\partial x} \left(D_V \frac{\partial C_V}{\partial x} \right) - zy\rho \frac{4\pi r^2}{\Omega_{MSi_x}} \frac{dr}{dt} + 4\pi r_{IV} (D_I + D_V) (C_I^{eq} C_V^{eq} - C_I C_V), \quad (14)$$

where r_{IV} is the reaction radius of I and V . Via the last terms on the right-hand-sides, Eqs. (13) and (14) have been written down assuming that the I and V recombination/generation reaction



where ϕ is a Si lattice atom, is operative. The I and V dynamical equilibrium condition according to reaction (15) is

$$C_I C_V = C_I^{eq} C_V^{eq}. \quad (16)$$

The assumption that Eq. (16) holds allow to continue the analysis of this case. First, Eq. (12) is now reduced to

$$C_M^* = C_M^{eq} \left(\frac{C_I}{C_I^{eq}} \right)^y \exp \left(\frac{2\Omega_{MSi_x} \sigma}{rk_B T} \right) = C_M^{eq} \left(\frac{C_V^{eq}}{C_V} \right)^y \exp \left(\frac{2\Omega_{MSi_x} \sigma}{rk_B T} \right) \quad (17)$$

Second, note that in Eqs. (12)-(14) z is not yet known, and we can now use Eqs. (13), (14), and (16) to obtain the value of z . Equation (16) yields

$$C_V \frac{\partial C_I}{\partial t} + C_I \frac{\partial C_V}{\partial t} = 0. \quad (18)$$

Assuming that Eq. (16) holds, then Eqs. (13) and (14) simplify to

$$\frac{\partial C_I}{\partial t} = \frac{\partial}{\partial x} \left(D_I \frac{\partial C_I}{\partial x} \right) + (1-z)y\rho \frac{4\pi r^2}{\Omega_{MSi_x}} \frac{dr}{dt}, \quad (19)$$

$$\frac{\partial C_V}{\partial t} = \frac{\partial}{\partial x} \left(D_V \frac{\partial C_V}{\partial x} \right) - zy\rho \frac{4\pi r^2}{\Omega_{MSi_x}} \frac{dr}{dt}. \quad (20)$$

Using Eqs. (18) and (19) in Eq. (17), we obtain

$$z = \frac{C_V}{C_I + C_V} + \frac{C_V \frac{\partial}{\partial x} \left(D_I \frac{\partial C_I}{\partial x} \right) + C_I \frac{\partial}{\partial x} \left(D_V \frac{\partial C_V}{\partial x} \right)}{y\rho \frac{4\pi r^2}{\Omega_{MSi_x}} (C_I + C_V) \frac{dr}{dt}}, \quad (21)$$

which is a function of t and x . The case can now be solved using Eqs. (1), (2), (17), (19), (20), and (21).

NUMERICAL SIMULATION RESULTS

The case of gettering precipitated Fe is of particular interest. Multicrystalline solar Si contain regions with high densities of dislocations and metal silicide precipitates, for which the minority carrier diffusion lengths are extremely low and cannot be improved by a normal gettering treatment using the Al-Si liquid layer at, e.g., 700°C for less than 2 h. Our results showed that the reason is that precipitate dissolution is involved which requires more extensive treatment.

As a test of the soundness of the model, numerical simulations were carried out for gettering Fe in Si wafers 200 μm in thickness by a 2 μm thick Al-Si liquid layer at the wafer backsurface (at the 200 μm position). It is assumed that Fe was introduced at 900°C to its solubility of $\sim 4.3 \times 10^{13} \text{ cm}^{-3}$ and then precipitated out to steady state at 700°C to the density of $\sim 10^{11} \text{ cm}^{-3}$. For the interstitial Fe atoms $\Omega_M = 0$ and for the FeSi₂ precipitates $\Omega_{MSi_x} = 1.85 \Omega_{Si}$,⁸ and hence $y = -0.15$ obtains from Eq. (4). The negative y value means that growth of FeSi₂ precipitates is associated with a volume contraction while their dissolution with a volume expansion. Thus, generation of V or consumption of I or both will result from FeSi₂ precipitate growth, while generation of I or consumption of V or both will result from FeSi₂ precipitate dissolution. Consequently, gettering will be faster if a V -supersaturation or I -undersaturation exists. Figure 1 shows two hypothetical cases for which $C_V = 10 C_V^{eq}$ holds for case (a) and $C_V = C_V^{eq}$ for case (b), and it is clear that gettering case (a) is more effective because of the V supersaturation. In Fig. 1 the Fe atom concentration in Si is plotted as a function of position in Si, and gettering time is used as a parameter. Another way of showing the results is to plot the total gettered Fe in the Al gettering layer and also to examine the precipitate size in Si, both as functions of gettering treatment time. First, Fig. 2 shows the result of the no volume misfit case, to be used to compare with results including the volume misfit. Fig. 3 shows the case assuming, at both Si surfaces, I concentration is either at thermal equilibrium value or undersaturated to 0.2 times of the thermal equilibrium value. As a function of gettering time, Fig. 3(a) shows the percentage of Fe depleted from the Si wafer by gettering into Al, while Fig. 3(b) shows the precipitate size change at the center of the wafer. Compared to the case of fig. 2, it is seen that the process of gettering is slowed down by

the volume misfit, but the gettering rate improves with V supersaturation at the Si wafer boundaries. Figure 4 shows a corresponding case for which V supersaturation is assumed instead, for which the V concentration at both boundaries are assumed to be either at thermal equilibrium or at 4 times of the thermal equilibrium values. Compared to Fig. 2, it is seen that the presence of the volume misfit slowed down the gettering process (“equilibrium” case, solid line), but with the presence of the Assumed V supersaturation, the gettering rate has essentially become that given by Fig. 2. Thus, gettering are more effective when the appropriate point defect supersaturation (V case, Fig. 4) or undersaturation (I case, Fig. 3) are present. For these calculations, the point defect thermal equilibrium concentration and diffusivity values are those given in the literature,⁹ as follows. For the I case, we used $C_I^{eq} = 5 \times 10^{30} \exp(-4.4eV/k_B T)$ cm⁻³ and $D_I = 10^{-5} \exp(-0.4eV/k_B T)$ cm²s⁻¹, which are believed to be accurate. For the V case, we used $C_V^{eq} = 2 \times 10^{23} \exp(-2eV/k_B T)$ cm⁻³ and $D_V = 0.1 \exp(-2eV/k_B T)$ cm²s⁻¹. It is noted that for while the product $D_V C_V^{eq}$ is fairly well known, individual values of D_V and C_V^{eq} are not, and the values used in obtaining Fig. 4 represent one extreme choice,⁹ while in the other extreme the V formation energy is ~ 3.5 eV and the V migration activation ion energy is ~ 0.5 eV, for which our results showed that the gettering process will be much slower than the case shown in Fig. 4.

CONCLUSIONS

In this study the effect of point defect injection on the dissolution process of metal silicide precipitate in Si has been modeled. Practically all metal silicide precipitate species possess volume misfits with the Si matrix material, and this is the basic reason why point defects are involved in the precipitate dissolution processes (as well as in the precipitate formation process). Fundamental equations describing such processes have been written. Using these equations, numerical simulations has been carried out for the case of FeSi₂, in which the role of point defects have been demonstrated: under the appropriate point defect super- or undersaturation conditions, precipitate will dissolve faster and hence speeding up the gettering process. This effect may be an important contributor to the highly effective optical gettering results obtained at NREL.¹⁰ Further works are needed for obtaining a more complete understanding on this subject.

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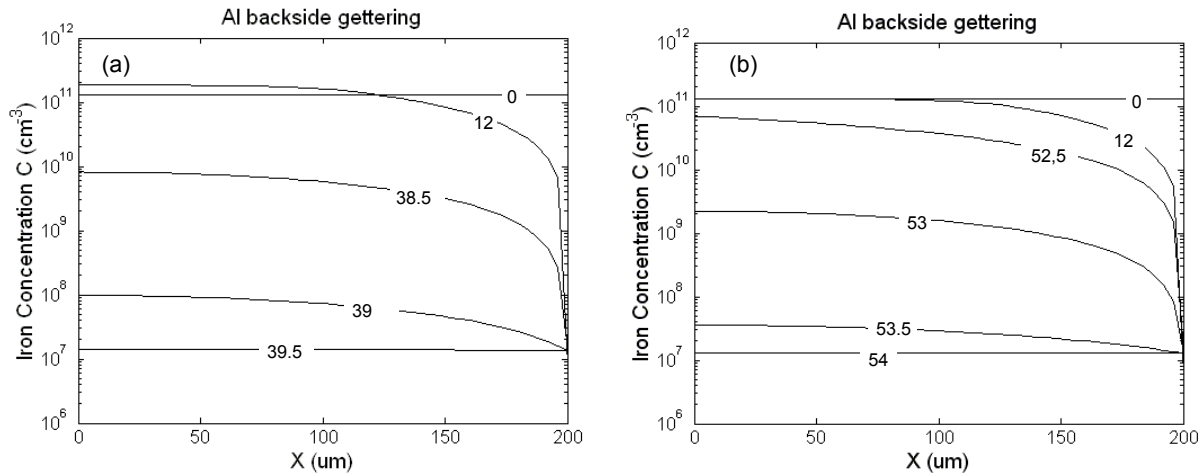


Fig. 1. Simulation results of gettering dissolved interstitial Fe and FeSi₂ precipitates from the Si matrix by an Al-Si liquid layer at the wafer backside (200 μm position). See text for initial and other experimental conditions. Plots (a) and (b) show the dissolved Fe concentrations at various gettering times in hrs at 700°C. It is seen that gettering is more effective in (a) than in (b), because in (a) a V supersaturation is assumed.

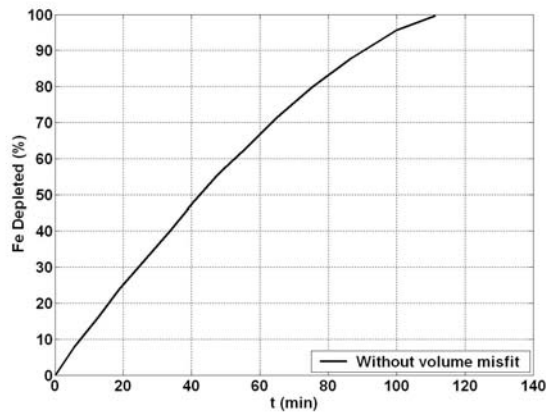


Fig. 2. Simulation results of gettering dissolved interstitial Fe and FeSi₂ precipitates from the Si matrix by an Al-Si liquid layer at the wafer backside (200 μm position). See text for initial and other experimental conditions. The plot shows the total % of Fe gettered into the Al layer as a function of time. In this case, it is assumed that the FeSi₂ precipitate is not associated with a volume misfit with the Si matrix.

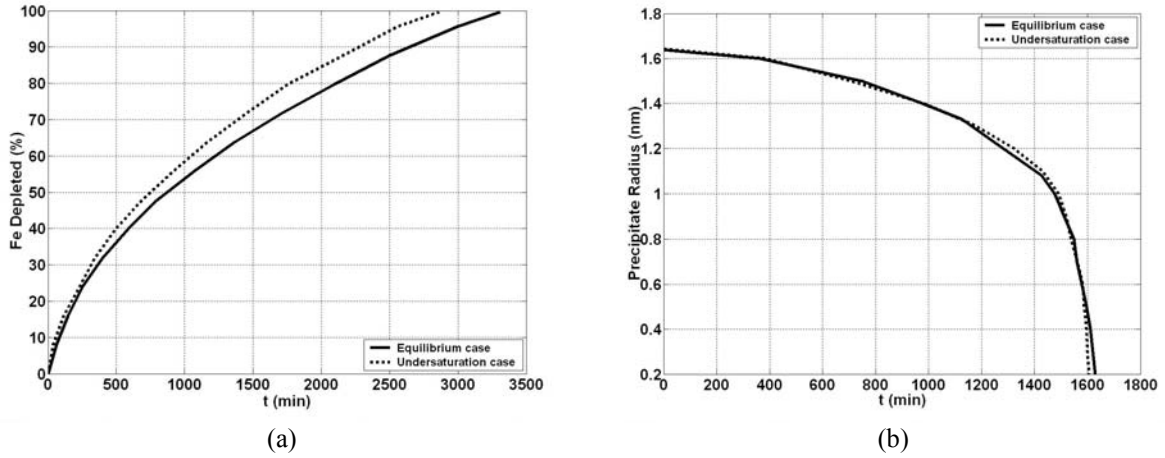


Fig. 3. Simulation results of gettering dissolved interstitial Fe and FeSi_2 precipitates from Si by an Al-Si liquid layer at the wafer backside ($200\ \mu\text{m}$ position). See text for initial and other experimental conditions. Plot (a) shows the percentage of Fe gettered to the Al layer at various gettering times in hrs at 700°C , under either the I thermal equilibrium assumption or I undersaturation assumption. Plot (b) shows the corresponding wafer center precipitate radius changes.

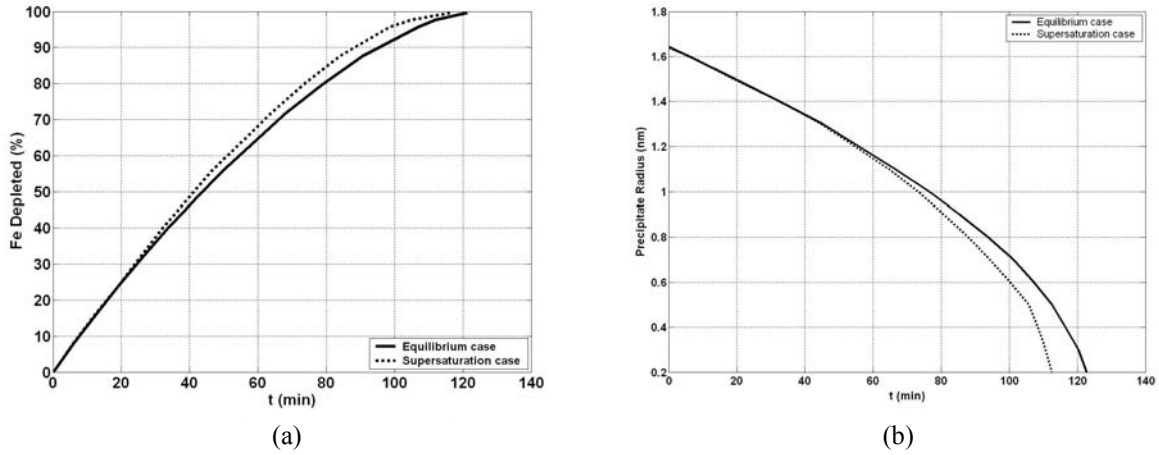


Fig. 4. Simulation results of gettering dissolved interstitial Fe and FeSi_2 precipitates from Si by an Al-Si liquid layer at the wafer backside ($200\ \mu\text{m}$ position). See text for initial and other experimental conditions. Plot (a) shows the percentage of Fe gettered to the Al layer at various gettering times in hrs at 700°C , under either the V thermal equilibrium assumption or V supersaturation assumption. Plot (b) shows the corresponding wafer center precipitate radius changes.

ABSTRACT

We have formulated the problem of gettering of metallic precipitates in Si for which there exists a volume misfit between the precipitate and the Si matrix material. The gettering process is modeled using an Al-Si liquid layer, and the volume misfit associated with dissolving the precipitates is assumed as accommodated by point defects, which may be dominated by vacancies (V), self-interstitials (I), or both contributing. Under the condition that V and I attained dynamical equilibrium, we found that for analytic purpose the problem reduces closely to either the V or the I alone case, with the fast diffusing one dominating. Our initial simulation results on gettering of FeSi_2 (with a misfit of -0.15) showed that the process can be sped up by the injection of V.

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