**SIMS Intensity Correction**

The relative response between the three isotopes of Mg for SIMS should be almost identical. However, we have seen biases of a few percent between them. Thus, the observed intensities $I_i$ for each of the isotopes $i$ should first be corrected $I'_i = I_i R_i$ where $R_i$ are the relative biases between each of the isotopes. The corrected abundance for each isotope becomes:

$$A_i = \frac{I'_i}{I'_{24} + I'_{25} + I'_{26}} = \frac{I_i R_i}{I_{24} R_{24} + I_{25} R_{25} + I_{26} R_{26}}$$  \hspace{1cm} (1)$$

Assuming the isotopic abundances of bulk Mg to be published values, we can determine the relative biases by minimizing the sum of the square of the abundance residuals as a function of depth for the regions representative of the natural isotopes with abundances $B_i$.

$$\sum_x [(A_{24}(x) - B_{24})^2 + (A_{25}(x) - B_{25})^2 + (A_{26}(x) - B_{26})^2]$$  \hspace{1cm} (2)$$

Setting $R_{25} = 1$, we find $R_{24} = 1.045$ and $R_{24} = 1.047$ for the canonical 350°C, 1-hour data in the depth region from 8µm to 12µm. After correction (1), a plot of the abundance deviation is shown in Figure 1. As expected, the deviation from natural abundance vanishes in the deep region where the tracer has not diffused.

![Abundance deviation from natural](image)

Figure 1. Deviation of the corrected Mg abundances from the bulk value as a function of depth.
Fitting for each isotope

As shown earlier, the solution to the one-dimensional diffusion equation for the excess concentration due to the tracer is:

\[
C(x,t) - C_1 = \frac{(C_0 - C_1)}{2} \left[ \text{erf} \left( \frac{x+h}{2\sqrt{Dt}} \right) - \text{erf} \left( \frac{x-h}{2\sqrt{Dt}} \right) \right]. \tag{3}
\]

Thus the atomic fraction of material consisting of the tracer with depth is:

\[
f(x) = \frac{1}{2} \left[ \text{erf} \left( \frac{x+h}{2\sqrt{Dt}} \right) - \text{erf} \left( \frac{x-h}{2\sqrt{Dt}} \right) \right]. \tag{4}
\]

For thin films (i.e., \( h \ll 2\sqrt{Dt} \)), the Gaussian approximation can also be applied, if desired:

\[
f(x) \approx \frac{h}{\sqrt{\pi Dt}} \exp \left( -\frac{x^2}{4Dt} \right). \tag{5}
\]

The fraction can be found by minimizing the sum of the square of the residuals and letting \( h \) and \( D \) be the fitting parameters

\[
\sum_x \left\{ \frac{A_i(x) - B_i}{T_i - B_i} - \frac{1}{2} \left[ \text{erf} \left( \frac{x+h}{2\sqrt{Dt}} \right) - \text{erf} \left( \frac{x-h}{2\sqrt{Dt}} \right) \right] \right\}^2,
\]

where \( T_i \) is the atomic abundance of each isotope in the tracer material. One can see that any of the isotopes can be used for fitting, even though the enriched tracer isotope should have the lower noise in SIMS measurement. This generalization is a nice discovery and allows further consistency checks in the data.

Figure 2 shows the fraction of tracer material \((A_i(x) - B_i)/(T_i - B_i)\) for the canonical 350°C data determined by each of the Mg isotopes. Note that even when \( T_i < B_i \) (e.g., for \(^{24}\text{Mg}\) and \(^{26}\text{Mg}\)), the plots are in good agreement. The root-mean-square noise for each is 0.08%, 0.06% and 0.49% for \(^{24}\text{Mg}\), \(^{25}\text{Mg}\), and \(^{26}\text{Mg}\), respectively. Obviously, the enriched tracer isotope, \(^{25}\text{Mg}\), has the lowest noise, as expected.

Careful examination also suggests slight differences in the slope for each isotope. Applying eq. (6) for each isotope in the 350°C data, we find diffusion coefficients of 9.71x10^{-12} \text{ cm}^2/\text{s}, 9.47x10^{-12} \text{ cm}^2/\text{s}, and 7.99x10^{-12} \text{ cm}^2/\text{s} for \(^{24}\text{Mg}\), \(^{25}\text{Mg}\), and \(^{26}\text{Mg}\), respectively. These are plotted in Figure 3 along with the same data for two other samples annealed at 300°C and 400°C. Plotting as diffusion lengths, \(2\sqrt{Dt}\), allows closer comparison, while scaling with temperature-equivalent velocity, \(\sqrt{2RT/m_a}\), allows extrapolation to the origin. (\(m_a\) is the atomic mass.) The variation is admittedly small, but the trend is consistent with slower diffusion of heavier isotopes.
Figure 2. Atomic fraction of diffused tracer material \( \left( A_i(x) - B_i \right) / \left( T_i - B_i \right) \), as determined by each of the Mg isotopes. The diffusion coefficient fitted from the \(^{25}\text{Mg}\) isotope is \( 9.47 \times 10^{-12} \text{ cm}^2/\text{s} \).

Figure 3. Diffusion length, \( 2\sqrt{D_t} \), determined by fitting with each of the Mg isotopes plotted against temperature-equivalent velocity. Linear fits through the origin are shown for data from three samples annealed at the indicated temperatures.