Process control loops using optical emission spectroscopy in reactive sputtering

C.Vitelaru(*), I. Pana, A.C Parau, M. Dinu, L.R. Constantin, A. Vladescu

National Institute of Research and Development for Optoelectronics - INOE 2000, Department for Advanced Surface Processing and Analysis by Vacuum Technologies, 409 Atomistilor St., Magurele, RO77125, Romania

(*)catalin.vitelaru@inoe.ro

Optical emission spectroscopy (OES) is a valuable tool for plasma diagnostics, giving direct access to the excited species in the plasma volume. The main advantage of OES is its non-invasive nature, being well adapted for the investigation and control of technological plasmas that are used for thin film deposition. The use of this technique is illustrated in this contribution for characterizing and controlling the reactive sputtering process in a variety of cases. The use of two reactive gas configurations with variable gas mixing is presented as a first example, for the deposition of TaOxNy compounds. The use of control loops to actively control the reactive gas flow is presented both for single reactive gas (O_2 and N_2 respectively) and mixture of two reactive gases. Moreover, a special design that uses the emission of an additional plasma is presented, for the control of a reactive process involving two reactive gases.

The plasma-based technologies used for surface processing and thin film deposition are of great interest, being labeled as ecological due to their lack of harmful byproducts, when compared to chemical deposition methods. Physical vapor deposition in general and magnetron sputtering in particular are valuable technologies, intensively used both in research and industry [1,2]. Reactive magnetron sputtering is of particular interest, enabling the creation of a large variety of compounds, including oxides, nitrides, carbides, oxynitrides, carbonitrides, by using a metal target and a gas mixture that contains reactive species, such as oxygen, nitrogen and hydrocarbon gases. The reactive gas present in the plasma volume leads to complex and interdependent phenomena that occur on all surfaces and in the volume, involving compound formation and destruction. One typical characteristic of the reactive sputtering process is the presence of Hysteresis phenomena [3,4]. This can be made visible as a different variation path of a process parameter, such as partial or total pressure, voltage, power, emission lines intensity, etc., corresponding to the variation of a control parameter, such as reactive gas flow, target current or power. The process window where hysteresis occurs can lead to process instability, the process parameters being dependent on the history of the system. The same interval also offers a potential advantage, being precisely the one that can be used to tune the properties of the thin films.

Optical emission spectroscopy is a powerful tool that can be successfully used for analyzing the reactive process [5,6,7], providing knowledge of process intervals and giving insight on the elementary processes. Using such process intervals, it becomes possible to choose the best suited process parameters, so that tunability of thin films properties can be achieved. In this contribution an illustration is given for different configurations, including one gas and two gas processes, with and without process control loops, presented in the form of 4 case studies.

TaO_xN_y, tuning in a 2 reactive gases process

The first case study refers to the processes involving two reactive gases, namely oxygen and nitrogen, for the deposition of oxynitride thin films. The contribution of each reactive gas is different, resulting in a more complex reactive environment. The process that will be described implies the sputtering of Ta target in an $Ar/N_2/O_2$ gas mixture, for the deposition of TaO_xN_y thin films with tunable composition and properties. The specific constant parameters for this process were: DC power applied to the target P_{DC} = 150 W, 5 mTorr of Ar pressure, Ar gas flow of 5 sccm.

The chosen emission lines to be followed for describing the process are: λ=337.13 nm N_2 line, λ=534.1 nm Ta line, λ=706 nm Ar line and λ=777.19 O line. The analysis of the hysteresis behavior, investigated individually for each of the reactive gas, reveals a hysteresis interval from 1.8 to 3 sccm of O_2, while for the Nitrogen process there is no visible hysteresis effect. The most reactive process conditions were identified for an oxygen flow of 2.5 sccm. In order to identify the most reactive conditions for nitrogen process, the derivative of the intensity variation vs gas flow was used, as described in [5], resulting that 1 sccm of N_2 corresponds to the most reactive conditions. In order to simplify the experimental procedure, we propose the use of only one control parameter. This parameter is the sum of the reactive gas flows, coupled with a fixed ratio of 2.5 between the two reactive gases flows, in order to account for their different reactivities. Using this approach, the hysteresis behavior for a two reactive gas process cand be derived, leading to the identification of the process interval best suited for obtaining tunable composition and properties. By choosing three experimental conditions situated in the interval that defines the hysteresis loop, the tunability of the film properties is proven in terms of composition and optical properties. Refractive index variation from 2.1 to 3.3 is achieved, while the optical bandgap variation lays in the interval from 1.6 to 3.2 eV.

CuO_x and CuN_y tunning by active control loop with one reactive gas

The second case study refers to the implementation of active control loops, for the sputtering of Cu target in a reactive environment containing Ar and one reactive gas, either O_2 or N_2. Plasma emission monitoring and control of the plasma is employed, by using a spectrometer that collects the emitted light from the vicinity of the magnetron plasma via an optical fiber. The ratio of selected line intensities, $I(\lambda O_{777\,nm})/I(\lambda Cu_{327\,nm})$ and $I(\lambda N_{337\,nm})/I(\lambda_{Cu327nm})$respectively, is transformed by a digital-analog convertor, becoming the „control signal" for a programable PID controller unit. This unit controls the reactive gas flow, such as the desired ratio between emission lines is kept.

The reactivity of the processes with one reactive gas, O_2 or N_2 is evaluated by using the reactive gas flow as a control parameter and following the evolution of voltage, pressure, or emission line intensities. The Hysteresis effect is more pronounced for the process involving oxygen, a clear interval being visible between 3.5 and 4.5 sccm of O_2. The correct setting of the PID parameters enables a precise control of the desired line intensity ratio, enabling both stable operation at exact setpoint and also quick transition to the desired setpoint in the event of a simulated perturbation of the system. Moreover, the gradual increase of the setpoint value enables stable operation of the system inside the hysteresis loop, represented either as voltage or line intensities ratio as a function of oxygen gas flow. Typical variation of the process parameters as a function of Oxygen flow is represented in Figure 1.

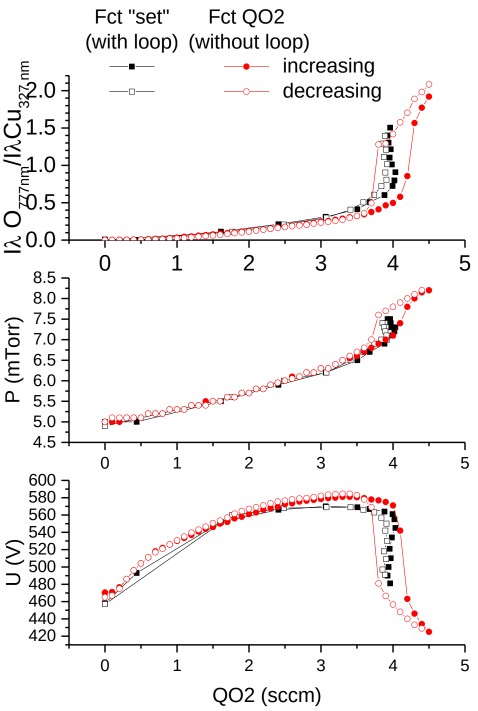

Fig. 1: Hysteresis behavior with and without control loop

The direct link between process conditions and thin film properties is revealed by evaluating the deposition rate, the Cu to O ratio, measured by EDS or the optical band gap variation. Moreover, the XRD spectra of the samples shows that by controlling the ratio of emission line intensities $I(\lambda O_{777\,nm})/I(\lambda Cu_{327\,nm})$it is possible to obtain different crystalline phases, including Cu_2O, Cu_4O_3 and CuO, depending on the composition and the O/Cu ratio.

TiOxNy tunning by active control loop with 2 reactive gases

For the third case study, a process with two reactive gasses is used, namely O_2 and N_2, for the sputtering of a Ti target. For this study the chosen ratio of selected line intensities is $I(\lambda N_{391\ nm})I(\lambda Ti_{504\ nm})$ and $I(\lambda O_{777\ nm})/I(\lambda Ti_{504\ nm})$ respectively, The „control signals" being transmitted to two controllers that use a PID algorithm. The simultaneous use of two reactive gases and two reactive control loops poses additional difficulties, the process being prone to instabilities and oscillations induced by the independent control of each reactive gas. Therefore, the careful choice of PID control parameters is of crucial importance for the stability of the process. Both independent control, with each individual gas, and simultaneous control with both reactive gases is achieved.

The direct relation between the set parameters and the thin film properties is revealed by the composition variation, expressed as N/Ti, Ti/(O+N) or N/O ratios. The optical properties can be also tuned in wide range, in terms of reflected color, transmittance and absorbance, refractive index etc.

Process stability in 2 reactive gas process by using additional plasma emission

The fourth case study proposes an indirect way to probe the gas composition, without need to use the emission from the sputtering plasma. For this purpose, a Penning gauge connected to the vacuum chamber is used as excitation source for plasma emission, the collected spectra being used for the process control. The emission line intensity, for Ar or reactive gases such as N_2 and O_2, is pressure dependent, giving an opportunity to analyze the gas composition without need to collect light from the sputtering plasma. This gives an important advantage, since the collecting optics is protected from deposition during the process and the collection of emitted light from the pressure gauge is much easier. The control signals in this case have to be linked to the Ar emission as a reference, since there is no emission from the sputtered metal. The chosen emission line intensities ratios are in this case $I(\lambda N_{391\ nm})I(\lambda Ar_{750\ nm})$ and $I(\lambda O_{777\ nm})/I(\lambda Ar_{706\ nm})$. By using these signals, it becomes possible to sweep the entire hysteresis loop going from metallic mode to compound mode, for each individual reactive process. The nitrogen process shows no visible hysteresis effect, whereas the Oxygen process shows a pronounced hysteresis loop. By actively controlling the set parameter $I(\lambda O_{777\ nm})/I(\lambda Ar_{706\ nm})$ it become possible to establish stable functioning that correspond to the interior of the Hysteresis loop, giving access to a completely new process window that can be exploited.

Acknowledgment: This research was funded by a grant of the Romanian National Authority for Scientific Research and Innovation, CCCDI – UEFISCDI, project number COFUND-M-ERANET-3-HardCoat-1, no. 311/2022 (INOE), within PNCDI III, and by the Romanian Ministry of Research, Innovation and Digitalization through the National Plan of Research, Development and Innovation 2022-2027, Core Program, Project no: PN 23 05, Contract no: PN11N-03-01-2023, and through Program 1- Development of the National Research-Development system, Subprogram 1.2 - Institutional Performance - Projects to Finance the Excellent RDI, Contract no. 18PFE/30.12.2021

[1] Baptista, A.; Silva, F.; Porteiro, J.; Míguez, J.; Pinto, G. *Coatings*, ***8*** (2018) 402.

[2] Bräuer, B. Szyszka, M. Vergöhl, R. Bandorf, *Vacuum* **84** (2010) 1354–1359.

[3] S. Berg, H.-O. Blom, M. Moradi, and C. Nender, *Journal of Vacuum Science & Technology A* **7**, (1989) 1225.

[4] K. Strijckmans, R. Schelfhout, and D. Depla, *Journal of Applied Physics* **124**, (2018) 241101.

[5] I. Pana, C. Vitelaru, N.C. Zoita, M. Braic, *Plasma Processes and Polymers*, **13** (2016) 208-216.

[6] C. Vitelaru, I. Pana, A.E. Kiss, N.C. Zoita, A. Vladescu, M. Braic, Journal of Optoelectronics and Advanced Materials, **21** (2019) 717-725.

[7] G.Zambrano, H.Riascos, P.Prieto, E.Restrepo, A.Devia, C.Rincón, Surface and Coatings Technology, **172**, (2003) 2–3.