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## Original Article

## Thermo-physical properties of as deposited and aged thermal barrier coatings (TBC) for gas turbines: State-of-the art and advanced TBCs

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## ABSTRACT

In the perspective of fuelling the future generations of gas turbines by hydrogen rich syngas, the evaluation of the effect of a higher water vapour content into the flue gases on the TBC used, or potentially usable, is a need. For this purpose YPSZ APS TBC with two different microstructures have been exposed for 500 h at different temperatures in the range 1000 °C–1250 °C either in air and air + 20% vol. H<sub>2</sub>O. The comparison between the different testing conditions has been performed in terms of sintering kinetics and phase stability, as evaluated by thermal diffusivity measurements and Synchrotron X-Rays diffraction, respectively. Furthermore the characterisation of thermal properties of two innovative TBCs (GZO-YSZ and YAG) potentially able to withstand the CMAS attack and erosive environments, respectively, has been carried out.

No clear evidence of a different behaviour of TBC has been observed, at least in the considered aging time and temperature range.

## 1. Introduction

Owing to climate changes, greenhouses gases emitted by fossil fuelled power generation plants should be drastically reduced. On the other hand, power plants are still necessary to guarantee the grid stability when the share of non-programmable renewables is predominant and enable the transition towards a green power generation, waiting for economically feasible energy storage technologies, such as electrochemical, mechanical, thermal and gravitational. In this perspective, Carbon Capture and Sequestration (CCS) is a viable option to minimise CO<sub>2</sub> emissions. Among the different potentially applicable solutions, pre-combustion CCS is attractive because of the relatively low energy consumption.

Integrated Gasification Combined Cycle (IGCC) is currently one of the most attractive technologies for the high-efficiency use of coal. It enables the conversion of coal and other solid or liquid fuels to a gaseous syngas fuel, while still maintaining aggressive emissions targets

and high efficiency. Pre-combustion methods are the preferred means of capturing CO<sub>2</sub> in IGCC systems. An IGCC plant equipped with pre-combustion CO<sub>2</sub> capture combined with low emissions of other gases, e.g. NO<sub>x</sub>, SO<sub>x</sub>, can be realised through major advancement in gas turbine technology.

The main aim of this future technology is to use an undiluted hydrogen-rich syngas as fuel, consequently, water vapour formed from H<sub>2</sub> in the combustion process along with differences in water input for various gasification processes (e.g. dry feed v. wet feed) result in significant differences in water vapour levels in the combustion gases flowing through IGCC turbine hot sections.

This can result in differences in hot section component degradation since increased water vapour levels have increased oxidation rates in laboratory tests of turbine materials [1].

Recently, B. Pint et al., M.H. Sullivan et al. and W. Nowak experimentally studied the effect of the increased steam content on TBC life [2–7].

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In particular, Pint and co-workers observed a significant reduction of Air Plasma Spray (APS) TBC life by cyclic oxidation tests when testing environment changed from dry air to air +10% water vapour. On the other hand, a further increase of water vapour to 50% did not affect TBC life significantly. Although no obvious impact on bondcoat and thermally grown oxide composition and microstructure was relieved after TBC failure in most of the cases, when exposition time was sufficiently long, some differences in TGO microstructure and TBC failure modes were observed providing some indications to explain the outcomes in terms of TBC lives [2]. When samples without the ceramics top layer were considered, no clear effect of water vapour content on TGO was noticed [3]. Furthermore in Pt-diffusion and Pt-modified aluminide bondcoats, in air +10% water vapour more  $\theta$ - $\text{Al}_2\text{O}_3$  formation and small alumina morphology and microchemistry differences compared to dry air are reported [4,5].

M.H. Sullivan and D. R. Mumm observed that at early oxidation stages the presence of higher water vapour partial pressures promoted and enhanced the (Ni, Co)(Al, Cr) $_2\text{O}_4$  spinel formation, on top of Co/Cr rich phases ( $\gamma$  phase) and even in correspondence to Al rich phase ( $\beta$ -phase) that is supposed to affect at later stages the bondcoat stability, the TGO development and TBC lifetime [6].

W. Nowak argued that the observed lifetime shortening by water vapour (20% vol.) on APS TBC samples could be correlated with an effect of water vapour on crack propagation in the ceramic topcoats. The proposed explanation was that the non-uniform Y distribution across powder particles could promote and enhance the transformation from tetragonal to monoclinic zirconia in the presence of water vapour at temperature below approximately 400 °C during cooling [7].

The aim of this work is to evaluate, if any, the effect of water vapour content in the flue gases on the state-of-the-art TBC in terms of sintering kinetics and/or phase stability by using thermo-physical and synchrotron radiation diffraction, respectively.

Furthermore, thermo-physical characterisation of advanced TBCs candidate for future applications also in next IGCC generation gas turbine hot path has been provided, as well. In particular, based on results from the literature [8–11] a two-phase TBC made of Gadolinium Zirconate (GZO) and YPSZ and a two-layer TBC made of YAG-YPSZ have been selected as potentially able to withstand CMAS attack and erosive environments.

## 2. Experimental

As representatives of YPSZ state-of-the-art TBC one highly porous and one dense vertically cracked TBC have been selected.

### 2.1. Sample deposition

#### 2.1.1. Highly porous and dense highly cracked YPSZ APS TBC

The powder for spraying freestanding coatings is a standard commercial  $\text{ZrO}_2$ -7Y $_2\text{O}_3$  TBC (Amperit 827 produced by H.C.Starck). The TBC porosity (see Fig. 1 and Table 1) produced by Image Analysis (IA) on as sprayed highly porosity and dense highly cracked samples resulted equal to an average value of 25–30% and 3–4%, respectively [11].

#### 2.1.2. Single YSZ-GZO composite and bilayer TBC of porous YSZ with top layer of YSZ-GZO composite

The single layer YSZ-GZO consists of a layer of ceramic composite nominally from 50% wt. 8YSZ and 50% wt. gadolinium zirconate (GZO) of  $500 \pm 50 \mu\text{m}$  thickness deposited with standard Atmospheric Plasma Spray (APS) equipment (Oerlikon Metco TriplexPro-210™).

The ceramic bilayer consists of two layers  $250 \pm 50 \mu\text{m}$  each; the internal layer is made of 8YSZ and an outer layer is nominally identical to the single layer composite. Powders used have been commercially available HOSP 8YSZ (Metco™ 204NS) and a blend of same 8YSZ powder with GZO powder of spherical shape (proprietary specification

produced by H.C. Starck), respectively [8,11]. Fig. 2 shows the SEM images of the microstructures of these advanced TBCs. Porosity resulted in the range 14–16% and 15–18% for single and bilayer samples, respectively. The GZO phase ranges from 42–47% and from 23–27% for single and bilayer TBCs (see Table 2).

#### 2.1.3. YAG

The single YAG (yttrium aluminium garnet) TBCs samples have been deposited with standard APS equipment (Oerlikon Metco TriplexPro-210™). The powder was synthesized by Treibacher (Austria) and agglomerated by spray drying at Forschungszentrum Jülich, respectively [11]. Spraying has been performed with low substrate temperature - resulting in an widely amorphous YAG layer in the as-deposited condition. Subsequent heat treatment at 900 °C initiated crystallization and formation of microscopic and “homogenous” crack network to form the brick-like structure, as pointed out in Fig. 3.

State of the art YPSZ TBC samples have been aged in air (R286&R320) and in air +20% vol. steam (R287&R267) at temperatures ranging from 1000 °C up to 1250 °C for 500 h to evaluate the effect of the aging atmosphere. This volumetric percentage of steam has been considered representative of the steam content in flue gases produced by the combustion of Hydrogen rich syngas. In particular, tests have been performed on freestanding coatings in order to study the sintering kinetics avoiding the superposition of other effects affecting thermal conductivity/diffusivity of TBC such as crack nucleation and propagation and bondcoat oxidation at the interface between bondcoat and TBC.

For this purpose all TBCs have been deposited on steel substrates which were priorly coated with a thin layer sodium chloride. The sodium chloride layer could be easily dissolved in water to obtain free-standing samples of the ceramic coatings. Advanced YPSZ + GZO TBCs have been characterised both in as sprayed conditions and after heat treatment in air at temperatures ranging from 1000 °C up to 1250 °C for 500 h. As concerns YAG TBCs measurements have been performed both before and after an heat treatment (900 °C for 24 h) finalised to promote the transition from amorphous to crystalline. As coating transforms from “dense” amorphous to brick-like crystalline this has a remarkable effect on thermal properties.

### 2.2. Microstructural characterisation

“The microstructural characterisation of TBCs was performed by scanning electron microscopy (SEM) (MIRA XMH, Tescan, Brno, CK). TBC specimens were impregnated in vacuum using epoxy resin. The TBC thickness has been evaluated as the average of 30 values measured by (IA) (Screen Measurement, Laboratory Imaging Ltd., Praha, CZ) along the whole sample section. The overall porosity has been evaluated from ten 700x back-scattered electron images (BEI) taken along each sample cross-section” [12].

“Pores have been classified depending on their elongation  $e$  (defined as the ratio between the maximum and the minimum Feret diameters of the object) and orientation in respect to the heat flux direction. In particular, as explained in details elsewhere [12], all pores having an elongation  $e \leq 2$  have been considered as “spherical” while those with  $e > 2$  have been considered lamellar shaped”.

For dual phase samples, the volumetric fraction of the both phases have been estimated by BEI images used for porosity evaluation.

Tables 1 and 2 summarise the name of samples, the main microstructural parameters (i.e. overall thickness and porosity, as measured by IA) and the heat treatment for the state-of-the-art and the advanced TBC samples, respectively. Furthermore, Tables 3 and 4 summarise detailed microstructural features, as well as phase composition for the state-of-the-art and advanced GZO + YPSZ and YAG samples, respectively. As quantitatively summarised in the previously mentioned tables and as also depicted by Fig. 4, the high temperature aging reduced the porosity content by a different extend depending on the microstructure

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