

Growth Kinetics of Refractory Metal Silicide under Radiation Induced Interstitial Mechanism: An Analytical Approach

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ABSTRACT

Refractory metal silicides are widely used in microelectronics and also important in hightemperature applications. This is due to their excellent thermal stability, high electrical conductivity, and mechanical strength. Conventional silicide formation typically occurs through thermal annealing. In this process, metal and silicon atoms interdiffuse. This interdiffusion occurs at elevated temperatures. However, under irradiation, atomic transport mechanisms are significantly altered. This change leads to enhanced diffusion. As a result, silicide formation can occur at lower temperatures. A theoretical model is developed to describe radiation-induced interstitial diffusion as the dominant mechanism during the formation of thin-film refractory metal silicide. Under irradiation, interstitial atoms are generated in both the metal and silicon layers. This process facilitates atomic transport. The metal interstitial atoms then diffuse toward the silicide/silicon interface through an interstitial mechanism. Similarly, silicon interstitial atoms migrate toward the metal/silicide interface. This also occurs via an interstitial mechanism. The silicide layer forms as a result of chemical reactions between metal and silicon interstitial atoms at these interfaces. The growth of the silicide layer follows a diffusion-limited rate governed by parabolic kinetics. The theoretical analysis indicates a distinct behaviour under irradiation. In such conditions, two interstitial atoms simultaneously contribute to silicide growth. This contrasts with non-irradiation conditions, where diffusion is typically governed by a single dominant atomic species. The effect is particularly pronounced in refractory metal silicides. The interstitial atomic densities were estimated from the model. These estimates correspond to both the irradiated refractory metal and silicon layers. The results indicate that silicide growth kinetics strongly depend on the defect generation rate in both layers. The influence of defect generation rate on silicide growth kinetics is significant. As the defect generation rate increases, the thickness of the silicide layer also increases. This enhancement is primarily due to the elevated interstitial densities resulting from higher defect generation rates in both irradiated layers. This effect is observed even at a low irradiation temperature. This temperature is significantly below the threshold required for silicide formation under non-irradiation conditions.

Keywords: Radiation induced interstitial, Refractory metal silicide, Diffusion-limited process, Interstitial diffusivity, Parabolic law

INTRODUCTION

In crystalline materials, irradiation induces structural modifications. This occurs through the generation of point defects. These defects significantly impact diffusion processes (Ustrzycka, Dominguez-Gutierrez., & Chromiński, 2024). When high-energy particles, such as neutrons or ions, interact with a solid, they transfer kinetic energy to the atoms in the crystal lattice. This energy transfer can displace atoms from their equilibrium positions. This displacement creates vacancies, or missing atoms. The displaced atoms are then placed into interstitial positions, forming interstitial defects. The simultaneous formation of a vacancy and an interstitial defect is known as a Frenkel pair (Balboa et al., 2018). The



defects creation of these follows а mechanism known as a collision cascade. In this process, initial energetic an displacement event is caused by the primary knock-on atom (PKA). This event initiates a series of atomic displacements. As a result, a localized region of disorder is formed. This leads to an increased concentration of interstitials and vacancies (Was, 2007).

Point defects such as vacancies and interstitials play a critical role in atomic diffusion. The formation energy of these defects depends on the crystal structure and material properties. In face-centered cubic (fcc) metals, for instance, elastic deformation models indicate that interstitials typically higher formation energies than have vacancies (Ma, & Dudarev, 2021). However, under irradiation, the continuous formation of Frenkel pairs sustains a non-equilibrium defect population, altering diffusion behaviour. Interstitials exhibit higher mobility than vacancies. This is due to their lower migration energy barriers. As a result, interstitial diffusion becomes a more efficient transport mechanism under certain conditions. (Zhang, 2024).

Diffusion processes play a crucial role in compound layer formation. These processes are often coupled with chemical reactions. This is especially important at the interfaces of immiscible substances. Diffusion allows atomic species to migrate from their respective bulk phases. At the interface, these atoms undergo chemical interactions that lead to the formation of new compounds.

Both irradiation and non-irradiation processes have been experimentally shown to influence compound layer formation. In many cases, the growth kinetics are governed by diffusion limited rate. (Jain & Agarwal, 2011; Murarka, 2012; Boussaa et al., 2005; Chakraborty et al., 2005; Agarwal et al., 2010).

However, the dominant diffusion mechanism under radiation-enhanced conditions remains under investigation. This is due to the complexity of defect-mediated transport. Multiple pathways contribute to atomic movement, making the mechanism difficult to isolate. Radiation-induced interstitial diffusion is considered as the dominant transport mechanism in irradiated metalsilicon systems. This dominance is particularly evident under conditions characterized by low defect generation rates. Under such conditions, the coupling between interstitial and vacancy fluxes becomes weaker. As a result, interstitials emerge as the primary diffusing species within the irradiated layers. Consequently, the growth rate of the silicide layer at the reaction interface is modeled as being proportional to the total flux driving its formation. To validate this hypothesis, we investigate the role of radiation-induced interstitials. This is done by analyzing their influence on silicide growth kinetics. Refractory metal silicides are used as a case study for this investigation.

METHOD AND MODELING

Suppose the coupling between interstitial and vacancy fluxes is weak. This assumption allows interstitial defects to be treated independently of vacancies. The assumption applies to an irradiated metal-silicon bilayer system. The metal layer occupies the region x < 0. The silicon layer extends over x > 0. After silicide formation, the silicide layer is located at x = h. Irradiation generates interstitial atoms in both the metal and silicon layers. Metal interstitial atoms migrate toward the metal/silicide interface via an interstitial mechanism. Similarly, silicon interstitial atoms move toward the silicide/silicon interface through the same process. At these interfaces, metal and silicon interstitial atoms chemically react to form the metal silicide layer.

The interstitial diffusivity of metal and silicon atoms under irradiation is denoted as $D_{m(si)}^{i,irr}$. This represents diffusion via the interstitial mechanism through the silicide layer.



Under irradiation, this diffusivity is described by an Arrhenius-type equation as follows:

$$D_m^{i,irr} = 0.1a^2 f_i \exp\left(\frac{-E_i^m}{K_B T}\right)$$
(1)
$$D_{si}^{i,irr} = 0.1a^2 f_i \exp\left(\frac{-E_i^m}{K_B T}\right)$$

where *a* is the lattice constant, f_i is the jump

frequency factor for interstitial atoms, E_i^m is

the interstitial migration energy, K_B is the Boltzmann constant, and T is the absolute temperature.

$$\frac{dn_{m}^{i}}{dt} = Kn_{m} - (K_{R} = 0) - \frac{n_{m}^{i}}{\tau_{m}}$$
(2)
$$\frac{dn_{si}^{i}}{dt} = Kn_{si} - (K_{R} = 0) - \frac{n_{si}^{i}}{\tau_{si}}$$

where K is the defect generation rate, K_R is the recombination rate, n_m , n_{si} , n_m^i , n_{si}^i , τ_m and τ_{si} are the lattice atomic densities, interstitial atomic densities, and the life times of interstitial atoms in metal and silicon-layers, respectively.

The relaxation of interstitial atoms distribution occurs faster than the growth of

 $Kn_m = \frac{n_m^i}{\tau_m} \quad \text{and} \quad Kn_{si} = \frac{n_{si}^i}{\tau_{si}} \quad (2^*)$ But $\tau_m = \frac{1}{\rho_{d_m} D_m^{i,irr}} \quad \text{and} \quad \tau_{si} = \frac{1}{\rho_{d_{si}} D_{si}^{i,irr}}$

After substituting τ_m and τ_{si} into Eqn. (2^{*}), rearrange the resulting expressions to make n_m^i and n_{si}^i the subjects of the new equations, respectively.

$$n_{m}^{i} = \frac{Kn_{m}}{\rho_{d_{m}} D_{m}^{i,irr}}$$

$$n_{si}^{i} = \frac{Kn_{si}}{\rho_{d_{si}} D_{si}^{i,irr}}$$
(3)

where ρ_{d_m} and $\rho_{d_{si}}$ are dislocation densities of metal and silicon respectively.

Equation (3) gives the expressions for the interstitial densities in the metal and silicon

layers under the radiation induced interstitial mechanism.

Neglecting transient diffusion, the Fick's equation for diffusion of interstitial atoms via interstitial mechanism under irradiation can be written as:

Neglecting the recombination between interstitial atoms and vacancies ($K_R = 0$), the changes in the densities of interstitial atoms and vacancies can be considered separately. The metal and silicon interstitial atomic densities in the silicide layer can then be obtained as follows:

metal silicide-layer (Akintunde and Selyshchev, 2016b). Therefore, it is assumed that interstitial atoms distribution is homogeneous and stationary inside the silicide layer i.e., $\frac{dn_m^i}{dt} = 0$ and $\frac{dn_{si}^i}{dt} = 0$ From Eqn. (2)

$$D_m^{i,irr} \frac{\partial^2 n^i(x)}{\partial x^2} = 0$$
$$D_{si}^{i,irr} \frac{\partial^2 n^i(x)}{\partial x^2} = 0$$

With corresponding boundary conditions:

 $J_m^i(x=0) = gn^i(x=0)$ at metal/silicide interface and $n^i(x=h) = n_{si}^i$ at silicide/silicon interface.

(4)

The flux of interstitial-atoms in metal layer is given by:

$$J_m^i(x=0) = -D_m^{i,irr}\frac{dn^i}{dx}$$
(5)

and

The flux of interstitial-atoms in silicon-layer is: $l^{i}(x = h) = -D^{i,irr}$ (6)

$$n^{i}(\mathbf{x}) = \mathbf{A}\mathbf{x} + \mathbf{B} \tag{7}$$

where A and B are integration constants and they are determined by boundary conditions as thus:

Using Eqn. (7), applying
$$n^{i}(x = h) = n_{si}^{i}$$

 $n_{si}^{i} = Ah + B$
 $B = n_{si}^{i} - Ah$ (8)

At x = 0, we apply the boundary condition for interstitial-atoms flux in metal-layer: $J_m^i(x = 0) = gn^i(x = 0)$

$$J_m^i(x=0) = gn$$
$$-D_m^{i,irr}A = gB$$

$$A = -\frac{gB}{D_m^{i,irr}}$$

 $\frac{\nu_m}{\text{Insert A into Eqn. (8)}}$

$$B = \frac{n_{si}^{l}}{1 - \frac{gh}{D_{m}^{l,irrr}}}$$

Insert A and B into Eqn. (7) $n^{i}(\mathbf{x}) = \frac{n_{si}^{i}}{D_{m}^{i,irr} - gh} \left(D_{m}^{i,irr} - gx \right)$ (9)

Equation (9) expresses the steady-state atomic density of diffusing interstitial atoms within the silicide layer.

The growth rate of silicide layer is described by:

$$\frac{dh}{dt} = VJ^{i}$$
(10)
where V is the volume of silicide layer, J^{i} is the total flux that drive the growth of silicide layer.

But

$$J^{i} = J^{i}_{m} + J^{i}_{si} = -D^{i,irr}_{m}A + (-D^{i,irr}_{si}A)$$
(11)
Insert A into Eqn. (11)
$$J^{i} = \frac{-gn^{i}_{si}}{D^{i,irr}_{m} - gh} (D^{i,irr}_{m} + D^{i,irr}_{si})$$



Substitute for
$$J^{i}$$
 in Eqn. (10)

$$\frac{dh}{dt} = \frac{gn_{si}^{i}V}{(gh - D_{m}^{i,irr})} \left(D_{m}^{i,irr} + D_{si}^{i,irr} \right)$$
Re-arrange and integrate

$$\int_{0}^{h} (gh - D_{m}^{i,irr}) dh = \int_{0}^{t} gn_{si}^{i}V \left(D_{m}^{i,irr} + D_{si}^{i,irr} \right) dt$$

$$\frac{1}{2}gh^{2} - D_{m}^{i,irr}h = gn_{si}^{i}V \left(D_{m}^{i,irr} + D_{si}^{i,irr} \right) t$$

$$h^{2} - \frac{2D_{m}^{i,irr}}{g}h = 2n_{si}^{i}Vt \left(D_{m}^{i,irr} + D_{si}^{i,irr} \right)$$
(12)
In accordance with parabolic law Eqn. (12) can be approxim

In accordance with parabolic law Eqn. (12) can be approximated as: $h^2 \approx 2n_{si}^i Vt(D_m^{i,irr} + D_{si}^{i,irr})$ (13)

RESULTS AND DISCUSSION

The growth kinetics of the silicide layer, as described by Eqn. (13), is diffusioncontrolled, with the layer thickness showing a parabolic dependence on time. This equation is consistent with experimental observations. Experimental studies (Chakraborty et al. 2005; Agarwal et al. 2010; Murarka, 2012; Chen, 2005; Jain and Agarwal, 2011) have shown that the growth kinetics of refractory metal silicide is diffusion-limited. This applies to both irradiation and non-irradiation conditions. The interfacial-controlled stage is difficult to identify experimentally due to its short duration. For this reason, we did not account for the interfacial-controlled growth rate in our study. Similarly, the critical thickness marking the transition from interfaciallimited to diffusion-limited growth was not considered. Experimental studies on the refractory metal silicide investigated here (Jain and Agarwal, 2011) have exclusively reported parabolic growth kinetics, further validating our model's agreement with experiment.

The experimental approach employed in the literature (Chakraborty et al. 2005; Agarwal et al. 2010; Arranz and Palacio, 2009; Jain and Agarwal, 2011) is ion beam mixing technique. Ion beam mixing (IBM) is a technique used for silicide formation at metal-silicon interfaces. High-energy ion irradiation induces intermixing of the metal and silicon layers. This process depends on parameters such as ion dose rate, ion energy, and substrate temperature. We did not consider the IBM approach because it does not take into account the independent contribution of point defects in the silicide formation.

Radiation-enhanced diffusion (RED) approach was taken into account under the irradiation of metal-silicon bilayer system. This approach facilitates silicide growth by generating a large number of point defects, such as interstitials, which enhance the transport of metal and silicon atoms through the lattice sites to the reaction interface. Unlike IBM, RED does not directly induce intermixing but instead accelerates diffusiondriven phase formation, similar to nonirradiation technique but at an enhanced rate due to radiation-induced defect generation (Akintunde and Selyshchev, 2016b).

Thin films of molybdenum and tungsten disilicides chosen are due to their exceptional electrical and metallurgical properties, as reported in the literature (Chen, The stable phases-molybdenum 2005). disilicide (MoSi₂) and tungsten disilicide (WSi₂)—are considered at low temperatures under irradiation conditions. Under nonconditions. irradiation where thermal diffusion dominates, silicon atoms are the primary diffusing species in both MoSi2 and WSi₂ phases (Murarka, 2012). In this study, we develop a model that accounts for the





contributions of both metal and silicon atoms as interstitial species during silicide formation under radiation-induced interstitial diffusion. The metal and silicon layers are irradiated at different defect generation rates $(K = 10^{-7}, 10^{-8}, \text{ and } 10^{-9} \text{dpa/s}).$

The interstitial diffusivity of silicon in MoSi₂ and WSi₂ is estimated using Eqn. (1) with the following parameters:

a = 5.43 x 10⁻¹⁰ m, f_i = 5 x 10¹² s⁻¹ (Smallman, 2016), E_i^m = 0.18 eV (Freysoldt et al., 2014), K_B = 8.617 x10⁻⁵ eV/atom, and T = 298 K.

Similarly, the interstitial diffusivity of tungsten and molybdenum is estimated using the same equation. The parameters for tungsten are $E_i^m = 0.022$ eV and a = 3.165×10^{-10} m, while for molybdenum, they are

 $E_i^m = 0.013$ eV and a = 3.150×10^{-10} m. All other parameters remain the same as those used for silicon. The resulting interstitial diffusivities for silicon, tungsten, and molybdenum are 1.3×10^{-10} , 2.13×10^{-8} , and 3.00×10^{-8} m²/s, respectively.

Both metal and silicon atoms are identified diffusing species. Their as interstitial diffusivities are higher than the interdiffusion coefficient of molybdenum and tungsten disilicides. This comparison is made with the interdiffusion coefficient under non-irradiation conditions. (Paul, 2019).

Using Eqn. (3), the interstitial metal and silicon atomic densities are determined. These calculations are based on the data in Table 1, with the results presented in Tables 2 and 3.

Table	1:	Useful	data	on	tungsten	and	moly	/bdenum	disilicides	and	their	component	atomic
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Lattice atomic density $n_{m(si)}$ (10 ²⁸ atoms/m ³)	Diffusing species under non irradiation	Diffusing species under irradiation	Volume of silicide V (10 ⁻²⁹ m ³)	Dislocation density (m ⁻²)
Mo, (6.45)	MoSi ₂ , (Si) Murarka, (2012)	MoSi ₂ , (Mo, Si) [Present work]	MoSi ₂ , (4.1)	Si, (10 ⁸) Wang, (2024)
Si, (5.00)	WSi ₂ , (Si) Murarka, (2012)	WSi ₂ , (W, Si) [Present work]	WSi ₂ , (4.3)	Mo, (3 x 10 ¹²)
W, (6.30)	× /			W, (3 x 10 ¹²)

Table 2: Estimation of interstitial tungsten and silicon atomic densities at different defect generation rates during tungsten disilicide formation.

K (dpa/s)	n_{m}^{i} (10 ¹⁶ m ⁻³)	$n_{si}^{i} (10^{23} \mathrm{m}^{-3})$	
10-7	9.86	3.76	
10 ⁻⁸	0.99	0.38	
10-9	0.10	0.04	

Table 3: Estimation of interstitial molybdenum and silicon atomic densities at different defect generation rates during molybdenum disilicide formation.

K (dpa/s)	n _m ⁱ (10 ¹⁶ m ⁻³)	$n_{si}^{i} (10^{23} \mathrm{m}^{-3})$	
10-7	7.17	7.52	
10-8	0.72	0.75	
10-9	0.07	0.08	





Table 2 shows that the interstitial atomic density of molybdenum increases with the defect generation rate in the molybdenum layer. This indicates a strong dependence on defect production. Similarly, Table 3 presents the interstitial atomic density of disilicide at different defect tungsten generation rates. The trends observed are consistent with those found for molybdenum disilicide.



Figure 1: The growth kinetics of molybdenum disilicilde (MoSi₂) at different defect generation rates $K = 10^{-7}$, 10^{-8} , and 10^{-9} dpa/s at 298K

Figures 1 and 2 illustrate that the layer thickness increases with defect generation rate in the irradiated layers. Additionally, both figures show a parabolic dependence of layer thickness on time. The growth kinetics of MoSi2 is diffusion-limited, as shown in Figure 1. Similarly, the growth kinetics of WSi₂ follows a parabolic law, as depicted in Figure 2. The results presented in Figures 1 2 show good agreement and with experimental observations reported by (Jain and Agarwal, 2011; Agarwal et al., 2010). These studies showed a parabolic time dependence in the growth of refractory metal silicide thickness.

At room temperature, the thermal energy alone is insufficient to drive the formation of tungsten and molybdenum disilicides in the absence of radiation. This is because the

Using the data from Tables 1-3 and the estimated interstitial diffusivities. we calculate the thickness of the molybdenum tungsten disilicide layers. and This calculation is done at different defect generation rates $(K = 10^{-7}, 10^{-8}, \text{ and }$ 10^{-9} dpa/s) using Eqn. (13). We then plot the silicide thickness as a function of time. Each graph corresponds to a specific defect generation rate, with results depicted in Figures 1 and 2.



Figure 2: The growth kinetics of tungsten disilicilde (WSi₂) at $K = 10^{-7}$, 10^{-8} , and 10^{-9} dpa/s at room temperature (298K).

required formation temperatures-798 K for MoSi₂ and 923 K for WSi₂ (Akintunde and Selvshchev, 2016a; Lytovchenko, 2016) under non-irradiation conditions—are significantly higher. At room temperature, the available thermal energy cannot overcome the covalent bonds in silicon or the metallic bonds in refractory metals. Consequently, it is incapable of facilitating the necessary diffusion of silicon and metal atoms to the reaction interface via thermal vacancies for silicide formation (Akintunde, Selyshchev, and Kehinde, 2025).

However, experimental studies in the literature (Jain and Agarwal, 2011; Rachid, 2019; Boussaa et al., 2005) confirm the feasibility of silicide thin-film formation at temperatures as low as room temperature, and even lower under irradiation. This



occurs because irradiation generates a high concentration of interstitial atoms in the metal and silicon layers. These interstitials facilitate atomic movement to the reaction interface via an interstitial mechanism. As a result, silicide is formed, particularly at very low irradiation temperatures.

CONCLUSION

The theoretical approach presented in this study shows the feasibility of producing thin films of refractory metal silicides through radiation-induced interstitial diffusion. This process occurs at low irradiation temperatures. Molybdenum and tungsten disilicides are used as case studies. The results indicate a diffusion-limited growth The silicide layer follows mechanism. parabolic growth kinetics, with its thickness varying as a parabolic function of time. Both metal and silicon atoms act as diffusing species, based on their estimated interstitial diffusivities. The required irradiation temperature for silicide formation is significantly lower than that under nonirradiation conditions. This confirms that the growth is driven by interstitial diffusion. Radiation-induced interstitial atoms in the metal and silicon lavers facilitate this diffusion process. Thermal energy at room temperature is not sufficient to overcome the strong covalent and metallic bonding in silicon and refractory metals. In contrast, radiation energy breaks these bonds and enables atomic transport.

Finally, the results show that the growth kinetics of refractory metal disilicides are strongly enhanced by increased defect generation rates.

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