On recycling in high power impulse magnetron sputtering discharges

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Introduction

- Magnetron sputtering has been a highly successful technique that is essential in a number of industrial applications.

- A magnet is placed at the back of the cathode target with the pole pieces at the center and perimeter.

- The magnetic field confines the energetic electrons near the cathode.

- The electrons undergo numerous ionizing collisions before being lost to a grounded surface.
High ionization of sputtered material requires very high density plasma.

In a conventional dc magnetron sputtering discharge the power density (plasma density) is limited by the thermal load on the target.

High power pulsed magnetron sputtering (HPPMS)

In a HiPIMS discharge a high power pulse is supplied for a short period

- low frequency
- low duty cycle
- low average power

Gudmundsson et al. (2012) JVSTA 30 030801

- Power density limits
  $p_t = 0.05 \text{ kW/cm}^2$ dcMS limit
  $p_t = 0.5 \text{ kW/cm}^2$ HiPIMS limit
On recycling in high power impulse magnetron sputtering discharges

**High power impulse magnetron sputtering discharge**

- Temporal and spatial variation of the electron density
- Ar discharge at 20 mTorr, Ti target, pulse length $100 \mu s$
- The electron density in the substrate vicinity is of the order of $10^{18} \sim 10^{19} \text{ m}^{-3}$ – ionization mean free path $\lambda_{iz} \sim 1 \text{ cm}$

(After Bohlmark et al. (2005), IEEE Trans. Plasma Sci. 33 346)
Ionization region model studies of HiPIMS discharges
The ionization region model (IRM) was developed to improve the understanding of the plasma behaviour during a HiPIMS pulse and the afterglow.

The main feature of the model is that an ionization region (IR) is defined next to the race track.

The IR is defined as an annular cylinder with outer radii $r_{c2}$, inner radii $r_{c1}$ and length $L = z_2 - z_1$, extends from $z_1$ to $z_2$ axially away from the target.

From Raadu et al. (2011) PSST 20 065007
Ionization region model of HiPIMS

- The temporal development is defined by a set of ordinary differential equations giving the first time derivatives of:
  - the electron energy
  - the particle densities for all the particles
- The species assumed in the of-IRM are:
  - cold electrons $e^C$ (Maxwellian), hot electrons $e^H$ (sheath acceleration)
  - argon atoms $\text{Ar}(3s^23p^6)$, warm argon atoms in the ground state $\text{Ar}^W$, hot argon atoms in the ground state $\text{Ar}^H$, $\text{Ar}^m$ ($1s_5$ and $1s_3$) (11.6 eV), argon ions $\text{Ar}^+$ (15.76 eV)
  - titanium atoms $\text{Ti}(a^3F)$, titanium ions $\text{Ti}^+$ (6.83 eV), doubly ionized titanium ions $\text{Ti}^{2+}$ (13.58 eV)
  - aluminium atoms $\text{Al}(^2P_{1/2})$, aluminium ions $\text{Al}^+$ (5.99 eV), doubly ionized aluminium ions $\text{Al}^{2+}$ (18.8 eV)

Detailed model description is given in Huo et al. (2017) JPD 50 354003
The model is constrained by experimental data input and fitted to reproduce the measured discharge current and voltage curves, $I_D(t)$ and $V_D(t)$, respectively.

Two model fitting parameters were found to be sufficient for a discharge with Al target:

- $V_{IR}$ accounts for the power transfer to the electrons
- $\beta$ is the probability of back-attraction of ions to the target

From Huo et al. (2017) JPD 50 354003

Experimental data from Anders et al. (2007) JAP 102 113303
A non-reactive discharge with 50 mm diameter Al target

Current composition at the target surface

From Huo et al. (2017) JPD 50 354003

Experimental data from Anders et al. (2007) JAP 102 113303
Ionization region model of HiPIMS

- When the discharge is operated at 400 V the contributions of Al\(^+\) and Ar\(^+\)-ions to the discharge current are very similar.
- At 800 V Al\(^+\)-ions dominate the discharge current (self-sputtering) while the contribution of Ar\(^+\) is below 10% except at the initiation of the pulse.

From Huo et al. (2017) JPD 50 354003
Experimental data from Anders et al. (2007) JAP 102 113303
Ionization region model of HiPIMS

- A primary current $I_{\text{prim}}$ is defined as ions of the working gas, here Ar$^+$, that are ionized for the first time and then drawn to the target.
- This is the dominating current in dc magnetron sputtering discharges.
- This current has a critical upper limit

$$I_{\text{crit}} = S_{RT} e p_g \sqrt{\frac{1}{2\pi m_g k_B T_g}} = S_{RT} e n_g \sqrt{\frac{k_B T_g}{2\pi m_g}}$$

- Discharge currents $I_D$ above $I_{\text{crit}}$ are only possible if there is some kind of recycling of atoms that leave the target, become subsequently ionized and then are drawn back to the target.

Anders et al. (2012) JPD 45 012003
Huo et al. (2014) PSST 23 025017
For the 50 mm diameter Al target the critical current is $I_{\text{crit}} \approx 7 \, \text{A}$

The experiment is operated from far below $I_{\text{crit}}$ to high above it, up to 36 A.

With increasing current $I_{\text{prim}}$ gradually becomes a very small fraction of the total discharge current $I_D$

The current becomes mainly carried by singly charged Al$^+$-ions, meaning that self-sputter recycling or the current $I_{SS}$—recycle dominates

From Huo et al. (2017) JPD 50 354003

Experimental data from Anders et al. (2007) JAP 102 113303
For discharges with Ti target the peak current is far above the critical current (up to 650 A, while $I_{\text{crit}} \approx 19$ A)

However, this discharge shows close to a 50/50 combination of self-sputter recycling $I_{\text{SS-recycle}}$ and working gas-recycling $I_{\text{gas-recycle}}$

Almost 2/3 of the current to the target is here carried by Ar$^+$ and Ti$^{2+}$-ions, which both can emit secondary electrons upon target bombardment, and this gives a significant sheath energization

From Huo et al. (2017) JPD 50 354003
Ionization region model of HiPIMS

- Recall that singly charged metal ions cannot create the secondary electrons – for metal self-sputtering ($\gamma_{SE}$ is practically zero)
- The first ionization energies of many metals are insufficient to overcome the workfunction of the target material
- For the discharge with Al target operated at high voltage, self-sputter dominated, the effective secondary electron emission is essentially zero

From Anders (2008) APL 92 201501
**Ionization region model of HiPIMS**

- **Reactive HiPIMS**
- **Ar/O₂ discharge with Ti target**
- For this system \( I_{\text{crit}} \approx 5 \) A
- In the metal mode \( \text{Ar}^+ \) and \( \text{Ti}^+ \)-ions contribute roughly equally to the current – combined **self-sputter recycling** and working gas recycling
- In the poisoned mode the current increases and \( \text{Ar}^+ \)-ions dominate the current – **working gas recycling**

From Gudmundsson et al. (2016) PSST 25(6) 065004
For the Al target, Ohmic heating is in the range of 87% (360 V) to 99% (1000 V).

The domination of Al\(^{+}\)-ions, which have zero secondary electron emission yield, has the consequence that there is negligible sheath energization.

The ionization threshold for twice ionized Al\(^{2+}\), 18.8 eV, is so high that few such ions are produced.
For a Ti target Ohmic heating is about 92%.
- Both $\text{Ar}^+$ and $\text{Ti}^{2+}$-ions contribute to creation of secondary electrons.

For Ti target in Ar/O$_2$ mixture:
- In the metal mode Ohmic heating is found to be 90% during the plateau phase of the discharge pulse.
- For the poisoned mode Ohmic heating is 70% with a decreasing trend, at the end of the pulse.

From Huo et al. (2017) JPD 50 354003
Ionization region model of HiPIMS

- Ohmic heating is also very significant in dc magnetron sputtering discharges.
- The relative contributions to the total ionization $\iota_{\text{total}}$ due to Ohmic heating, $\iota_{\text{Ohmic}}$, and sheath energization, $\iota_{\text{sheath}}$.
- A blue circle marks the HiPIMS study modelled by Huo et al. (2013).
- Note that this HiPIMS case $\gamma_{\text{SE,eff}}$ is consistent with the dcMS cases.

From Brenning et al. (2016). PSS 125 065024
The generalized recycling model
Generalized recycling

- A working gas-sputtering parameter

\[ \pi_g = \alpha_g \beta_g \xi_{\text{pulse}} \]

where

- \( \alpha_g \) is ionization probability
- \( \beta_g \) is back attraction probability
- \( \xi_{\text{pulse}} = 1 \) is return fraction in a pulse

- The total current carried by working gas ions

\[ I_g = I_{\text{prim}} + I_{\text{gas-recycle}} = I_{\text{prim}} \left( 1 + \frac{\pi_g}{1 - \pi_g} \right) \]

From Brenning et al. (2017) PSST 26 125003
Generalized recycling

The total self-sputter current is

\[ I_{SS} = I_g \left( \frac{Y_g}{Y_{SS}} \frac{\pi_{SS}}{1 - \pi_{SS}} \right) \]

where the self-sputter parameter is

\[ \pi_{SS} = \alpha_t \beta_t Y_{SS} \]

The total discharge current is

\[ I_D = I_{prim} + I_{gas-recycle} + I_{SS} \]

\[ = I_{prim} \left( 1 + \frac{\pi_g}{1 - \pi_g} \right) \left( 1 + \frac{Y_g}{Y_{SS}} \frac{\pi_{SS}}{1 - \pi_{SS}} \right) \]

From Brenning et al. (2017) PSST 26 125003.
Generalized recycling

- The discharge current

\[ I_D = I_{\text{prim}} \Pi_{\text{gas-recycle}} \Pi_{\text{SS-recycl}} \]

- \( I_{\text{prim}} \) is the seed current acts as a seed to the whole discharge current and has an upper limit \( I_{\text{crit}} \)

- \( I_{\text{prim}} \Pi_{\text{gas-recycle}} \) is the seed current for the self-sputter process

- If \( \pi_{\text{SS}} > 1 \) the discharge goes into SS-runaway

From Brenning et al. (2017), PSST 26 125003
Generalized recycling

- Recycling map
- A graph in which the ion current mix of \( I_{\text{prim}} \), \( I_{\text{gas-recycle}} \), and \( I_{\text{SS}} \) to the target in a magnetron discharge is defined by a point
- The value of \( I_{\text{prim}} / I_D = 39 \% \), can be read on the diagonal lines (\( Y_{SS} = 0.5 \))
- \( I_{\text{prim}} / I_D \geq 0.85 \) defines the dcMS regime
- For \( I_{\text{SS}} / I_D > 0.5 \) we have the SS-recycle dominated range A
- For \( I_{\text{gas-recycle}} / I_D > 0.5 \) we have the gas-recycle dominated range B

From Brenning et al. (2017) PSST 26 125003
Generalized recycling

- The discharge with Al target moves from the dcMS regime to the HiPIMS discharge regime with increased discharge voltage – **type A**
- A discharge with carbon target jumps from the dcMS regime to the HiPIMS regime – both SS recycling and working gas recycling play a role – intermediate **type AB**

From Brenning et al. (2017) PSST 26 125003
Generalized recycling

- Recycling map for five different targets with varying self-sputter yield
  - Cu – \( Y_{SS} = 2.6 \)
  - Al – \( Y_{SS} = 1.1 \)
  - Ti – \( Y_{SS} = 0.7 \)
  - C – \( Y_{SS} = 0.5 \)
  - TiO\(_2\) – \( Y_{SS} = 0.04 – 0.25 \)

- For very high self-sputter yields \( Y_{SS} > 1 \), the discharges above \( I_{crit} \) are of type A with dominating SS-recycling

- For very low self-sputter yields \( Y_{SS} < 0.2 \), the discharges above \( I_{crit} \) are of type B with dominating working gas recycling

From Brenning et al. (2017), PSST 26 125003
Generalized recycling

- Recycling loops
- Discharge with Al target – SS recycling dominates
  - high self sputter yield
- Reactive discharge with TiO₂ target – working gas recycling dominates
  - low self sputter yield

From Brenning et al. (2017) PSST 26 125003
HiPIMS - Voltage - Current - time

- For Ar/O₂ discharge with Ti target
- At high frequencies, oxide is not able to form between pulses, and **self-sputtering recycling by Ti⁺-ions** is the dominant process
- At low frequency, the long off-time results in an oxide layer being formed (TiO₂) on the target surface and **working gas recycling dominates** – triangular current waveform

From Gudmundsson (2016), PPCF 58:014002
Magnus et al. (2012), JVSTA 30:050601
Summary

For high currents the discharge with Al target develops almost pure **self-sputter recycling**, while the discharge with Ti target exhibits close to a 50/50 combination of **self-sputter recycling** and **working gas-recycling**.

For very high self-sputter yields, above approximately $Y_{SS} \approx 1$, the discharges above $I_{crit}$ are of type A with
- dominating SS-recycling
- very little secondary electron emission
- little sheath energization of electrons

For very low self-sputter yields, below approximately $Y_{SS} \approx 0.2$, the discharges above $I_{crit}$ are of type B with
- dominating working gas recycling
- significant secondary electron emission
- significant sheath energization of electrons.

The fraction of the total electron heating that is attributable to Ohmic heating is over 90% in the HiPIMS discharge.
Thank you for your attention

The slides can be downloaded at
http://langmuir.raunvis.hi.is/~tumi/ranns.html
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References


