Analysis of Electron Transport Coefficients in Binary Mixtures of TEOS Gas with Kr, Xe, He and Ne Gases for Using in Plasma Assisted Thin-film Deposition

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Abstract – The electron transport coefficients in not only pure atoms and molecules but also in the binary gas mixtures are necessary, especially on understanding quantitatively plasma phenomena and ionized gases. Electron transport coefficients (electron drift velocity, density-normalized longitudinal diffusion coefficient, and density-normalized effective ionization coefficient) in binary mixtures of TEOS gas with buffer gases such as Kr, Xe, He, and Ne gases, therefore, was analyzed and calculated by a two-term approximation of the Boltzmann equation in the E/N range (ratio of the electric field E to the neutral number density N) of 0.1 - 1000 Td (1 Td = 10^{-17} V.cm²). These binary gas mixtures can be considered to use as the silicon sources in many industrial applications depending on mixture ratio and particular application of gas, especially on plasma assisted thin-film deposition.

Keywords: TEOS, thin-film, Electron transport coefficient, Plasma assisted thin-film deposition, Boltzmann equation.

1. Introduction

Tetraethoxysilane (TEOS), Si(OC₂H₅)₄, which is a gas with non-toxic and non-explosive characteristics; and is mainly used as the silicon source to deposit SiO₂-like films by plasma-enhanced chemical vapor deposition (PECVD) in remote microwave oxygen plasma reactors because the deposited films show good step coverage compared with films deposited from SiH₄ [1]. A simple model for the oxygen rich O₂ and TEOS gas discharge plasmas was constructed [2, 3]. The electron transport coefficients in TEOS-O₂ and TEOS-Ar mixture gases were calculated and measured [4]. The binary mixtures of TEOS-N₂ have been also used to instead of SiH₄-based gas mixtures to reduce the dangers of explosion and toxicity while improving quality of the deposition of SiO₂ by PECVD. Recently, the electron transport coefficients in TEOS-N₂ mixture gases were also calculated [5]. On the other hand, electron transport coefficients in the mixture gases are necessary data to obtain the plasma models.

For all of above reasons along with the lack of measured electron transport coefficients in the binary mixtures of TEOS gas with buffer gases such as Kr, Xe, He and Ne gases, in this study, we have calculated and analysed electron drift velocity, density-normalized longitudinal diffusion coefficient, ratio of longitudinal diffusion coefficient to electron mobility, Townsend first ionization coefficient using a two-term approximation of the

Boltzmann equation over the wide E/N range (ratio of the electric field E to the number density of neutral atoms or molecules N) and the reliable electron collision cross sections of the TEOS, Kr, Xe, He and Ne gases. These results are the first calculations and the best available so far for the quantitative modeling of plasma discharge for processing procedures using these binary mixture gases.

2. Analysis

We used the electron swarm method to determine a set of accurate electron collision cross sections for TEOS, TMS, and BF₃ molecules [4, 6, 7]. The two-term approximation of the Boltzmann equation for the energy has been applied to calculate the electron transport coefficients in pure gases and binary mixture gases such as electron drift velocity, density-normalized longitudinal diffusion coefficient, ratio of longitudinal diffusion coefficient to electron mobility, Townsend first ionization coefficient over the wide E/N range. The electron transport coefficients calculated from the present set of TEOS gas and binary mixtures of TEOS gas with Ar and O₂ gases are agreement with those in experiments [4]. The sets of electron collision cross sections for Kr, Xe, He and Ne gases were slightly modified to obtain the accurate sets of electron collision cross sections. The electron transport coefficients calculated by using the present sets of these gases are also compared and good agreement with those in experiments.

In the present study, the electron collision cross sections for TEOS molecule determined by Tuan and Jeon [4]. This

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set includes one momentum transfer, one attachment, two vibrational excitation cross sections (threshold energies of 0.16 and 0.37 eV), one dissociation excitation (threshold energy of 3.6 eV), one electronic excitation (threshold energy of 1.7 eV), and one total ionization (threshold energy of 10.6 eV). The set of electron collision cross sections for Kr atom determined by Hayashi [10] includes one momentum transfer, fourteen electronic excitation cross sections (threshold energies of 9.915 to 13.437 eV), and one total ionization (threshold energy of 14.0 eV) cross section. The set of electron collision cross sections for Xe atom determined by Hashimoto and Nakamura [11] includes one momentum transfer, fourteen electronic excitation cross sections (threshold energies of 8.315 to 11.58 eV), and one total ionization (threshold energy of 12.13 eV) cross section. The set of electron collision cross sections for He atom determined by Hayashi [10] have been used as initial sets. This set includes one momentum transfer, twenty five electronic excitation cross sections (threshold energies of 19.82 to 24.19 eV), and one total ionization (threshold energy of 24.59 eV) cross section. The set of electron collision cross sections for Ne atom determined by Hayashi [10] have been used as initial sets. This set includes one momentum transfer, seven electronic excitation cross sections (threshold energies of 16.62 to 19.66 eV), and one total ionization (threshold energy of 21.56 eV) cross section. A brief summary of several reaction processes of TEOS molecule and other gases for plasma modeling is listed in Table 1.

The present two-term approximation of the Boltzmann

equation for the energy given by Tagashira *et al.* [12] was also previously used for the pure gases and binary mixture gases [4-9]. The centre-of-mass drift velocity of an electron swarm (W_r) and the longitudinal diffusion coefficient (D_L) are calculated in the time-of-flight (TOF) parameters. The diffusion-corrected ionization and attachment coefficients are also calculated in the steady state Townsend (SST) parameters. A description of the Boltzmann equation analysis in the previous publications [4-9, 12-16] is also briefly following represented.

The electron transport coefficients in given gases are functions only of the ratio E/N , the gas temperature T , and when a magnetic field is present, of B/N . They are related to the electron collision cross sections by complex integral expressions involving the electron energy distribution function (EEDF) [13]. The electron energy distribution function (EEDF), $f(\epsilon, E/N)$, is normalized by

$$\int_0^{\infty} f(\epsilon, E/N) d\epsilon \cong 1 \quad (1)$$

where ϵ is the electron energy. Electron transport properties directly depend on EEDF [13]. The EEDF is normally non-Maxwellian in the non-equilibrium plasma used for applications. The EEDF can be obtained theoretically by solving the Boltzmann Eq. [13]

$$\frac{\partial f}{\partial t} + \bar{v} \nabla_r f + \bar{a} \nabla_v f = \left(\frac{\partial f}{\partial t} \right)_{coll} \quad (2)$$

Table 1. Reaction processes with threshold energy for plasma modeling in atoms and molecules

Types of collision	Reactions	Threshold energies (eV)	References and comments
* In TEOS molecule			
Momentum transfer	$\text{Si}(\text{OC}_2\text{H}_5)_4 + e \rightarrow \text{Si}(\text{OC}_2\text{H}_5)_4 + e$	0	Ref. 4
Vibrational excitation	$\text{Si}(\text{OC}_2\text{H}_5)_4 + e \rightarrow \text{Si}(\text{OC}_2\text{H}_5)_4(v=1) + e$	0.16	Ref. 4
Vibrational excitation	$\text{Si}(\text{OC}_2\text{H}_5)_4 + e \rightarrow \text{Si}(\text{OC}_2\text{H}_5)_4(v=2) + e$	0.37	Ref. 4
Electronic excitation	$\text{Si}(\text{OC}_2\text{H}_5)_4 + e \rightarrow \text{Si}(\text{OC}_2\text{H}_5)_4^* + e$	1.7	Ref. 4
Dissociation excitation	$\text{Si}(\text{OC}_2\text{H}_5)_4 + e \rightarrow \text{Si}(\text{OC}_2\text{H}_5)_4^* + e$	3.6	Refs. 4 and 17
Ionization	$\text{Si}(\text{OC}_2\text{H}_5)_4 + e \rightarrow \text{Si}(\text{OC}_2\text{H}_5)_4^+ + e$	10.6	Refs. 4, 17, and 18
* In Kr atom			
Momentum transfer	$\text{Kr} + e \rightarrow \text{Kr} + e$	0	Ref. 10
Electronic excitation	$\text{Kr} + e \rightarrow \text{Kr}^* + e$	9.915 ÷ 13.437	Ref. 10, includes fourteen electronic excitations
Ionization	$\text{Kr} + e \rightarrow \text{Kr}^+ + e$	14.0	Ref. 10
* In Xe atom			
Momentum transfer	$\text{Xe} + e \rightarrow \text{Xe} + e$	0	Ref. 11
Electronic excitation	$\text{Xe} + e \rightarrow \text{Xe}^* + e$	8.315 ÷ 11.58	Ref. 11, includes fourteen electronic excitations
Ionization	$\text{Xe} + e \rightarrow \text{Xe}^+ + e$	12.13	Ref. 11
* In He atom			
Momentum transfer	$\text{He} + e \rightarrow \text{He} + e$	0	Ref. 10
Electronic excitation	$\text{He} + e \rightarrow \text{He}^* + e$	19.82 ÷ 24.19	Ref. 10, includes twenty five electronic excitations
Ionization	$\text{He} + e \rightarrow \text{He}^+ + e$	24.59	Ref. 10
* In Ne atom			
Momentum transfer	$\text{Ne} + e \rightarrow \text{Ne} + e$	0	Ref. 10
Electronic excitation	$\text{Ne} + e \rightarrow \text{Ne}^* + e$	16.62 ÷ 19.66	Ref. 10, includes seven electronic excitations
Ionization	$\text{Ne} + e \rightarrow \text{Ne}^+ + e$	21.56	Ref. 10

where $f = f(\vec{r}, \vec{v}, t)$ is the distribution function of the positions $\vec{r}$ and velocities of electrons $\vec{v}$, $\vec{a}$ is the acceleration due to external forces, and $(\partial f / \partial t)_{\text{coll}}$ is the collision induced rate of change of the number of electrons per unit volume of phase space. The solution normally involves the assumptions that the fields are time independent, that spatial gradient terms can be omitted, and that the distribution in velocity space is only slightly disturbed from spherical.

The electron drift velocity calculated from the solution of electron energy distribution function, $f(\varepsilon, E/N)$, of the Boltzmann equation is defined as

$$W = -\frac{1}{3} \left(\frac{2}{m} \right)^{1/2} \frac{eE}{N} \int_0^\infty \frac{\varepsilon}{q_m(\varepsilon)} \frac{df(\varepsilon, E/N)}{d\varepsilon} d\varepsilon \quad (3)$$

where m is the electron mass, e is the elementary charge and $q_m(\varepsilon)$ is the momentum-transfer cross section.

The density-normalized longitudinal diffusion coefficient is defined as

$$ND_L = \frac{V_1}{3N} \left(E \int_0^\infty \frac{\varepsilon}{q_T} \frac{\partial}{\partial \varepsilon} (F_1 \varepsilon^{-1/2}) d\varepsilon + \int_0^\infty \frac{\varepsilon^{1/2}}{q_T} F_0 d\varepsilon \right) - (\varpi_0 A_2 - \varpi_1 A_1 - \varpi_02). \quad (4)$$

where V_1 is the speed of electron, q_T is the total cross section, here F_n and ϖ_n ($n = 0, 1, 2$) are respectively the electron energy distributions of various orders and their eigen values. V_1 , ϖ_n , ϖ_{0n} , and A_n are given by

$$V_1 = \left(\frac{2e}{m} \right)^{1/2} \quad (4.1)$$

$$\varpi_0 = V_1 N \int_0^\infty \varepsilon^{1/2} q_i F_0 d\varepsilon \quad (4.2)$$

$$\varpi_1 = -\frac{V_1 E}{3N} \int_0^\infty \frac{\varepsilon}{q_T} \frac{\partial}{\partial \varepsilon} (F_0 \varepsilon^{-1/2}) d\varepsilon + (\varpi_0 A_1 - \varpi_{01}) \quad (4.3)$$

$$\varpi_{0n} = V_1 N \int_0^\infty \varepsilon^{1/2} q_i F_n d\varepsilon \quad (4.4)$$

$$A_n = \int_0^\infty F_n d\varepsilon \quad (4.5)$$

where q_i is the ionization cross section.

The Townsend first ionization coefficient is defined as

$$\alpha / N = \frac{1}{W} \left(\frac{2}{m} \right)^{1/2} \int_I^\infty f(\varepsilon, E/N) \varepsilon^{1/2} q_i(\varepsilon) d\varepsilon \quad (4.6)$$

where I is the ionization onset energy and $q_i(\varepsilon)$ is the ionization cross section.

3. Electron transport coefficients

The electron drift velocities, W , the density-normalized longitudinal diffusion coefficients, ND_L , the ratio of the longitudinal diffusion coefficients to the electron mobility, D_L/μ , the Townsend first ionization coefficients, α/N , as functions of E/N for the binary mixtures of TEOS gas with other gases such as Kr, Xe, He and Ne have been calculated in the E/N range $0.1 < E/N < 1000$ Td by a two-term approximation of the Boltzmann equation for energies.

The calculated electron drift velocities in TEOS-Kr, TEOS-Xe, TEOS-He, and TEOS-Ne mixture gases are shown in Figs. 1-4. In Figs. 1 and 2, the W values in TEOS-Kr and TEOS-Xe mixture gases are suggested to be between those of the pure gases over the range of $E/N > 100$ Td. In the range of $E/N < 100$ Td, the W values in these binary mixtures are higher than those of Kr, Xe, and TEOS gases, especially on $E/N = 10$ Td corresponding to electron energy of about 0.202 eV. To the best of our

Fig. 1. Electron drift velocity, W , as functions of E/N for the TEOS-Kr mixtures with 10%, 30%, 50%, 70%, and 90% TEOS. The solid curves show present W values calculated for the pure Kr atom and TEOS molecule.

Fig. 2. Electron drift velocity, W , as functions of E/N for the TEOS-Xe mixtures with 10%, 30%, 50%, 70%, and 90% TEOS. The solid curves show present W values calculated for the pure Xe atom and TEOS molecule.

Fig. 3. Electron drift velocity, W , as functions of E/N for the TEOS-He mixtures with 10%, 30%, 50%, 70%, and 90% TEOS. The solid curves show present W values calculated for the pure He atom and TEOS molecule.

Fig. 4. Electron drift velocity, W , as functions of E/N for the TEOS-Ne mixtures with 10%, 30%, 50%, 70%, and 90% TEOS. The solid curves show present W values calculated for the pure Ne atom and TEOS molecule.

Fig. 5. Density-normalized longitudinal diffusion coefficient, ND_L , as functions of E/N for the TEOS-Kr mixtures with 10%, 30%, 50%, 70%, and 90% TEOS. The solid curves show present ND_L values calculated for the pure TEOS molecule and Kr atom.

Fig. 6. Density-normalized longitudinal diffusion coefficient, ND_L , as functions of E/N for the TEOS-Xe mixtures with 10%, 30%, 50%, 70%, and 90% TEOS. The solid curves show present ND_L values calculated for the pure TEOS molecule and Xe atom.

Fig. 7. Density-normalized longitudinal diffusion coefficient, ND_L , as functions of E/N for the TEOS-He mixtures with 10%, 30%, 50%, 70%, and 90% TEOS. The solid curves show present ND_L values calculated for the pure TEOS molecule and He atom.

Fig. 8. Density-normalized longitudinal diffusion coefficient, ND_L , as functions of E/N for the TEOS-Ne mixtures with 10%, 30%, 50%, 70%, and 90% TEOS. The solid curves show present ND_L values calculated for the pure TEOS molecule and Ne atom.

knowledge, the electron drift velocity strongly depends on momentum transfer cross sections and vibrational excitation cross sections. In these cases, the reasons could be suggested that at this energy the vibrational excitations in TEOS gas occurred, the momentum transfer cross section of TEOS gas is higher than that of Kr and Xe gases and the momentum transfer cross sections of Kr and Xe gases are deeply decreasing to the minimum points. Therefore, these curves have the same tendency as that of pure TEOS gas. For TEOS-He and TEOS-Ne mixture gases in Figs. 2 and 4, the W values are suggested to be between those of the pure gases over the all range of E/N . However, the W values of 10% TEOS-He and 10% TEOS-Ne mixtures are higher than those of pure TEOS, He, and Ne gases in the range of $E/N < 20$ Td (corresponding to electron energy of about

0.202 eV of about 0.15 eV). In these cases, the reasons could be suggested that at this energy the momentum transfer cross section of TEOS gas is higher than that of He and Ne gases and the momentum transfer cross sections of He and Ne gases are slightly increasing.

The calculated density-normalized longitudinal diffusion coefficients and calculated ratio of the longitudinal diffusion coefficients to the electron mobility in TEOS-Kr, TEOS-Xe, TEOS-He, and TEOS-Ne mixture gases are shown in Figs. 5-8 and Figs. 9-12, respectively. The calculated values of these coefficients in the binary mixtures of TEOS with Kr, Xe, He, and Ne gases are suggested to be between those of the pure gases over the all range of E/N .

The calculated Townsend first ionization coefficients in TEOS-Kr, TEOS-Xe, TEOS-He, and TEOS-Ne mixture

Fig. 9. Ratios of longitudinal diffusion coefficient to the electron mobility, D_L/μ , as functions of E/N for the TEOS-Kr mixtures with 10%, 30%, 50%, 70%, and 90% TEOS. The solid curves show present D_L/μ values calculated for the pure TEOS molecule and Kr atom.

Fig. 10. Ratios of longitudinal diffusion coefficient to the electron mobility, D_L/μ , as functions of E/N for the TEOS-Xe mixtures with 10%, 30%, 50%, 70%, and 90% TEOS. The solid curves show present D_L/μ values calculated for the pure TEOS molecule and Xe atom.

Fig. 11. Ratios of longitudinal diffusion coefficient to the electron mobility, D_L/μ , as functions of E/N for the TEOS-He mixtures with 10%, 30%, 50%, 70%, and 90% TEOS. The solid curves show present D_L/μ values calculated for the pure TEOS molecule and He atom.

Fig. 12. Ratios of longitudinal diffusion coefficient to the electron mobility, D_L/μ , as functions of E/N for the TEOS-Ne mixtures with 10%, 30%, 50%, 70%, and 90% TEOS. The solid curves show present D_L/μ values calculated for the pure TEOS molecule and Ne atom.

gases are shown in Figs. 13-16. In Figs. 13 and 14, the α/N values in TEOS-Kr and TEOS-Xe mixture gases are suggested to be between those of the pure gases over the ranges of $E/N < 200$ Td and $E/N < 700$ Td, respectively. The α/N values in 10% TEOS-Kr and 10% TEOS-Xe gas mixtures are closed to those of pure Kr and Xe gases. These values could be greater than those of pure Kr and Xe gases over the ranges of $E/N > 200$ Td and $E/N > 700$ Td, respectively.

To the best of our knowledge, the Townsend first ionization coefficient strongly depends on ionization cross sections and dissociation excitation cross sections. The reasons could be suggested that the ionization and dissociation excitation cross sections of TEOS gas are much higher than those of Kr and Xe gases. For TEOS-He and TEOS-Ne mixture gases in Figs. 15 and 16, the α/N values are suggested to be between those of the pure gases

over the all range of E/N . However, the α/N values of 10% TEOS-He and 10% TEOS-Ne mixtures are higher than those of pure TEOS, He, and Ne gases over the all range of E/N . In these cases, the reasons could be suggested that ionization cross sections of He and Ne gases are much lower than that of TEOS gas.

The present results, that have been also calculated for the first time, are reliable over the E/N range of 0.1 to 1000 Td because of the accuracy of the electron collision cross sections for the present gases and the validity of the Boltzmann equation. More experiments of the electron transport coefficients for these binary mixtures need to be performed over the wide range of E/N in the future. From ionization coefficients of these binary mixtures, we could make the source of plasma corresponding equivalent industrial applications in manufacturing nano-material, depending on mixture ratio and particular application of

Fig. 13. The Townsend first ionization coefficients, α/N , as functions of E/N for the TEOS-Kr mixtures with 10%, 30%, 50%, 70%, and 90% TEOS. The solid curves show present α/N values calculated for the pure TEOS and Kr molecules.

Fig. 14. The Townsend first ionization coefficients, α/N , as functions of E/N for the TEOS-Xe mixtures with 10%, 30%, 50%, 70%, and 90% TEOS. The solid curves show present α/N values calculated for the pure TEOS and Xe molecules.

Fig. 15. The Townsend first ionization coefficients, α/N , as functions of E/N for the TEOS-He mixtures with 10%, 30%, 50%, 70%, and 90% TEOS. The solid curves show present α/N values calculated for the pure TEOS and He molecules.

Fig. 16. The Townsend first ionization coefficients, α/N , as functions of E/N for the TEOS-Ne mixtures with 10%, 30%, 50%, 70%, and 90% TEOS. The solid curves show present α/N values calculated for the pure TEOS and Ne molecules.

gas, especially on plasma assisted thin-film deposition. Especially on models for the binary mixture gases of TEOS gas with high concentration of buffer gases such as Kr, Xe, He, and Ne should be experimented in gas discharge plasma conditions in order to reduce the dangers of explosion and toxicity while improving quality of the deposition of SiO_2 .

4. Conclusions

The electron transport coefficients (electron drift velocity, density-normalized longitudinal diffusion coefficient, and density-normalized effective ionization coefficient) in binary mixtures of TEOS gas with buffer gases such as Kr, Xe, He, and Ne gases was calculated for the first time and analyzed by a two-term approximation of the Boltzmann equation in the E/N range of 0.1 - 1000 Td. Models of gas discharge plasmas could be constructed using the binary mixture gases of TEOS gas with high concentration of buffer gases such as Kr, Xe, He, and Ne. These binary gas mixtures can be considered to use as the silicon sources in many industrial applications depending on mixture ratio and particular application of gas, especially on plasma assisted thin-film deposition.

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