High Power Density Pulse Magnetron Sputtering - Process and Film Properties

Peter Frach*, Christian Gottfried, Fred Fietzke, Heidrun Klostermann, Hagen Bartzsch, Daniel Gloess

Fraunhofer-Institut für Elektronenstrahl- und Plasmatechnik FEP, Dresden
*corresponding author: Tel.: 0351-2586-370; Fax 0351-2586-55-370;

Abstract. In this paper specific advantages and disadvantages of different pulse magnetron sputtering processes (unipolar and bipolar pulse sputtering at high and very high power density including HIPIMS) as well as current and potential fields of application will be discussed.
On the examples of Ti and TiO_2 the typical effects and their influence on film properties occurring during the transition from classical medium frequency pulse magnetron sputtering to high energy pulse sputtering (HIPIMS) and the influence on film properties will be described. The discharge current density was varied between 0.2 and 3.5 A/cm^2. Aspects of energy feed-in, magnetron design and methods of reactive process control in the transition mode will be considered. Ideas for upscaling of the HIPIMS process will be discussed. Furthermore the influence of increasing ionisation on the occurrence of crystalline phases and on mechanical, optical and photocatalytic properties of the layers will be presented.

Keywords: HIPIMS, HPPMS, high power density pulse magnetron sputtering,TiO_2, reactive sputtering

1. Introduction

Since the idea of high power pulsed magnetron sputtering (HPPMS or HIPIMS) was born more than 10 years ago [1], there has been ever increasing interest within the scientific community to use this new technology, to carry out extensive R&D, and to clarify the mechanisms involved [2]. In some areas, the technology has attained special technical significance, for example for the coating of deep trenches in microelectronics by "Highly Ionized Metal Sputtering" [3]. First investigations regarding industrial use for hard coatings have been presented. In contrast, no applications for large surfaces have been developed, so far. In industry a certain reticence to use the technology, and to transfer the acquired knowledge to real processes, has prevailed.

HIPIMS, like conventional PMS, implies feeding energy pulses into a magnetron discharge – albeit with much higher power densities and hence drastically reduced values for the pulse-pause ratio and pulse frequency. Typically conventional PMS has power / current densities on the target during the pulse-on-time in the range 5 ... 50 W/cm^2 / < 0.5A/cm^2 for pulse-pause ratios of 1:1 and pulse frequencies between 20 kHz and 200 kHz. The relevant values for HIPIMS are 0.5 ... 5 kW/cm^2 / > 0.5A/cm^2 at pulse-pause ratios of 1:10 ...1:1000, in a frequency range of 50 Hz to 500 Hz. The high power density in the pulse leads to a high degree of ionization of the layer-forming particles, allowing the deposition of layers which are exceptionally dense, smooth, and homogenous. Besides this advantage, there are a number of disadvantages and in particular the drastically reduced deposition rate and the much higher tendency for arc discharges. Furthermore there are problems of up-scaling towards larger magnetron size due to limitations regarding powering and arc management.

In order to build up a knowledge base at Fraunhofer FEP for evaluating this technology and its potential for industrial use, the HIPIMS generator TruePlasma HighPulse 4008 (Trumpf Hüttinger Elektronik) with performance parameters 20 kW DC, 2 kV / 4 kA pulse at max. 500 Hz , was compared with established MF pulse generators under a wide range of conditions (coating plant, magnetron type, target material, pressure, gas composition). The discharges themselves, the possibilities of reactive process control and also the properties of the deposited layers were evaluated.

2. Experimental

Two different sputtering systems were used for this investigation.

System 1 is the Double Ring Magnetron sputter source DRM 400 on the Cluster 300 plant [4, 5]. The inner target of the DRM 400 was powered against the anode (Fig. 1a). The experiments in the unipolar HIPIMS mode were carried out using the TruePlasma HIPIMS generator. The relatively small target size of 116 cm^2 allowed to achieve a maximum power density up to 4kW/cm^2. For comparison the pulse unit UBS-C2 of Fraunhofer FEP was used for the normal unipolar pulse mode. Amongst the materials which were studied, comparative tests were carried out with a Ti target, with and without reactive gas.

System 2 is a Double Magnetron System DMS 500 with two 120 x 500 mm^2 rectangular targets in the batch coating plant UNIVERSA. The DMS 500 was powered by a voltage-fed rectifier made by MagPuls (60 kW DC, 1 kV / 1 kA pulse at max. 50 kHz). The power was introduced in the bipolar pulse mode, where the polarity of the two magnetrons is switched in each half cycle (Fig. 1b). By varying the duty cycle between 90% and 14%, the

current densities in the pulse changed between 0.2 and 1.2 A/cm^2, i.e. between normal bipolar pulse mode to bipolar HIPIMS-mode.

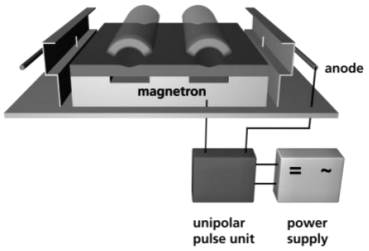

Figure 1a. Schematic of the unipolar powering configuration of the inner target of the DRM 400 against the anode in the normal pulse and in the HIPIMS mode

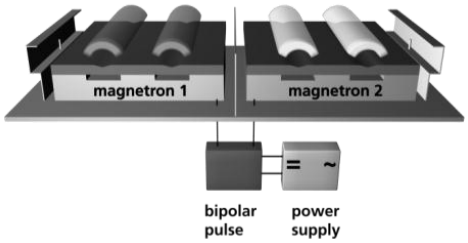

Figure 1b. Schematic of the bipolar powering configuration of the two targets of the DMS 500 in the normal pulse and in the HIPIMS mode

3. Results and discussion

3.1. Ti and TiO_2 layers deposited with DRM400 (system 1)

The DRM 400 has moveable magnet systems so that the dependence of the energy input in HIPIMS mode on the magnetic field strength could be studied in detail. It was found that the power density on the target is strongly dependent on the magnetic field strength for sputtering in pure argon (Fig. 2). The maximum power density in the pulse increases from 2.4 kW/cm^2 to 4 kW/cm^2 (67% increase) when the magnetic field strength is increased by 15%, only. To achieve high power densities, strong magnetic fields are therefore favorable.

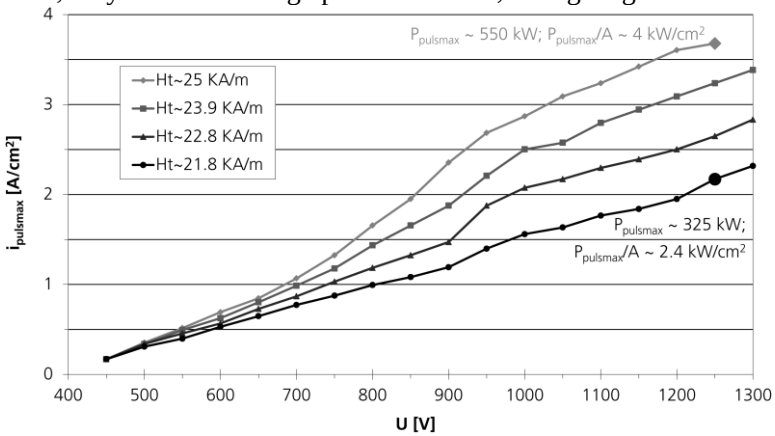

Figure 2. Maximum discharge power density $i_{pulsmax}$ in the pulse for a Ti target (magnetron DRM 400) as a function of the discharge voltage U at different magnetic field strengths (p = 1 Pa Ar, f = 12 Hz, t_{on} = 200 µs). The values in the figure for pulse power density $P_{pulsemax}/A$ of 2.4kW/cm^2 and 4kW/cm^2 correspond to the enlarged data points of the lower and the upper curve, respectively

In the HIPIMS mode, the Ti coating rates are only between 10 to 14% of the coating rates in normal pulse mode at the same average power. This could be confirmed in the reactive mode for TiO_2-films (about 17%, see Tab. 1). Furthermore, it was found that the thermal substrate load per unit layer thickness was comparable to that of the PMS process. The temperature of glass substrates started at room temperature and raised to about 190°C±5°C for normal pulse mode and HIPIMS mode after depositing 1 µm Ti-layer. The deposition time in the HIPIMS mode was by a factor of about 7 longer than in the unipolar pulse mode. As a consequence of this, the substrate temperature would be much higher in the HIPIMS mode if the deposition rate could be improved using higher power level or more process stations. This needs be considered in the case of temperature sensitive substrates.

The following results were achieved from experiments in reactive mode with a Ti target in argon-oxygen atmosphere. In HIPIMS mode, the reactive process showed hysteresis, similar to the normal PMS process. It has been operated in the transition mode using optical plasma emission spectrometry as described below.

The maximum achievable coating rate for stoichiometric TiO_2 in the HIPIMS mode is significantly lower than that of normal PMS. It is only 5% (fully reactive) to 17% (highest rate for stoichiometric films in the transition mode) of the deposition rate in normal pulse mode. The TiO_2 layers deposited in the HIPIMS mode are much finer grained than those deposited in the normal pulse mode. Starting from the substrate, they grow first in amorphous phase and then in the crystalline rutile form. Similar behavior was observed for TiO_2 layers deposited in normal bipolar pulse mode or in pulse packet mode [6]. The TiO_2 layers deposited in the HIPIMS mode have, however, a higher hardness of up to 20 GPa compared to a maximum of 16 GPa for those deposited in pulse packet mode. They also have a somewhat higher refractive index n than the layers deposited in normal pulse mode [7].

Table 1. Layer properties for different pulse modes and different reactive working points for TiO_2 deposition

unipolar pulse mode	working point	layer thickness [nm]	deposition rate [nm/min]	hardness [GPa]	n @ 550 nm	k @550 nm
normal	stoichiometric	560	110.0	~7.0	2.480	$< 10^{-5}$
HIPIMS	substoichiometric	848	28.0	20.0	2.552	$4.70 \cdot 10^{-3}$
HIPIMS	fully reactive	180	6.0	16.5	2.514	$4.20 \cdot 10^{-3}$
HIPIMS	stoichiometric (transition mode)	675	18.4	19.1	2.565	$2.95 \cdot 10^{-3}$

3.2. TiO_2 layers deposited with DMS 500 (system 2)

The experiments with DMS 500 were carried out to investigate the properties of the process and of the films during gradual transition between normal pulse mode and HIPIMS mode. By varying the duty cycle between 90% and 14% the current densities in the pulse changed from 0.2 to 1.2 A/cm^2 (Tab. 2). The value of 0.5 A/cm^2 was reached at a duty cycle of about 33%, i.e. the discharge is in the HIPIMS mode for smaller duty cycle. The optical plasma intensity of different lines was investigated in dependence on duty cycle (Fig. 3) showing a monotonous variation. It is assumed that the reduction of Ar I-line might be due to gas rarefaction effects. The reduction of Ti I-line corresponds to the deposition rate reduction.

Table 2. Duty cycle, frequency and current densities of Ti layers deposited with DMS 500

Duty cycle [%]	90	50	33	20	14
frequency [kHz]	10	5	3.3	2	1.4
current density [A/cm^2]	0.2	0.3	0.55	0.8	1.2

The reactive process was controlled using the plasma control unit S-PCU (Fraunhofer FEP) having closed feedback system for the piezo valve of the oxygen gas inlet. A spectrometer inside S-PCU was used and 2 lines were chosen: Ti 500 nm and O_2 777 nm. The intensity ratio of these two lines was used as control value for the reactive gas flow. By controlling the process in this way, the discharge can be stabilized within any desired working point and stoichiometric TiO_2 layers can be deposited in both normal pulse mode and in HIPIMS mode. Reduced hysteresis was observed for lower duty cycles (Fig. 4), but the hysteresis does not disappear.

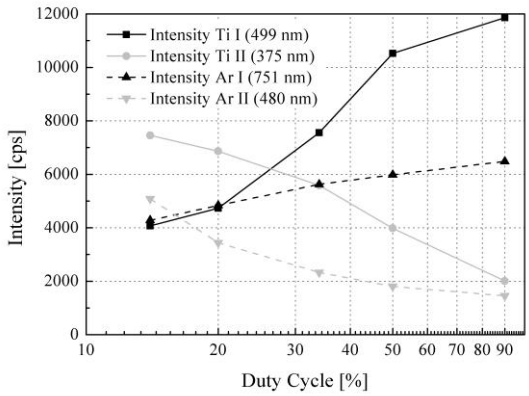

Figure 3. Optical plasma intensity vs. duty cycle for Ar I and II as well as for Ti I and II

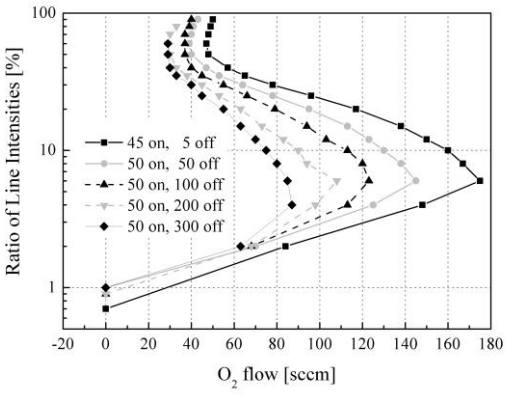

Figure 4. Ratio of line intensities (in %) vs. oxygen flow within the hysteresis region of the reactive sputtering process of TiO_2 for different parameters of duty cycle

Rotating substrates (X5CrNi18.10) were coated with TiO_2 at different duty cycles. At high duty cycle (normal pulse mode) clear facetted anatase crystallites were found (Fig. 5). At small duty cycle a reduction of crystal size and a growing rutile fraction as a second phase with lower roughness were observed.

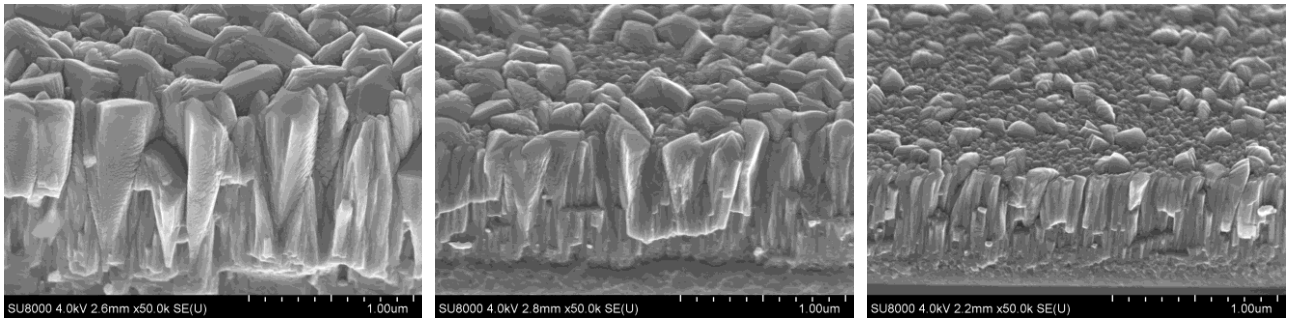

Figure 5 a-c. SEM images of TiO_2-layers at different duty cycle (left: 90%, middle: 34%, right: 14%)

Keeping the total power constant, the deposition rate decreased to about one third at a duty cycle of 14% (Fig. 6). The Ti I-line intensity behaves in the same way representing the number of sputtered Ti atoms in the plasma. The roughness reduces to about 53%, which corresponds to the change of microstructure and apparent roughness in the SEM images. The hardness rises from 8.7 GPa (typical value for anatase) at 90% duty cycle to 16 GPa (typical value for rutile) at 14% duty cycle. The elastic modulus correlates well with the hardness and changes from 160 GPa to 210 GPa, respectively. Furthermore the refractive index increases from 2.35 to 2.65 as a consequence of the growing fraction of rutile phase in the film. Another indication of the reduced anatase fraction is the decreasing photocatalytic activity derived from methylene blue decomposition measurements [to be published elsewhere].

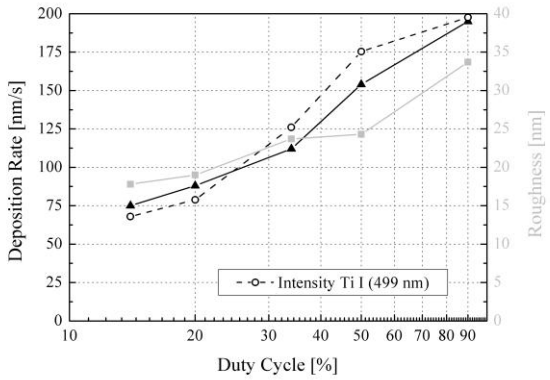

Figure 6. Deposition rate, roughness and Ti I-line intensity vs. duty cycle

4. Summary

The studies on TiO_2 have shown that HIPIMS is a promising technology for achieving layer properties that cannot be achieved using other sputtering techniques. These properties include a higher hardness and fine crystallinity. The disadvantages of the technique are in particular the strongly reduced coating rate at higher power density and the worse process stability. Given the current state of HIPIMS generators, laboratory scale process development should ensue. For the further development of the technology, suitable magnetron sputter sources with a high magnetic field and power supplies with sufficient performance to allow up-scaling of the process are required. This will then enable HIPIMS processes to be developed for industrial applications.

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