International Journal of Applied Mathematics
Volume 39 No. 1, 2026
ISSN: 1311-1728 (printed version); ISSN: 1314-8060 (on-line version)

SEMI-QUANTITATIVE ANALYSIS ON OPTICAL OXIDE THIN FILMS USING CS COMPLEX SECONDARY IONS IN TOF-SIMS

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Abstract

This study investigates the capabilities of "Cs_nM^+" and "Cluster Counting" methods for semi-quantitative analysis of optical oxide thin films in TOF-SIMS, including SiO_2 thin film, Ta_2O_5-SiO_2 multilayer thin film and different ratios of HfO_2-SiO_2 mixed thin films systems. Quantitative XPS results of HfO2-SiO2 mixed thin films were obtained to compare with SIMS results. The study confirms that cluster counting of Cs complexes yields significantly better estimates the matrix composition than the "Cs_nM^+" approach in optical thin films when ignoring other trace elements and contaminants. Different element RSF and surface Cs coverage on each layer should be taken into consideration when applying "MCs_n^+" and "Cluster Counting" methods on any multilayer optical thin films.

Key words: SIMS, Optical thin films, Semi-quantitative analysis

1. Introduction

Time-of-Flight Secondary Ion Mass Spectrometry (TOF-SIMS) is a surface analysis technique with high mass and spatial resolution, high sensitivity and low limit of detection (LOD) at ppm level[1], making it a powerful tool for depth profiling analysis[2-6] and the detection of trace elements[7,8]. However, the capability of quantitative analysis in TOF-SIMS has long been hindered by "Matrix Effect". An element's secondary ion yields are influenced by not only the abundance but also its surrounding sample chemistry, namely, the sample matrix[1]. This phenomenon makes quantitative analysis in TOF-SIMS rather difficult. Normally, the use of "ions implantation"[9] or "standard sample calibration"[10] method will compensate matrix effect and make it possible to quantify the elements interested. But these methods not

Received: 13 Aug 2025

International Journal of Applied Mathematics
Volume 39 No. 1, 2026
ISSN: 1311-1728 (printed version); ISSN: 1314-8060 (on-line version)

only require standard materials that consist of homogeneous matrix-matched materials, with the same matrix as the samples of interest and whose elements composition are known but also face limitations at thin film interfaces.

Solutions to the problem of matrix effect have been reported in early researches. Among these solutions, "Cs_nM^+" and "Cluster Counting" techniques allow to reduce matrix effects. "Cs_nM^+" technique measures Cs_nM^+ molecular ions, the combination of a neutral atom M that is detached from the surface and Cs_n^+(n=1, 2) ions formed in the sputtering process. Because of the high Cs_n^+ ions yield, under steady-state the flux of Cs_nM^+ ions will reflect the atomic concentration of the element M[11]. This method has been proven effective in III-V semiconductors[12-14] with high precision but has limitations in some other types of materials where tangible matrix effects can be observed[15,16]. "Cluster Counting" is an evolution method based on "Cs_nM^+" method. In this method, all Cs complex cluster ions that consist of the interested element M will be taken into account instead of only counting Cs_nM^+ ions. This type of data processing has been confirmed to better estimates the matrix composition than the "Cs_nM^+" approach[17,18]. To our knowledge, no systematic researches were conducted on dielectrics like optical glasses and thin films based on these 2 methods. Therefore, in this article we conducted a series of experiments to investigate the possibility of using Cs_nM^+ and cluster counting methods on optical elements and compare the differences between these 2 methods.

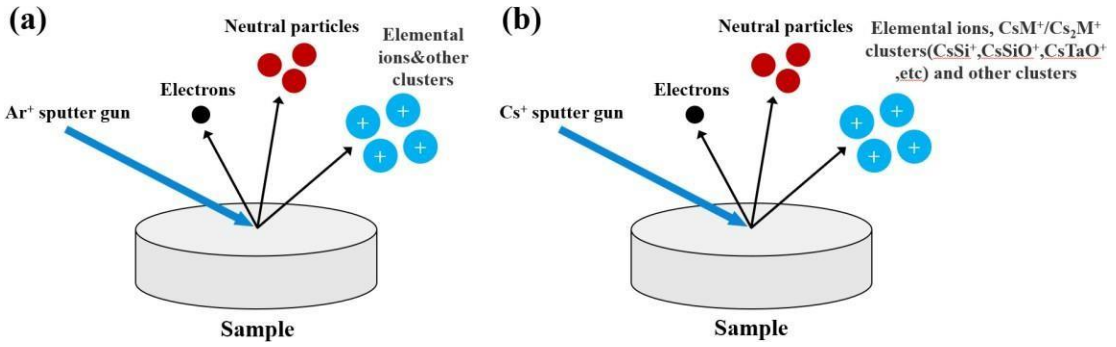

Fig 1. Possible ions and clusters when applying Ar^+ and Cs^+ sputter guns under positive ion mode.

2. Experimental

2.1 Sample preparation

7 different ratios of HfO2/SiO2 mixed thin films were deposited on 30mm-SiO2 substrates by dual e-beam evaporation with a base pressure of 9×10^{-6} mbar and a deposition pressure of 2×10^{-4} mbar in the vacuum chamber. The temperature was set to 200°C during the coating process. The designed values of HfO2 in all 7 thin films are: 100%, 83%, 66%, 50%, 33%, 17%, 0%. The true values were measured using XPS. Besides, SiO2 thin film and SiO2-Ta2O5-SiO2 oxide multilayer samples were provided by Shanghai Institute of Optics and Fine Mechanics (SIOM).

Received: 13 Aug 2025

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Volume 39 No. 1, 2026
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2.2 Methods

2.2.1 Theory

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The detailed explanation of "Cluster Counting" theory was discussed in reference 16 and 17. In short, for an interested element *ei*, the secondary ion intensity *Ie* is computed as the linear combination for the intensities I_{sj} of all Cs clusters

$$I_{ei} \approx \sum_{j=1}^{N_S} n_{ij} I_{sj} \quad (2\text{-}1)$$

Where N_S is the total number of Cs cluster species, n_{ij} is the number of atoms of element e_i in the species s_j. In this article, trace elements, contaminants will be ignored due to the fact that only main elements in the sample matrix were analyzed. The aim is to improve MCs_n^+ method by including previously ignored Cs clusters.

2.2.2 Instruments

The XPS measurements on HfO_2-SiO_2 mixed thin films were conducted on K-Alpha(Thermo Fisher Scientific Inc.,USA) with a monochromatic Al Kα X-ray source(400μm diameter, photon energy of 1486.6eV). Surface etching was performed using 1 keV Ar ions with a scanned area of 4×2 mm². The pass energy was set to 50 eV. Data acquisition was carried out using the Advantage software provided by the equipment manufacturer.

The TOF-SIMS measurements were carried out using Nano-TOF II(ULVAC-PHI Inc., Japan). Sputtering was performed using Ar^+ sputter gun(Ar-3kV-100nA) and Cs^+ sputter gun(Cs-2kV-100nA/Cs-1kV-50nA)with 600×600μm^2 sputtering area and 45° incidence angle. A 30keV-Bi_3^{++} liquid metal ion gun(LMIG) with 100×100μm^2 analyzing area in the center of the sputter crater and 45° incidence angle was used for depth profiles acquisition. In addition, ion-neutralized and electron-neutralized guns were activated for charge compensation on the sample surface under positive ion mode. The pressure of the vacuum chamber during the experiments was lower than 5×10^{-8}mbar. Data acquisition was carried out using the TOF-DR software.

3. Results and discussion

3.1 Semi-quantitative abilities in SiO_2 thin film and multilayer thin film

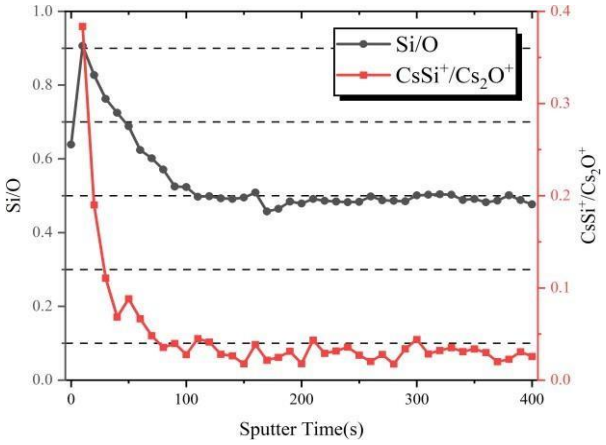
In this section, we fisrt assessed the capabilities of MCs_n^+ and Cluster Counting Method in SiO_2 thin film. For "Cs_nM^+" method, only $^{133}Cs^{28}Si^+$ and $^{133}Cs_2{}^{16}O^+$ were analyzed; For "Cluster Counting" method, Table 1. shows the clusters analyzed in this study. All data The Si/O ratio by using equation (2-1) and $CsSi^+/Cs_2O^+$ ratio was calculated and the results are

Received: 13 Aug 2025

International Journal of Applied Mathematics
Volume 39 No. 1, 2026
ISSN: 1311-1728 (printed version); ISSN: 1314-8060 (on-line version)

shown in Fig.2.

Fig 2. Si/O and $CsSi^+/Cs2O^+$ ratio changes over sputter time using Cs-1kV-50nA sputter



gun(positive ion mode)

As shown in the figure, both Si/O and $CsSi^+/Cs2O^+$ ratio first show fluctuation due to surface contamination and surface concentration of Cs, then gradually decrease until each reaches a steady value after approximate 150s of sputtering. The steady average values for Si/O and $CsSi^+/Cs2O^+$ ratio are 0.489 and 0.0297 respectively. "Cluster Counting" method shows a close Si/O ratio to the theoretical Si/O ratio of pure $SiO2$ thin film (0.5) with 2.2% deviation, where "$CsnM^+$" method seems to show a big Si/O ratio deviation when not applying relative sensitivity factors (RSFs). This could suggest that for $SiO2$ thin films, all analyzed clusters in table. 1 could be seen as having RSFs of 1.

To further investigate capabilities of "Cluster Counting" method in different matrices, $SiO2$-$Ta2O5$-$SiO2$ multilayer thin film was applied to compare the ion ratio differences in different layers. For $Ta2O5$ layer, $CsTa^+$, $CsTaO^+$ and $CsTaO2^+$were used to characterize the overall Ta intensity. The intensity counts, Si/O and Ta/O ratio results are shown in Fig 3.

Table. 1 Clusters analyzed in $SiO2$ thin films

$^{133}Cs^{16}O$	$^{133}Cs^{28}Si$
$^{133}Cs^{28}Si^{16}O$	^{16}O $^{133}Cs2$
$^{28}Si^{16}O$ $^{133}Cs2$	$^{28}Si^{16}O$ $^{133}Cs2$ 2
$^{28}Si^{16}O$ $^{133}Cs2$ 3	

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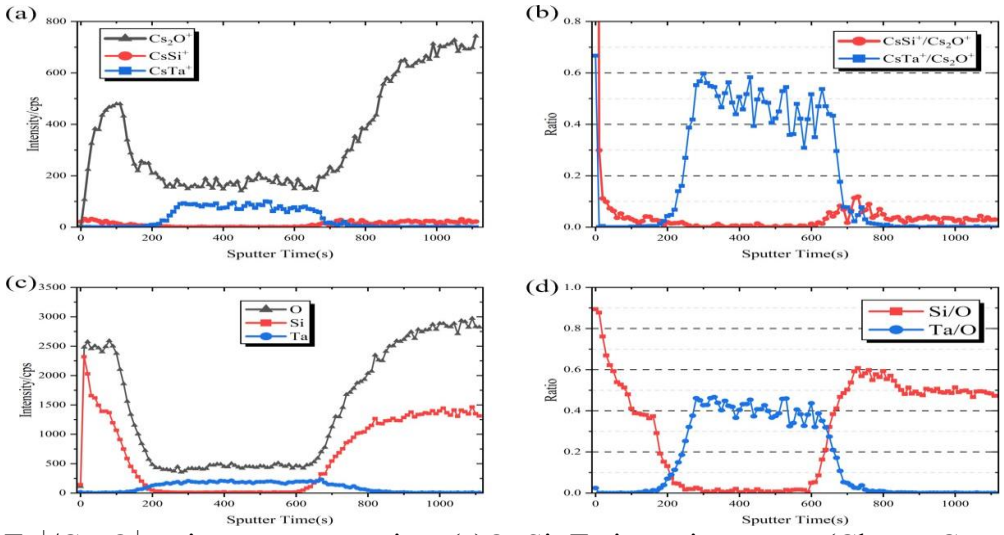
Fig 3. (a)Cs_2O^+, $CsTa^+$ and $CsSi^+$ intensity counts over sputter time; (b) $CsSi^+/Cs_2O^+$ and



$CsTa^+/Cs_2O^+$ ratio over sputter time (c)O, Si, Ta intensity counts (Cluster Counting Method) over sputter time; (d)Si/O and Ta/O ratio over sputter time (using Cs-1kV- 50nA sputter gun & positive ion mode)

Fig 3(a) shows a considerable deviation to the theoretical ratio in both SiO_2 and Ta_2O_5 layer, with 0., Fig 3(c) clearly shows the significant ion intensity difference in 479 CsTa/Cs_2O and 0.0312 CsSi/Cs_2O ratio(Fig.3(b)). In Fig 3(c), both Si/O and Ta/O ratio in each layer show similar variations to the previous SiO_2 thin film results. The steady average values of Ta/O and Si/O after 100s of sputtering in each layer is 0.413 and 0.487, and the deviations to the theoretical ratio are 3.25% and 2.6% respectively(Fig 3(d)). Besides 2 different layers. This phenomenal was observed previously where high surface concentration of Cs will change the surface chemistry and decrease Cs^+ and CsM^+ ion yield[18]. To evaluate instantaneous surface Cs coverage, both Cs^+ and Cs^+_2 ions were measured. Suppose that a Cs^+_2 cluster is formed by Cs and Cs^+ recombination or association:

$$Cs + Cs^+ \rightarrow Cs_2^+ \qquad (3\text{-}1)$$

Then the $I(Cs_2^+)$ could be written as the product of instantaneous surface Cs coverage C_{Cs} and the flux of Cs^+ ions J_{Cs}:

$$I(Cs_2^+) = C_{Cs} J_{Cs} \qquad (3\text{-}2)$$

Or

$$C_{Cs} \propto \frac{I(Cs_2^+)}{I(Cs^+)} \qquad (3\text{-}3)$$

$$J_{Cs} \qquad I(Cs^+)$$

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Where $I(Cs^+)$ represents instantaneous surface Cs coverage. If we assume a statistical distribution of surface Cs atoms, instantaneous surface Cs coverage should be proportional to $I(Cs2^+)/I(Cs^+)$ ratio. The $I(Cs2^+)/I(Cs^+)$ ratio for multilayer thin film is shown in Fig 4.

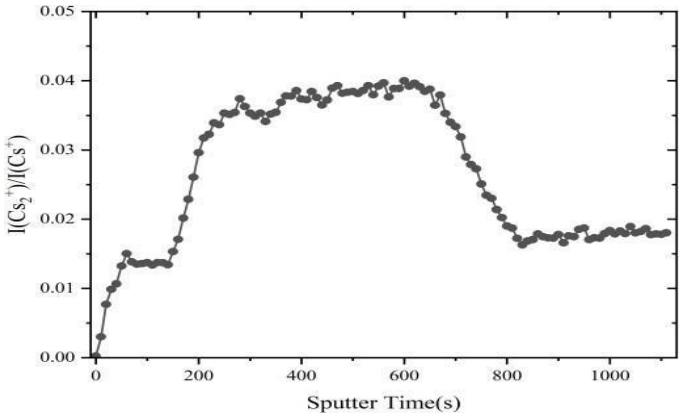

Fig 4. $I(Cs2^+)/I(Cs^+)$ change over sputter time.

The result in Fig 4. shows a significant Cs coverage difference in 2 layers, which could explain the low ion yield in $Ta2O5$ layer. This suggests that apart from element RSFs, different Cs coverage in different layer(or can be seen as layer RSFs) should be taken in to consideration if semi-quantitative analysis in multilayer samples needs to be made. In conclusion, "$CsnM^+$" and "Cluster counting" methods are used in $SiO2$ thin film and multilayer thin film to investigate the quantitative abilities of these 2 methods. For $SiO2$ and $Ta2O5$ thin films, direct semi-quantitative analysis can be made by using "Cluster counting" method. But different Cs coverage should be monitored when profiling multilayer samples.

3.2 Semi-quantitative abilities in $HfO2$-$SiO2$ mixed thin films

In this section, we first investigate the SIMS response for various $HfO2$-$SiO2$ mix films second ions under different sputter conditions. Every normalized intensity and intensity ratio are the mean value after 40 cycles of sputtering. Table 2 shows the actual abundance of Hf and Si in every mixed thin film.

Table.2 XPS results of the composition in 7 mixed thin films

Sample No.	Si(at. %)		Hf(at. %)
	Designed value	Measured abundance	Measured abundance
1	100%	100.0%	0.0%
2	83%	83.2%	16.8%
3	67%	68.4%	31.6%
4	50%	58.0%	42.0%
5	33%	38.0%	62.0%
6	17%	13.0%	87.0%

Received: 13 Aug 2025

International Journal of Applied Mathematics
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ISSN: 1311-1728 (printed version); ISSN: 1314-8060 (on-line version)

7	0%	0%	100%

When considering monatomic ions, matrix effects are present for Hf^+ ions under 3 sputter conditions. The results in Fig 5. shows that the dependence of the normalized

intensity of Hf^+ ions versus Hf abundance is not linear. For Hf^+ ions under Ar-3kV- 100nA bombardment, the normalized intensity of Hf^+ secondary ions increases almost linearly with Hf abundance until 31.6 at. % and drops 42 at. % and increases nonlinearly and finally drops at 100 at. %. Similar results are observed when using Cs-1kV-50nA sputter gun. When using Cs-2kV-100nA sputter gun, the normalized intensity of Hf^+ ions seems to increase linearly until 83.2% and then drops like other sputter conditions. Under such conditions, Hf quantification in HfO_2-SiO_2 mixed thin films could still be possible but probably with a large measurement error.

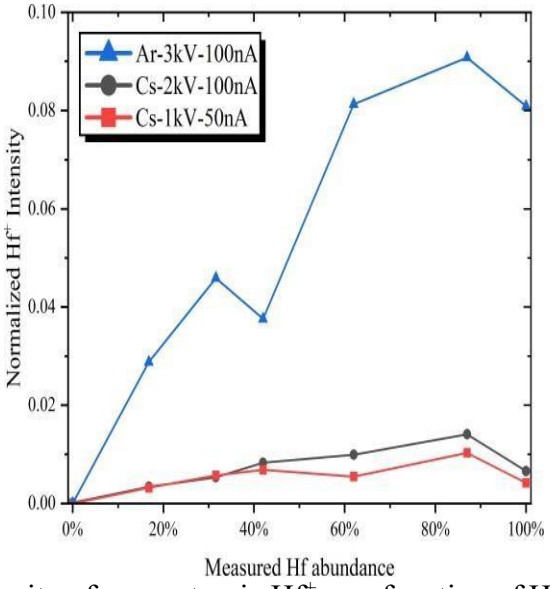

Fig.5 Normalized intensity of monoatomic Hf^+ as a function of Hf abundance under 3 different conditions. (Positive ion mode)

The normalized intensities of polyatomic Hf-based secondary ion $CsHf^+$ with the Hf^+ abundance in the thin films under different sputter conditions are presented in Fig. 6(a). When considering $CsHf^+$ ions, no major differences between 2 sputter conditions are found. The results show that although the variation of normalized intensity is univocal, the intensity increase is still not linear, matrix effects are still detected. Besides, severe matrix effects are detected when considering $CsSi^+$ secondary ions(Fig.6(b)). When Si abundance increases, the $CsSi^+$ signal increases nonlinearly to 58.0% Si abundance and then decreases for higher Si abundance. The similar result was reported by Mikami at al.[14] who observed that in $Si_{1-x}Ge_x$ semiconductor CsSi+ normalized intensity increased until about x=0.6 and then decreased after x=0.6. Both figures reveal that for HfO_2-SiO_2 mixed thin films, matrix effect

cannot be ignored when using Cs_nM^+ method.

Received: 13 Aug 2025

International Journal of Applied Mathematics
Volume 39 No. 1, 2026
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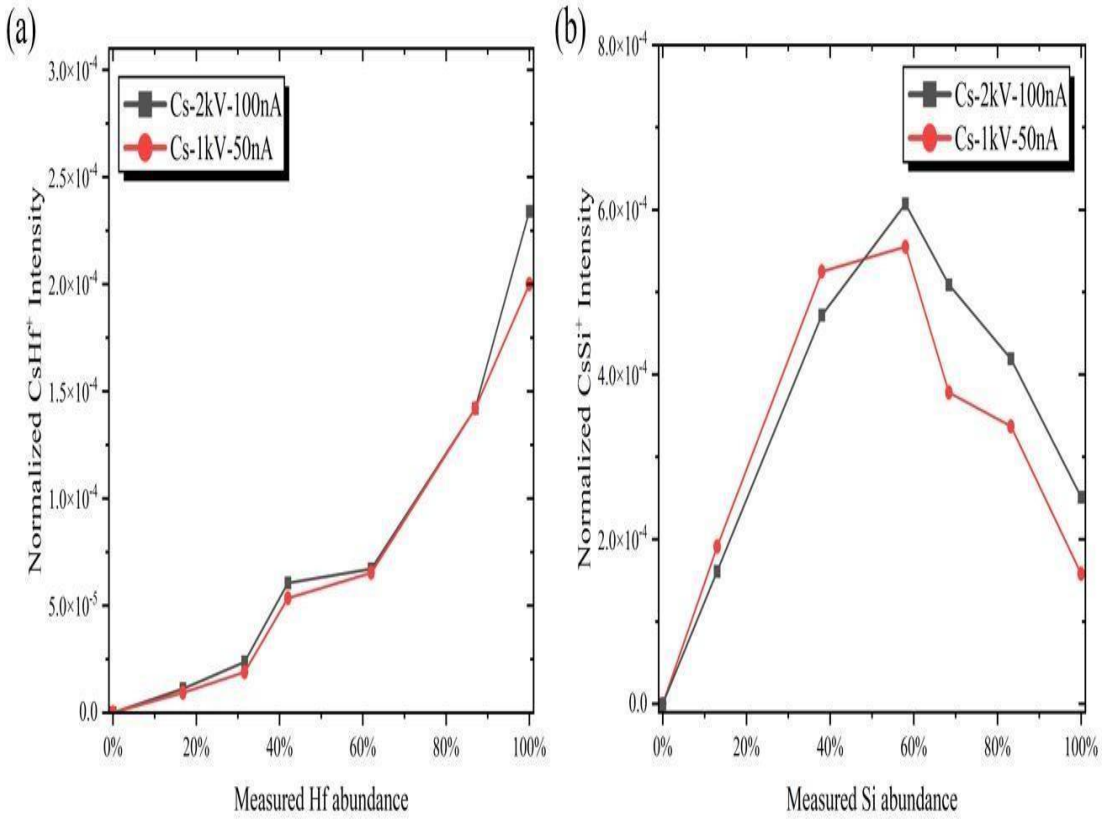
Fig.6 Normalized intensities of (a)$CsHf^+$ and (b) $CsSi^+$ as a function of Hf or Si



abundance under 2 different sputter conditions. (Positive ion mode) However, by adopting a calibration method based on the linear dependence of $I(CsHf^+)/I(CsSi^+)$ ratio as a function of Si/Hf ratio, which is similar to the method used in semiconductor samples[13,14], matrix effects can be depressed. Normally, RSFs are needed to compensate matrix effects and achieve higher precision. The RSF values are determined by the equation below:

$$C_m = RSF_m \cdot I_m \qquad (3\text{-}4)$$

$$\frac{C}{n} = \frac{RSF}{n} \cdot \frac{I}{n}$$

Where C and I represent mean abundance(determined by XPS analysis) and mean intensity(determined by SIMS depth profiles). The subscripts ‘m’ and ‘n’ refer to the element interested.

In this study, eq. (3-4) can be written as:

$$\frac{I_{CsSi}}{I_{CsHf}} = k \cdot \frac{C_{Si}}{C_{Hf}} \qquad (3\text{-}5)$$

Received: 13 Aug 2025

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Where $I(CsSi^+)/I(CsHf^+)$ represents the intensity ratio of $CsSi^+$ and $CsHf^+$ clusters, CSi/CHf represents the measured Si:Hf ratio, k represents the line slope. The results are shown in Fig.7. Linear correlations between $I(CsSi^+)/I(CsHf^+)$ and CSi/CHf can be obtained. Linear fittings give good fits with R^2 value of 0.970, k value of 7.286 under Cs-2kV-100nA sputter condition and R^2 value of 0.973, k value of 6.844 under Cs-1kV- 50nA sputter condition.

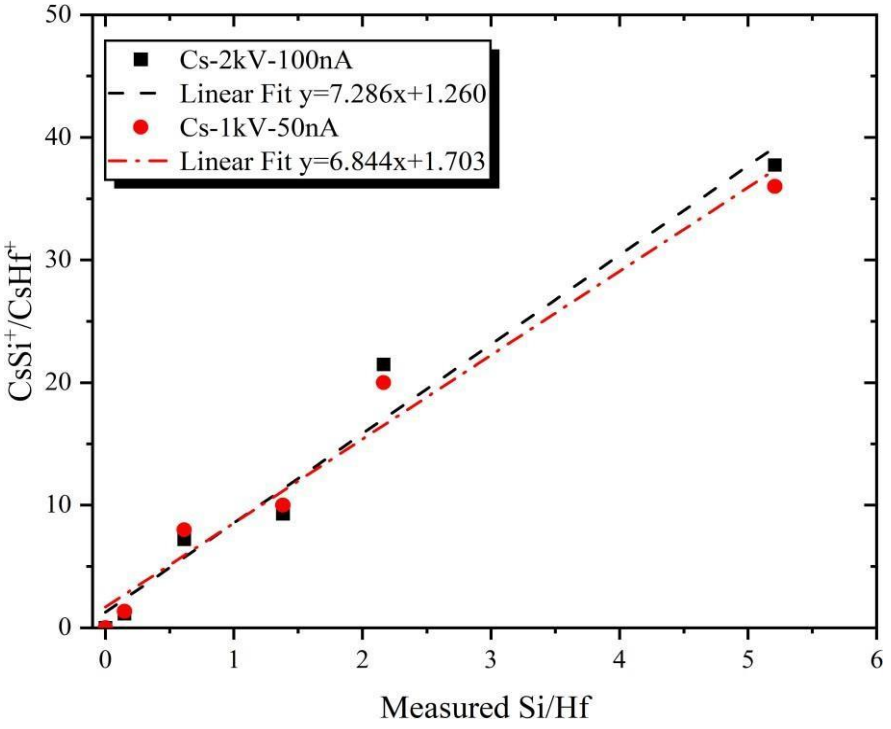

Fig.7 Intensity ratio $I(CsSi^+)/I(CsHf^+)$ as a function of the measured ratio Si/Hf under different sputter conditions.

For the last part of the study, "Cluster Counting" method was evaluated using mixed thin films. In this part, $CsHf^+$, $CsHfO^+$ and $CsHfO2^+$ clusters were used to characterize the overall Hf intensity. Overall Si intensity was not under research due to the peak overlap($^{133}Cs^{28}Si^{16}O^+$ and $^{177}Hf^+$.On our instrument these 2 species are not discernible on the mass spectra). The relation between Hf abundance and normalized Hf intensity and the comparison between "Cs_nM^+" and "Cluster Counting" method are shown in Fig 8.

Obviously, for "Cs_nM^+" method, severe matrix effects are present in Fig. 8(a) and (c), with R^2 of 0.917 and 0.882 respectively. For "Cluster Counting" method, linear correlations between normalized Hf intensity and measured Hf abundance can be found with R^2 of 0.988 and 0.980 in Fig. 8(a) and (c) respectively. This proves that "Cluster Counting" method could reduce matrix effects and achieve better element detection precisions than traditional "Cs_nM^+" method.

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Volume 39 No. 1, 2026
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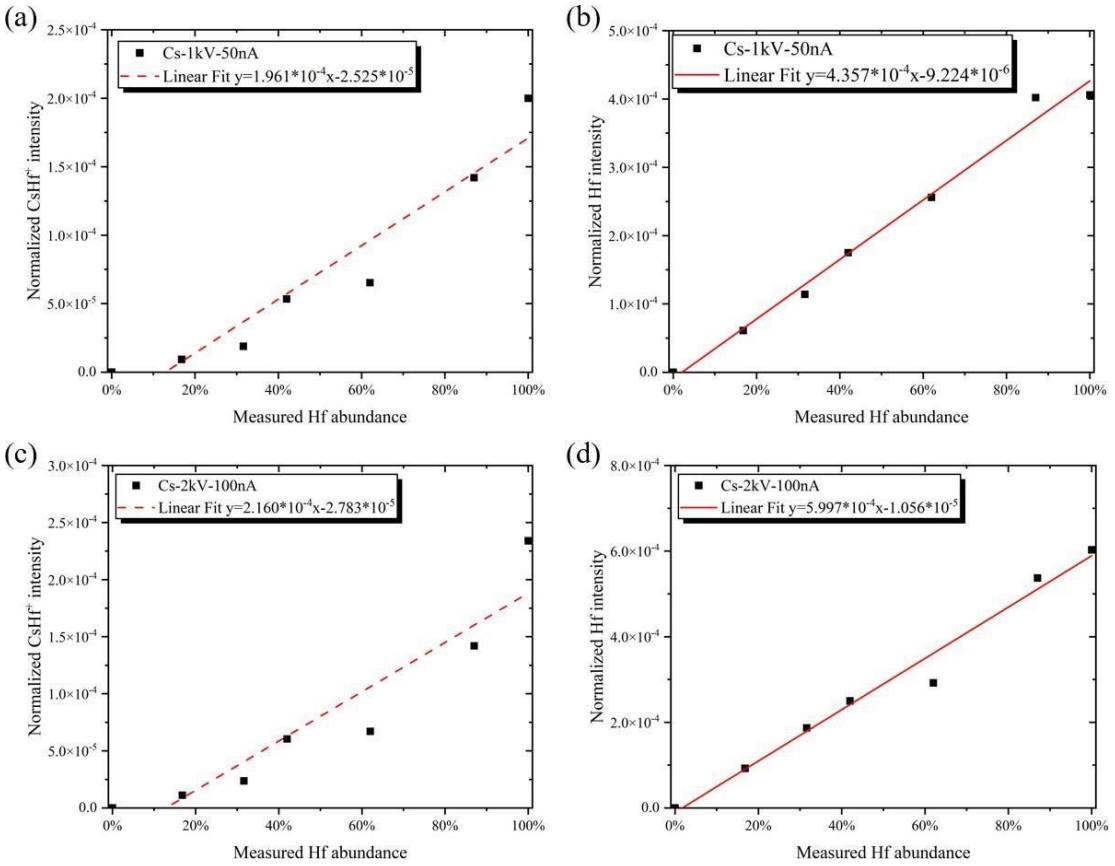

Fig. 8 (a)(c) linear fits of "Cs_nM^+" method under 2 different sputter conditions.

(b)(d) linear fits of "Cluster Counting" method under 2 different sputter conditions. In summary, this section shows the SIMS response and the results of applying "MCs_n^+" and "Cluster Counting" methods on mixed HfO2-SiO2 thin films. "Cs_nM^+" method presents visible matrix effects under different sputter conditions, except when adopting a calibration method(eq. (3-4) and eq.(3-5)).When applying "Cluster Counting" method, matrix effect can be depressed, semi-quantitative analysis with linear correlations between XPS element composition results and SIMS normalized element intensities can be achieved.

4. Conclusions

In this paper, we have presented a study of investigating the capabilities of "Cs_nM^+"and "Cluster Counting" methods in analyzing three types of commonly used optical oxide thin films: SiO_2 thin film, Ta_2O_5-SiO2 multilayer thin film and multiple HfO_2-SiO_2 mixed thin films of different ratios. For SiO2 thin film and Ta2O5-SiO2 multilayer thin film, the "Cluster Counting" method shows close Si/O and Ta/O ratios to the theoretical value on our instrument. Whether this phenomenal can be observed on any other types of thin films needs to be investigate in the future. Different Cs coverage needs to be investigated in order to compensate the ion intensity difference in multilayer samples if semi-quantitative depth profile needs to be conducted.

A systematic HfO2-SiO2 mixed thin films experiment using "Cs_nM^+" and "Cluster Counting" methods was conducted on our instrument. We showed that severe matrix effects

Received: 13 Aug 2025

ISSN: 1311-1728 (printed version); ISSN: 1314-8060 (on-line version)

were observed when considering monatomic Hf^+ ions and $CsHf^+/CsSi^+$ ions. By adopting the calibration method, matrix effects were depressed when using "Cs_nM^+" method, linear correlations between intensity ratio $I(CsSi^+)/I(CsHf^+)$ and element abundance ratio CSi/CHf can be obtained with R^2 of 0.970 and 0.973 depending on different sputter conditions(Fig 7.). For "Cluster Counting" method, linear correlations between normalized Hf intensity and measured Hf abundance can be found with R^2 of 0.988 and 0.980 depending on different sputter conditions(Fig. 8(b)&(d)). These findings reveal the promising future for cluster counting method as a powerful tool for SIMS data analysis. If proper computer algorithm is applied, this method could contribute to huge advancements in dielectrics like optical thin films.

CRediT authorship contribution statement

Xiaoyu Yang: Writing – original draft, Methodology, Investigation, Formal analysis, Data curation. **Yun Cui:** Supervision, Writing – review and editing, Project administration, Resources. **Chunxian Tao:** Supervision, Writing – review and editing. **Jianfei Chen:** Writing – original draft, Investigation. **Shijie Liu:** Supervision, Resources, Project administration. **Jianda Shao:** Supervision, Resources, Project administration, Writing – review and editing.

Acknowledgements

The author would like to thank Yun Cui and SIOM for providing samples. We appreciate Zhengguo Lai for sample preparation and all the pre-work before TOF-SIMS experiments.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be available on reasonable request.

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Received: 13 Aug 2025

ISSN: 1311-1728 (printed version); ISSN: 1314-8060 (on-line version)

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Received: 13 Aug 2025