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BRASS SMELTING DUST AS A SOURCE OF ZnO IN THE PRODUCTION OF TARGETS USED IN MAGNETRON SPUTTERING THIN FILM DEPOSITION

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ABSTRACT
The feasibility of using a zinc oxide rich waste (ZnO>95%), dust originated in the brass smelting industrial process, as a raw material to produce targets to be used in radio-frequency magnetron sputtering (RFMS) deposition of ZnO thin films was investigated. The ZnO waste was characterized in terms of chemical and mineralogical compositions and particle size distribution. The targets were prepared by uniaxial pressing of the ZnO waste followed by sintering in air at a temperature in the range 900-1200 ºC. The density of sintered targets increased with sintering temperature, and zincite (ZnO) was the predominant crystalline phase identified by X-ray diffraction (XRD). Scanning electron microscopy (SEM) revealed zincite grains, with an average size ~2 μm and a non-homogeneous microstructure due to the presence of dense aggregates. Preliminary MS thin film deposition tests on a glass substrate produced transparent nanostructured ZnO thin films with a homogeneous microstructure. Research work is underway to process ZnO waste-based targets with an improved microstructure in order to obtain RFMS deposited ZnO thin films with optimized final electrical and optical properties.

Keywords: ZnO waste, zincite, ceramic target, sintering, thin film, optical properties.

INTRODUCTION
Recycling waste is considered as the most important activity to preservation of natural resources on the substitution of critical raw materials that are increasingly used in emerging technologies, as it was identified by European Comission. Apart from the economic benefits, recycling waste is mandatory due to environmental and social impacts. Therefore, recycling of wastes through the development of innovative methodologies that turn waste into added-value products is of paramount importance helping to change waste into wealth.
Recycling of metal oxide rich wastes from different sources has been normally achieved by incorporation in civil construction products. In fact, the use of metallurgical slags, coal fly ashes and bottom ashes as raw materials to obtain structural materials has been investigated by some of the authors [1-4]. All the developed materials can be used by construction industry and hence are good options for the management of such solid wastes in an environmentally correct way.
In this work, an innovative recycling process was applied to ZnO rich waste aiming to investigate the transformation of this industrial waste into valuable advanced functional materials potentially useful in the fields of optoelectronics and photonics. This waste comes from the brass melting industrial process, where it is filter-collected as dust. The production in Portugal estimated as about 100 ton/year, and it is sent to landfill posing serious environmental and health issues.
The processing and characterization of ceramic targets from this non-conventional ZnO source and the use of these targets to obtain ZnO thin films by RFMS deposition were investigated together with the study of films properties. Research will be continued in order to optimize processes and materials.

MATERIALS AND METHODS

The chemical and mineralogical compositions of the as-received waste powders were analyzed by X-ray fluorescence (XRF) and X-ray diffraction (XRD), respectively. ZnO was the major component (96 wt%), together with CuO (2 wt%), SiO_2 (0.7 wt%), PbO (0.3 wt%), Fe_2O_3 (<0.2 wt%), and other minor oxides (each of them <0.1 wt%). XRD revealed that zincite (ZnO) was the predominant crystalline phase present in the waste. Particle size analysis by a laser granulometer indicated a particle size totally lower than 3 μm. The powders were uniaxially pressed, using a pressure of ~110 MPa, and green targets with ~60 mm diameter and ~5 mm height were obtained (Fig. 1). The targets were sintered in air using an electric furnace at a maximum selected temperature in the range 900-1200 ºC for 2 h, and then the sintered targets were characterized by XRD and SEM. The sintered targets (with a final diameter of 50 mm) were used to deposit thin films on glass substrates by RFMS. The films were characterized by XRD and SEM and the optical and electrical characteristics of the obtained transparent films were investigated.

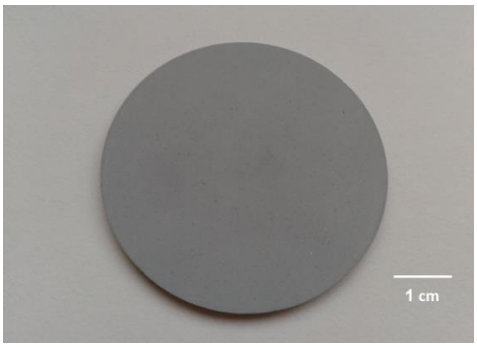

Fig. 1- Target of ZnO waste

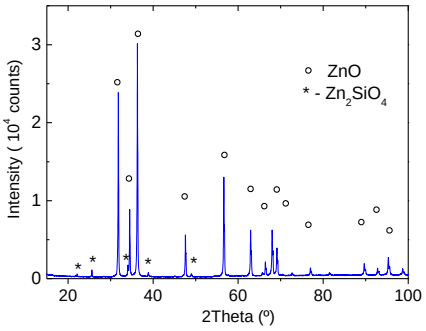

Fig.2- XRD results for ZnO-waste sintered target

RESULTS AND DISCUSSION

After sintering at 900 ºC, the powder compacts exhibited still some crumbling, while after sintering at 1200 ºC, a linear shrinkage of ~9% was observed and a well consolidated powder compact was obtained. XRD results for ZnO-waste target sintered at 1200 ºC, shown in Fig. 2, indicated that zincite (ZnO) was the predominant crystalline phase together with some peaks of zinc orthosilicate (Zn_2SiO_4). SEM observations of targets sintered at 1200 ºC (Fig. 3) revealed a non-homogeneous microstructure that is caused by the presence of highly dense micro-aggregates of zincite grains, as illustrated at high magnification in Fig.4. Targets sintered at 1200 ºC were robust enough to be easily manipulated and were selected to prepare thin films on glass substrates by RFMS deposition at room temperature, under similar conditions as used in previous work with commercial ZnO targets [5]. Transparent ZnO thin films were produced (Fig. 5), and the change of transmittance as a function of wavelength for films obtained at different powers is shown in Fig. 6. The structural characterization of the films was performed by XRD and the results revealed the presence of a very sharp diffraction peak (Fig. 7) that corresponds to the (002)plane peak of zincite crystals, which indicates a preferred orientation along the c-axis perpendicular to the substrate surface due to the self-texturing mechanism. Microstructural characterization of the thin films by SEM demonstrated a nanostructured microstructure containing rather uniform ZnO nanocrystals as shown in Fig. 8.

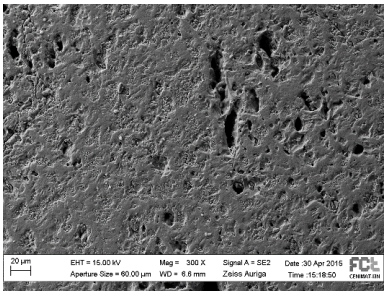

Fig. 3- SEM of a target sintered at 1200 ºC

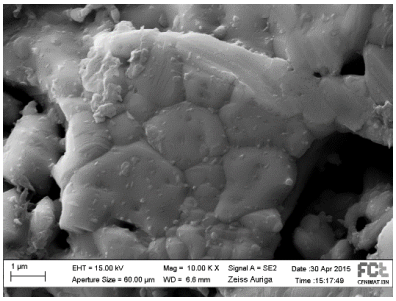

Fig. 4- Detail of SEM at high magnification

Fig. 5- Glass substrate with a transparent ZnO thin film deposited by RFMS

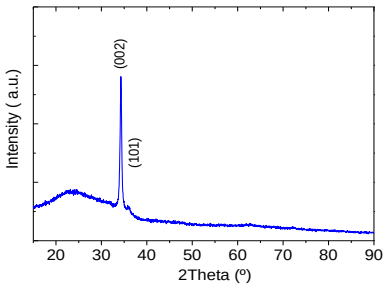

Fig. 6- XRD results of produced thin film

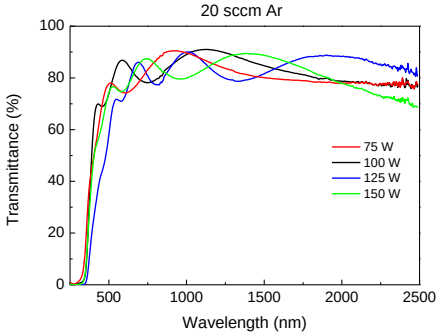

Fig. 7 – Optical characteristics of thin films produced from a ZnO-waste target

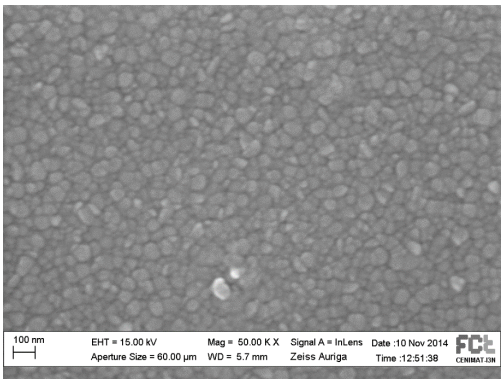

Fig. 8 – SEM of thin film produced from a ZnO-waste target

CONCLUSIONS

The possibility of producing targets from a ZnO rich waste and their application on the processing of ZnO nanostructured thin films by RFMS deposition have been demonstrated. Further experimental work should be developed in order to obtain homogenous microstructures of the sintered targets, avoiding the formation of dense micro-aggregates. The effect of dopant addition (eg Al_2O_3) to ZnO waste should also be investigated in order to achieve final optically transparent thin films exhibiting simultaneously adequate electrical properties to be used in optoelectronics.

Acknowledgments

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