Deposition of Gd_2O_3 by Reactive Anodic Evaporation in Arc with Self-heated Hollow Cathode

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Thin Gd_2O_3 coatings

Properties

- High thermal and chemical stability
- Bang gap – 5.2 eV
- Dielectric constant ~16
- Transparency in wavelength range 0.19-16 µm

Application of thin (>1 µm) coatings

- Microelectronics (metal oxide semiconductor field effect transistors)
- Optics (UV and deep UV laser)

Deposition methods (*low-rate – less than 100 nm/h*)

- Magnetron sputtering
- Molecular-beam epitaxy
- Electrolytic deposition

Thick Gd_2O_3 coatings

Application of thick (<1 μm) coatings

- High temperature oxidation protection
- Neutron detectors

Deposition problem

Polymorphism of Gd_2O_3 enhanced adhesion problem of thick coatings:

- Phase transformation occurs with an increase in coating thickness
- Significant (~ 8 %) difference in the molar volumes of Gd_2O_3 phases
- High value of the CTE ($15.5 \cdot 10^{-6}$ C^{-1}) and stabilization temperature (>600 ^oC) of the monoclinic phase

Deposition methods

(*low-rate – less than 100 nm/h*)

- Electron beam physical vapor deposition

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Process–structure–property relations of micron thick Gd_2O_3 films deposited by reactive electron-beam physical vapor deposition (EB-PVD)

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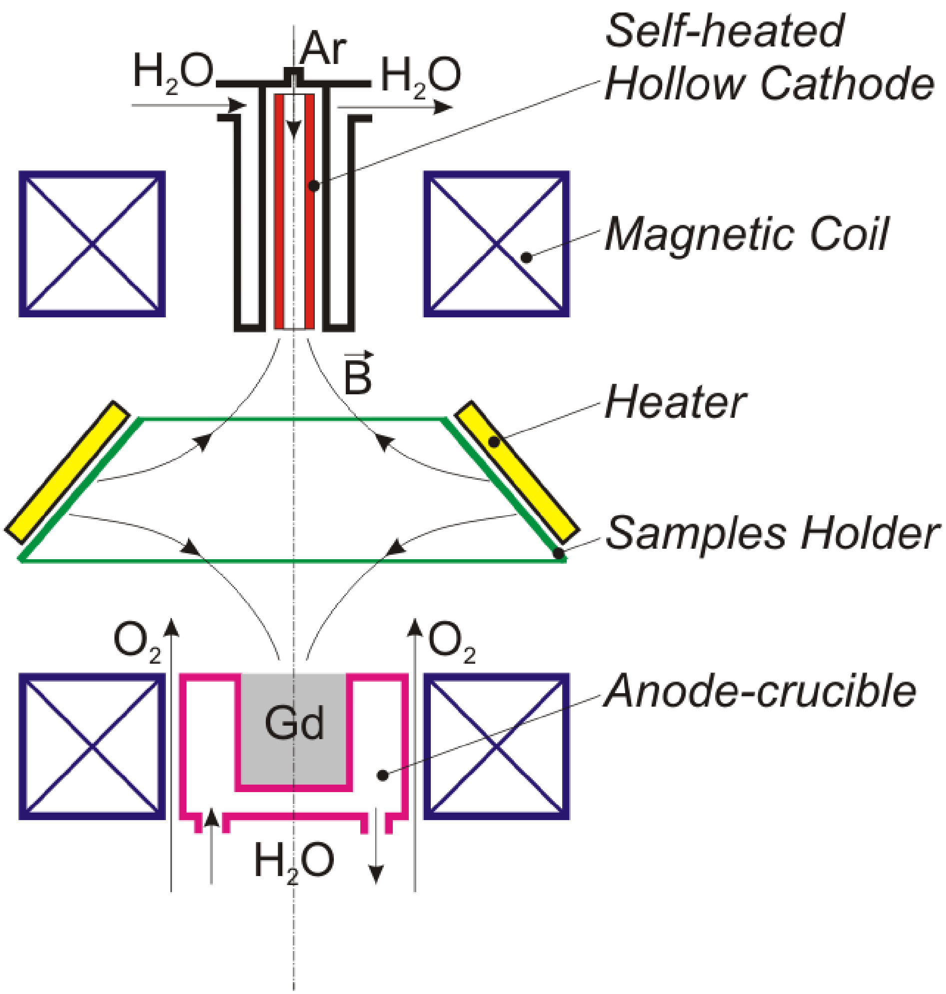
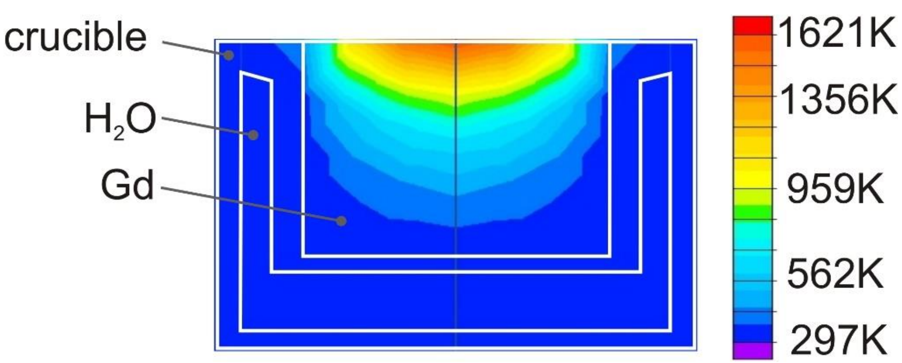
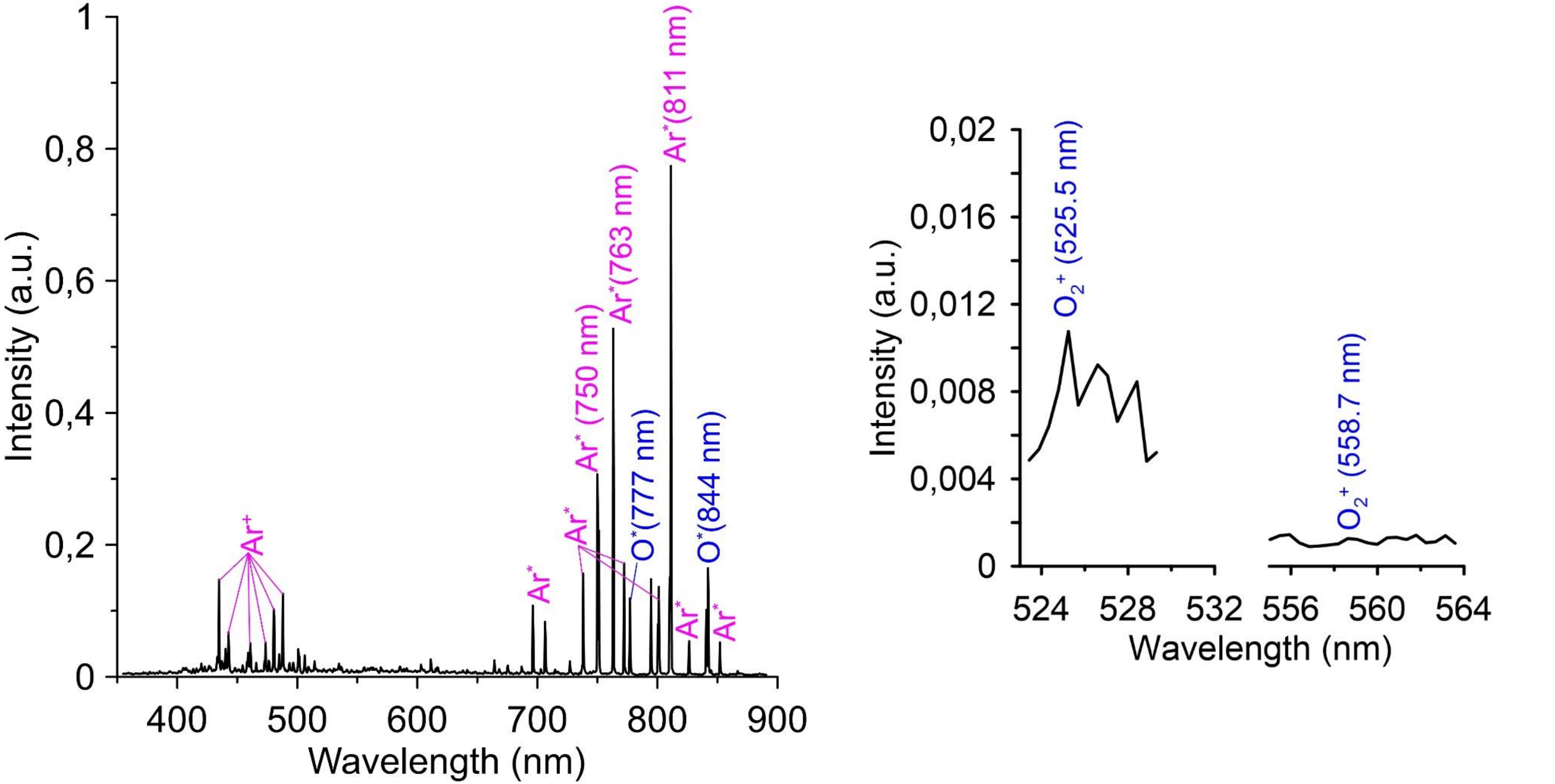
Deposition method



Discharge current	>30 A
Power density on the anode-crucible surface	>0.5 W/cm^2
Magnetic induction (maximum)	20 mT
Ion current density	>5 mA/cm^2
Ar/O_2 flow rate	30/10 sccm
Samples temperature	150 – 600 ^oC

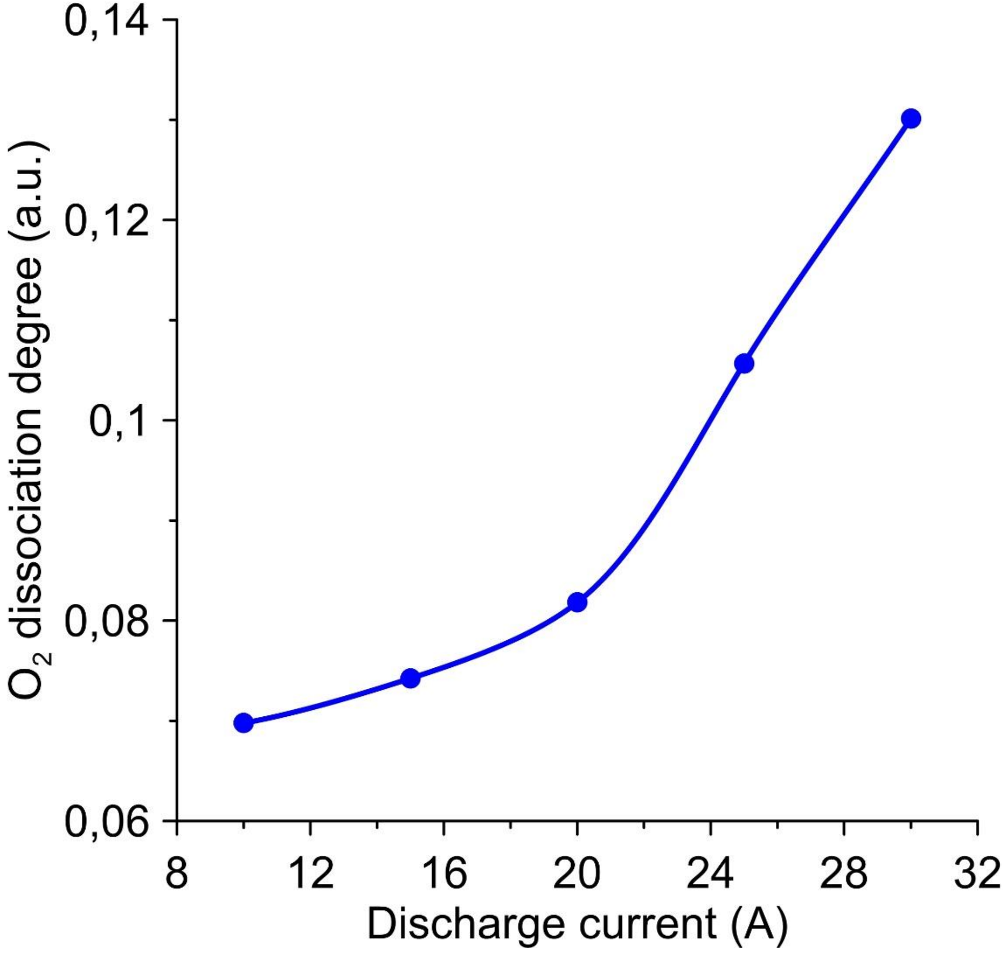


Optical emission of plasma



HR2000 (Ocean Optics) spectrometer
Wavelength range 190-1100 nm
Exposure time – 25-1000 ms

Oxygen dissociation degree



$$\frac{I_{O(777\ nm)}}{I_{Ar(750\ nm)}} = \frac{h\nu_{777} A_{ij}^{5P}}{h\nu_{750} A_{ij}^{3p_1}} \frac{\sum A_{ij}^{2p_1}}{\sum A_{ij}^{5P} + k_Q^{5P} n_{O2}} \frac{k_e^{5P} n_O + k_{de}^{5P} n_{O2}}{k_e^{2p_1} n_{Ar}}$$

I – intensity of emission line

n_O and n_{O2} - concentrations of atomic and molecular oxygen

$h\nu_x$ – photon energy with the wavelength *x*

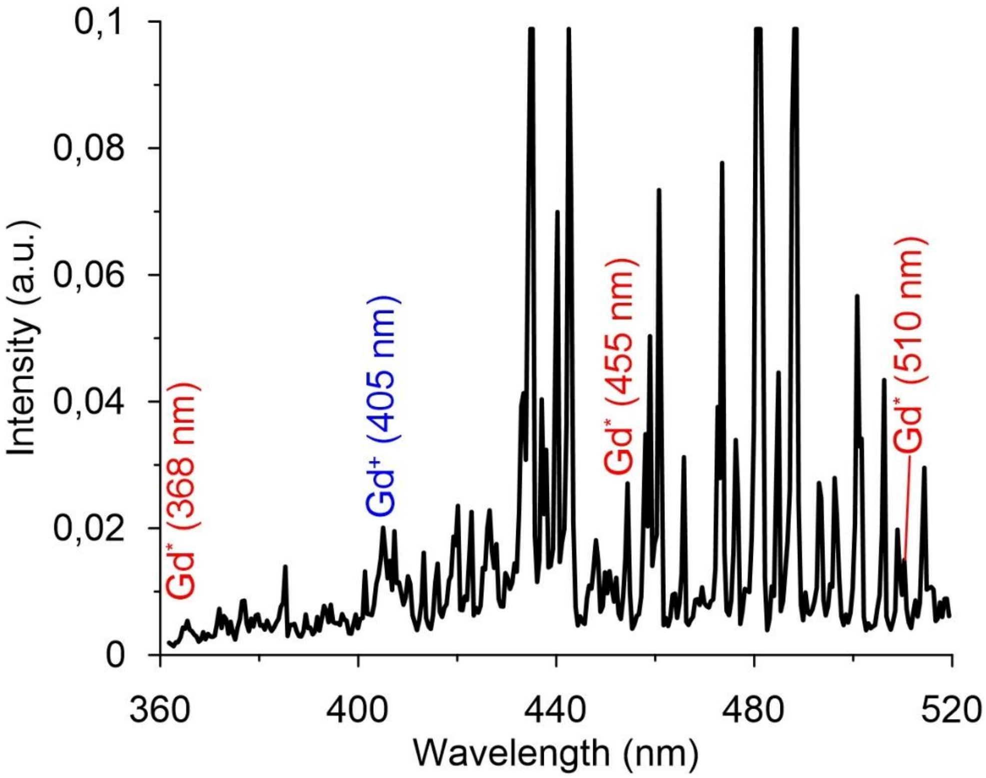
A_{ij} – probability of spontaneous transition between the levels *i* and *j*

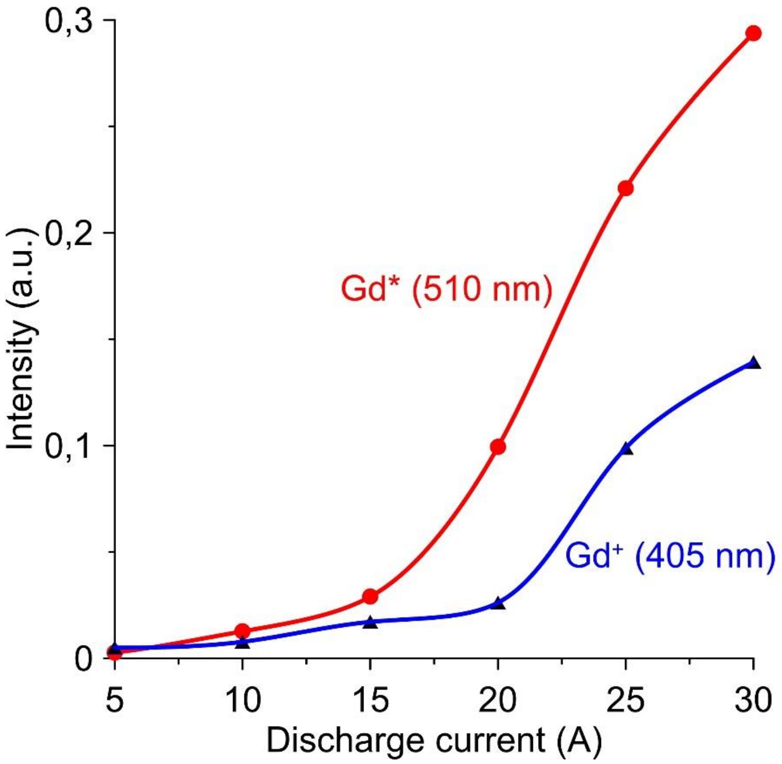
ΣA_{ij}^Y – summ of probabilities of all processes of quenching from the excited level *Y*

k_e, k_{de}, k_Q – constants for reactions of excitation by direct electronic impact, dissociative excitation and quenching involving molecular oxygen

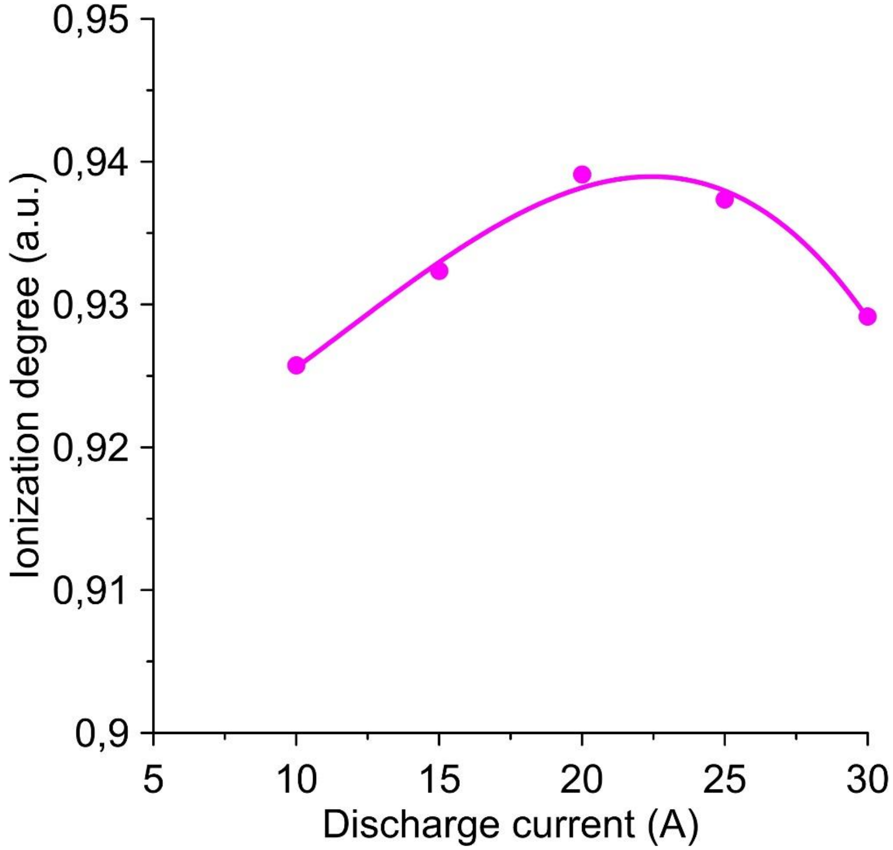
n_{Ar} – Ar concentration

Gadolinium content





Gadolinium ionization degree



$$I_{ij} = A_{ij}\, h\nu_{ij}\, n_a\, g_i / Z_a \exp\left(-E_i/kT\right)$$

I_{ij} - intensity of the spectral line of spontaneous transition between the levels i and j

n_a - concentration of particles

T - particle temperature

g_j – statistical weight

E_j – energy of the excited state

Z_q – electronic partition function

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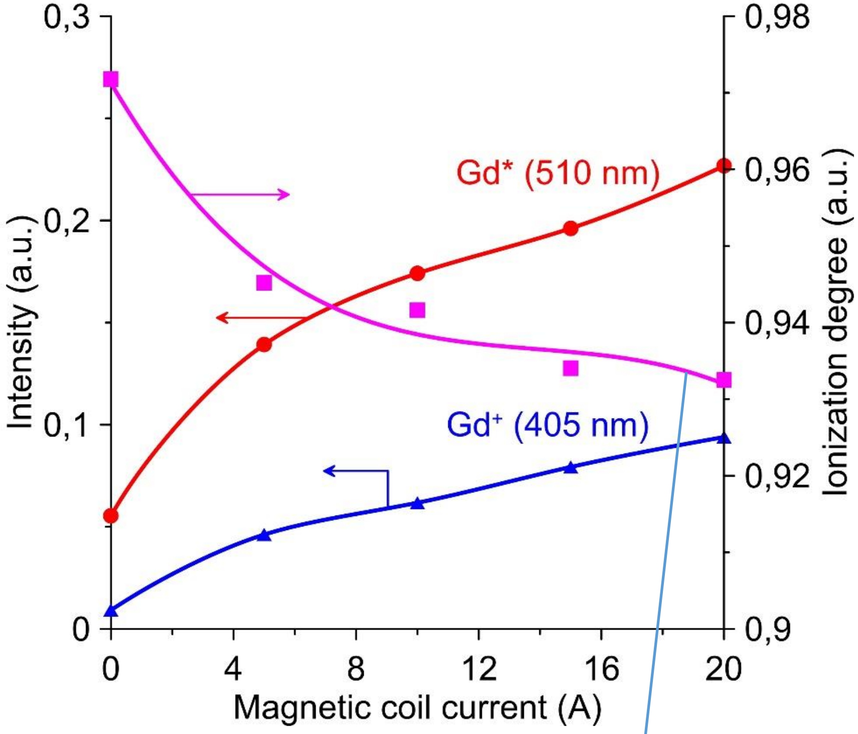
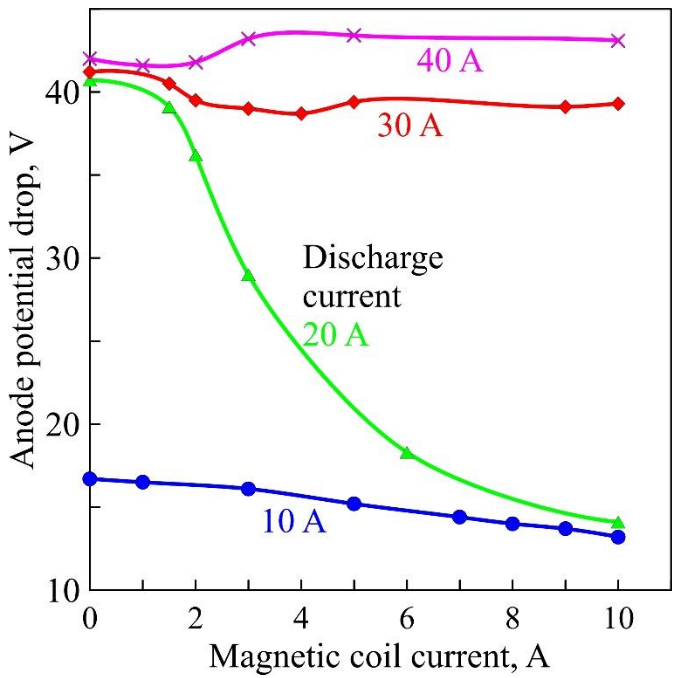
Magnetic focusing effect



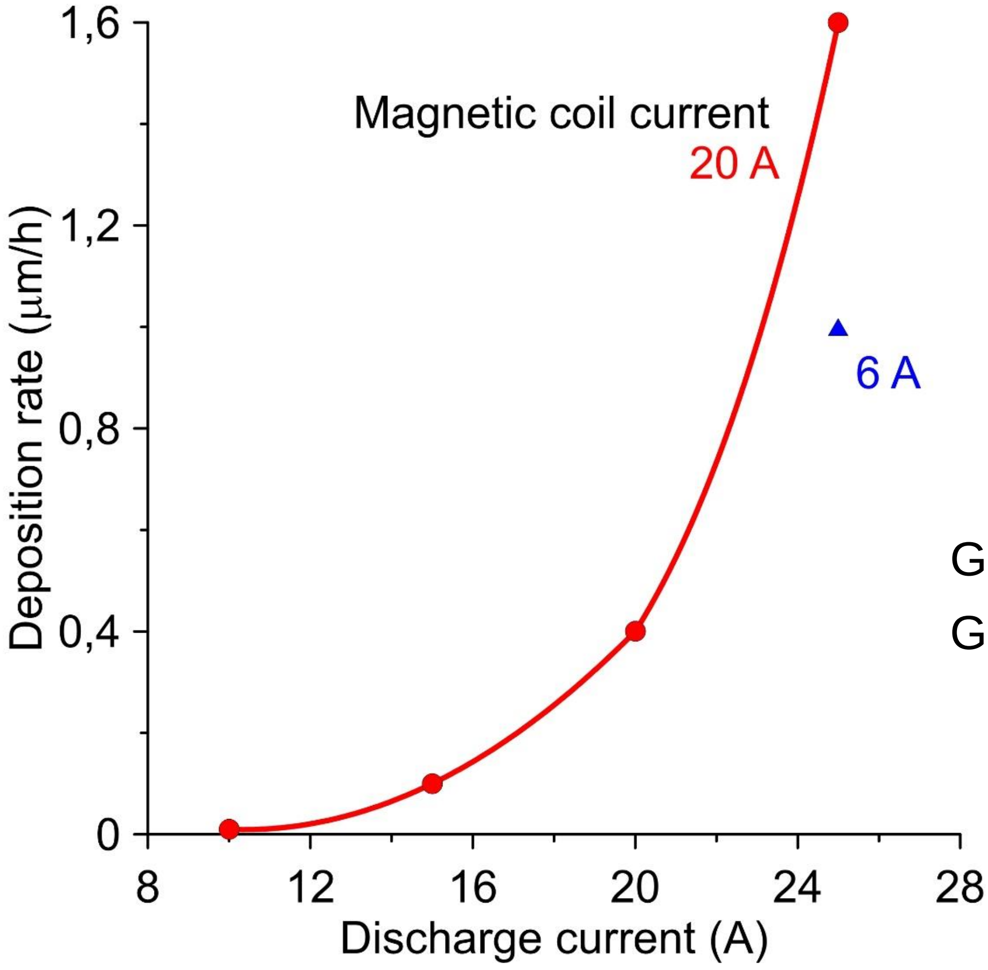


$$\frac{n_i}{n_i + n_0} \sim \frac{\sigma_i(E)\sqrt{E}}{\sigma_i(E)\sqrt{E} + 1}$$

n_i, n_o – concentration of ions and neutrals
σ_i – electron impact ionization cross section
E – electron energy

N.V. Gavrilov, A.S. Kamenetskikh, S.V. Krivoshapko, P.V. Tretnikov. Discharge with a self-heated hollow cathode and a vaporizable anode in an inhomogeneous magnetic field. Journal of Physics: Conf. Series 1115 (2018) 032005

Gd-O coating deposition rate



Gd evaporation rate up to $2.6 \cdot 10^{-5}$ g/(s·cm^2)

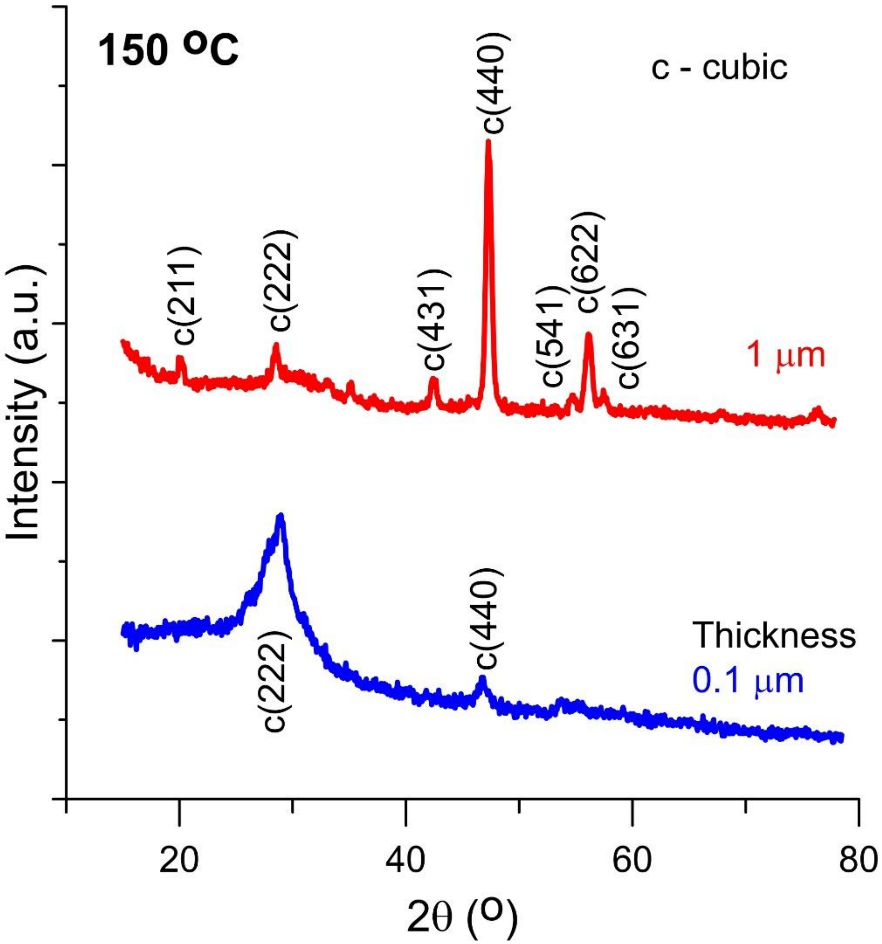
Gd vapor pressure up to 0.13 Pa

Gd-O coating deposition condition

- Oxygen dissociation degree ~ 0.13
- Metal vapor ionization degree ~ 0.93
- Deposition rate ~1,6 μm/h
- Ar/O pressure 0.1 Pa
- Low O_2 flow rate (10 sccm)

- Substrate – quartz
- Temperature range 150 – 600 ^{o}C

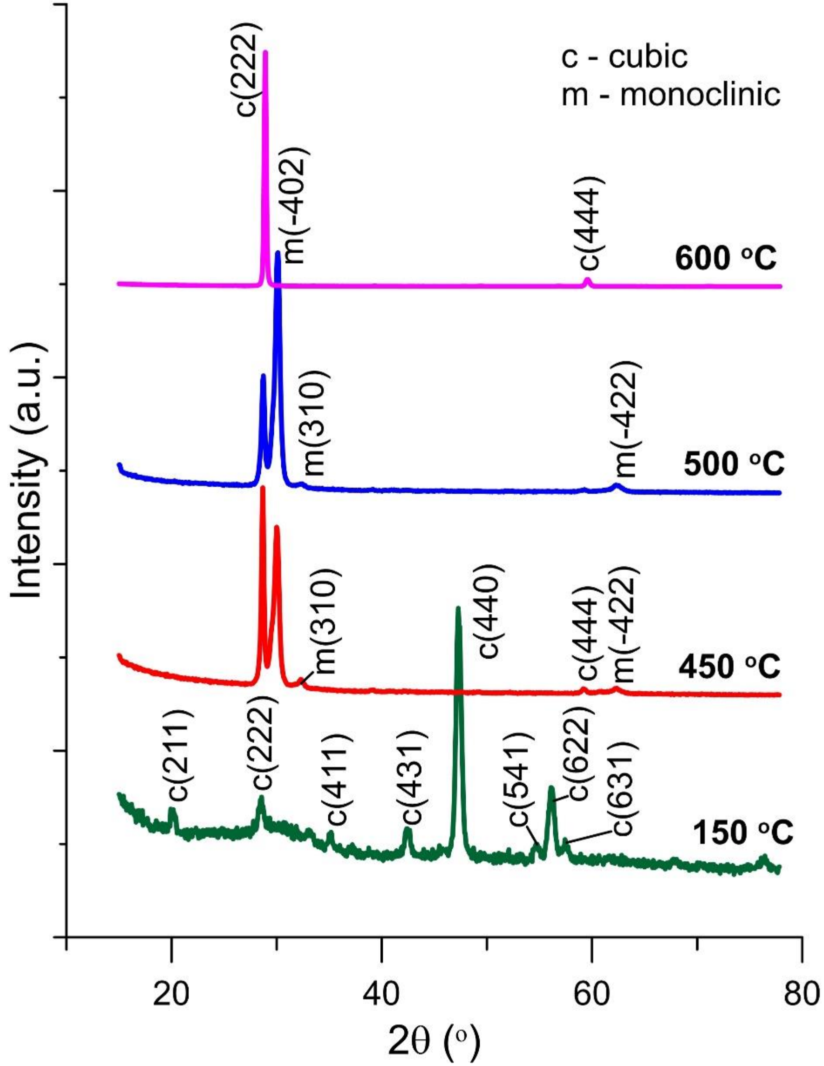
Phase composition of coatings

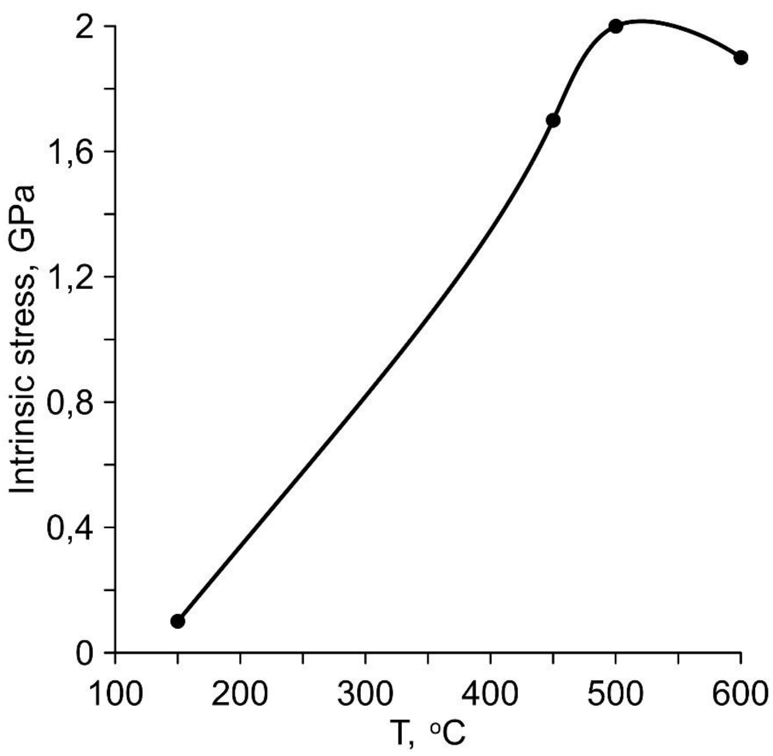


Features of the Gd_2O_3 coating deposited by anodic arc evaporation method

- Low (0.1 GPa) value of intrinsic stress
- High (up to 1 μm) thickness of monophasic cubic coating
- Strong (440) texture which appearance is not typical for coatings deposited by other methods
- Low temperature (150 °C) deposition of the monophasic cubic coating take place under low O_2 flow rate (10 sccm which is an order of magnitude less than in other methods)

Temperature effect





Conclusion

- The reactive anodic arc evaporation method provides high-rate (1,6 μm/h) deposition of the nanocrystalline Gd_2O_3 coating
- Low-temperature (less than 200 ^{o}C) deposition of monophasic cubic Gd_2O_3 coating up to 1 μm thick is provided under high Gd ionization (~90 %) and O_2 dissociation (~13 %) degree and low (10 sccm) O_2 flow rate
- The coatings are characterized by low level of intrinsic stress (~0.1 GPa) which ensures their adhesive strength

Thank you for your attention!

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