

# Study of the evolution of the residual stress state in thermal barrier coatings sprayed on AISI 304 stainless steel

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## Abstract

Thermal barrier coatings (TBC) were manufactured by thermal spraying of CoNiCrAlY and YSZ powders, which constituted the bond coat (BC) and top coat (TC), respectively. The BC was deposited on stainless steel substrates by a high velocity oxygen fuel (HVOF) system whilst the TC was deposited on the BC by an atmospheric plasma spraying (APS) gun. TBCs were heat treated at 1200 °C for 8 and 16 h in order to determine the evolution of the residual stress state as a function of time. The residual stress profiles were obtained by employing the Modified Layer Removal Method (MLRM) and an adaptation of this for bilayer coatings called Modified Layer Removal Method for Duplex Coatings (MLRMDC). It was observed that as the exposure time was increased, the compressive residual stresses increased, which was more noticeable in the BCs. The TBCs without heat treatment revealed tensile residual stresses.

**Keywords:** thermal barrier coating, residual stress, modified layer removal method, thermal spraying, Modified Layer Removal Method for Duplex Coatings.

# Estudio de la evolución del perfil de esfuerzos residuales en recubrimientos barrera térmica depositados sobre acero inoxidable AISI 304

## Resumen

Se fabricaron recubrimientos barrera térmica depositando polvos de CoNiCrAlY y YSZ, los cuales constituyeron las capas de enlace y superior, respectivamente. La primera se depositó sobre el acero inoxidable AISI 304 mediante un sistema de oxígeno combustible de alta velocidad; mientras que la segunda se depositó mediante un sistema de rociado por plasma atmosférico. Los recubrimientos se trataron térmicamente a 1200 °C por 8 y 16 h con el fin de determinar la evolución del estado de esfuerzos residuales en función del tiempo. Los perfiles de esfuerzos residuales se determinaron empleando el método de remoción de capa modificada y una adaptación de éste para recubrimientos bicapa, llamado método de remoción de capa modificada para recubrimientos dúplex. Se encontró que conforme el tiempo de exposición se incrementó los esfuerzos residuales de compresión incrementaron, siendo más notorios en las capas de enlace. Los recubrimiento sin tratamiento térmico revelaron esfuerzos residuales de tensión.

**Palabras clave:** recubrimiento barrera térmica, esfuerzos residuales, método de remoción de capa modificada, rociado térmico, método de remoción de capa modificada para recubrimientos dúplex.

## 1. Introduction

Thermal barrier coatings (TBC) are commonly employed in hot sections of aeronautic and land turbines to provide protection against hot corrosion and oxidation of critical components [1,2].

A TBC is formed by two layers of different materials deposited on a metallic substrate. The external layer, also known as the top coat (TC), is commonly formed by yttria stabilized zirconia (YSZ), a ceramic with low thermal conductivity, good thermal-shock resistance and a relatively low thermal expansion coefficient [3-5].

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5]. On the other hand, the layer in contact with the substrate, known as the bond coat, is normally fabricated from a MCrAlY alloy ( $M = \text{Co, Ni, or Co+Ni}$ ). Such a layer promotes good adherence of the TC to the substrate, as it has a thermal expansion coefficient similar to that of the substrate [7]. It also provides protection against oxidation [5,8] and hot corrosion [2,8] by forming a stable alumina thermally grown oxide (TGO) at the interface with the TC. TBCs can be processed entirely by thermal spraying; the BC is normally produced by employing the HVOF process, whereas the ceramic TC is fabricated by high temperature processes, such as atmospheric plasma spraying (APS). Both processes are cost-effective and produce good quality TBCs[9].

Residual stresses are always present in thermal spray coatings. They are produced by two known mechanisms: i) quenching of splats after impact the substrate and restricted contraction of them by adherence to the substrate; and ii) differential thermal expansion of the coating and the substrate during cooling. The stress state of a coating is important as it has a strong influence on its mechanical properties, which, in turn, affect its performance and durability [10, 11]. According to Widjaja *et al.* [12], the final stress state of a TBC is influenced not only by quenching of splats and thermal mismatch but also by phase transformation during coating deposition.

In the HVOF process the particles projected onto the substrate reach high velocities, and temperatures lower than those measured in the APS process, which in contrast produces a low velocity and high temperature plume, adequate for the deposition of ceramic powders. Despite those differences, it has been suggested that the coatings processed by each process to make a TBC, i.e. the BC and TC, respectively, have a compressive residual stress state [13], which opposes to the tensile stresses that are responsible for coating delamination during service [14].

Another important variable when processing a TBC by thermal spraying is substrate roughness. A substrate having high roughness promotes a compressive residual stress state in the BC, which improves the cracking resistance of the TBC [15]. Therefore, it is important to promote a compressive stress state in a TBC as a tensile stress state increases the coating susceptibility to delamination and cracking [16].

During the last decade, different techniques have been proposed to study the final stress state of TBCs that are produced by thermal spraying: x-ray diffraction [6, 16], indentation [17], finite element simulation [18, 20], material removal [21,22], hole drilling [23], 3D imaging correlation [24], and curvature evolution [17]. In this work, the residual stress state was evaluated by employing the Modified Layer Removal Method (MLRM) [25]. The method is based on the monitoring of strains in the coating after removing thin layers by fine grinding. This technique was developed for mono-layer systems; however, the method has also been employed in duplex systems but only considering the physical properties of one material in the calculations [21]. Here, the MLRM technique was modified in order to calculate the residual stress state of a TBC employing the physical properties of the BC and TC substrates. This modified procedure was named as the Modified Layer Removal Method for Duplex Coatings (MLRMDC). A comparison of the residual stress state of a TBC, with and without heat treatment at 1200 °C, obtained by employing the MLRM and MLRMDC methods, is presented in this work.

## 2. Materials and Methods

In this investigation, the MLRM and MLRMDC methods were used to calculate the residual stresses profiles of TBCs prior and after heat treatment at 1200 °C for 8 and 16 h. The following section describes each experimental stage.

### 2.1. Deposition of TBCs

Three TBCs were deposited on stainless steel AISI 304 substrates of 25.4 mm X 25.4 mm X 6.35 mm in size. The BC and TC were obtained by deposition of powders of a CoNiCrAlY alloy (AMDRY 9954, Co32Ni21Cr8Al0.5Y), and yttria stabilized zirconia (NS-204, YSZ), respectively: both from Sulzer Metco. Before deposition, the substrates were cleaned with acetone and grit blasted by an air stream carrying alumina particles that impacted at a 45° angle at a distance of 200 mm, they were then re-cleaned with acetone; the surface roughness ( $R_a$ ) ranged from 3 to 7  $\mu\text{m}$ . The BC was deposited by a HVOF system, model DJH2700 by Sulzer, with a powder feed rate of 38 g/min and a distance of 203 mm. The mean thickness of this layer was  $180\pm 5 \mu\text{m}$ . On the other hand, the TC was deposited by an APS system, model 9MB by Sulzer, with a powder feed rate of 45 g/min and a distance of 120 mm. The potential and current of the gun were set at 67 V and 600 A, respectively. The mean thickness of this layer was  $120\pm 3 \mu\text{m}$ . Each layer was built-up applying 16 passes at a gun lateral displacement speed of 1.5 m/s. Table 1 shows the pressures and flow rates of the gases employed.

### 2.2. Heat treatment of coatings

Once the TBCs were deposited, two samples were heat treated at 1200 °C using a closed muffle (by Nabertherm) in argon atmosphere, one for 8 h and the other for 16 h, Table 2. The heating rate was 6 °C/min. The sample without heat treatment (WT) was used for comparison.

### 2.3. Fixing and soldering of strain gauges

After heat treatment, the back faces of the samples were carefully ground and cleaned to eliminate oxide and dirt, thus ensuring an adequate attaching of the strain gauges. These devices were supplied by Vishay Precision Group Inc.

Table 1.  
Pressure and flow rate of the gases.

	Pressure (bar)	Flow rate (SLPM)
Oxygen	10	154
Propane	8.9	38
Air	6.8	294
APS	Pressure (bar)	Flow rate (SLPM)
Argon	5.2	42
Hydrogen	3.4	7

Source: The authors

Table 2.  
Heat treatment of samples.

Sample	Temperature (°C)	Time (h)
8H	1200	8
16H	1200	16
WT	Without treatment	

Source: The authors

(model CEA-09-125UT-350). The gauges were joined to connecting wires by applying a tin-based solder. The connection verification and strain measurements were carried-out by means of a strain meter model P3500, also provided by the same supplier.

#### 2.4. Residual stresses determination by MLRMDC

The method followed for the removal of thin layers of the TBC coating is described elsewhere [21,25]. The physical properties of all materials involved were considered when determining the residual stresses; stainless steel AISI 304 for substrate, CoNiCrAlY for BC, and YSZ for TC, Table 3.

The mathematical analysis of the stresses was performed using only a quarter of the sample, as shown in Fig. 1. The force and momentum acting on the removed layer along and around the  $X$  axis are  $F_x$  and  $M_x$ , respectively. Thickness of the removed layer, remaining layer, substrate, BC, and TC are  $h$ ,  $h'$ ,  $H$ ,  $h_{BC}$  y  $h_{TC}$ , respectively. The strain gauge was attached on samples' back face;  $b_x$  and  $b_y$  are half of both length and width of the actual sample. Plane located at  $z = 0$  is the interface substrate/BC.

Strain changes of the remaining part of the sample, after removing a thin layer by fine grinding, are given by:

$$\varepsilon_x = \varepsilon_{x0} + k_{xz} \quad \text{and} \quad \varepsilon_y = \varepsilon_{y0} + k_{yz} \quad (1)$$

Where  $k_{xz}$  and  $k_{yz}$  are sample curvatures.

For a plain stress state, the stress-deformation equation is given by:

$$\begin{Bmatrix} \sigma_x \\ \sigma_y \end{Bmatrix} = E' \begin{bmatrix} 1 & v \\ v & 1 \end{bmatrix} \begin{Bmatrix} \varepsilon_x \\ \varepsilon_y \end{Bmatrix} \quad (2)$$

Where  $E'$  is a relationship that involves the Young modulus, and  $v$  the Poisson ratio of the materials.

$E_s$  is the Young modulus of the substrate,  $E_{BC}$  of the BC, and  $E_{TC}$  of the TC:

$$E_s = \frac{E_s}{(1-v_s)}, E_{BC} = \frac{E_{BC}}{(1-v_{BC})}, \text{ and } E_{TC} = \frac{E_{TC}}{(1-v_{TC})} \quad (3)$$

Where  $E_t$  is the overall Young modulus of the TBC:

$$E_t = E_{BC} + E_{TC} \quad (4)$$

If  $\sum h \geq h_{TC}$ , then  $E_{TC} = 0$ , but if  $\sum h \geq h_{TC} + h_{BC}$  then  $E_{TC} = E_{BC} = 0$ .

Table 3.  
Physical properties of the TBC materials.

Property	CoNiCrAlY [26]	YSZ [10]	AISI 304 [27]
Young modulus (GPa)	200	70	200
Poisson ratio	0.30	0.23	0.29

Source: The authors

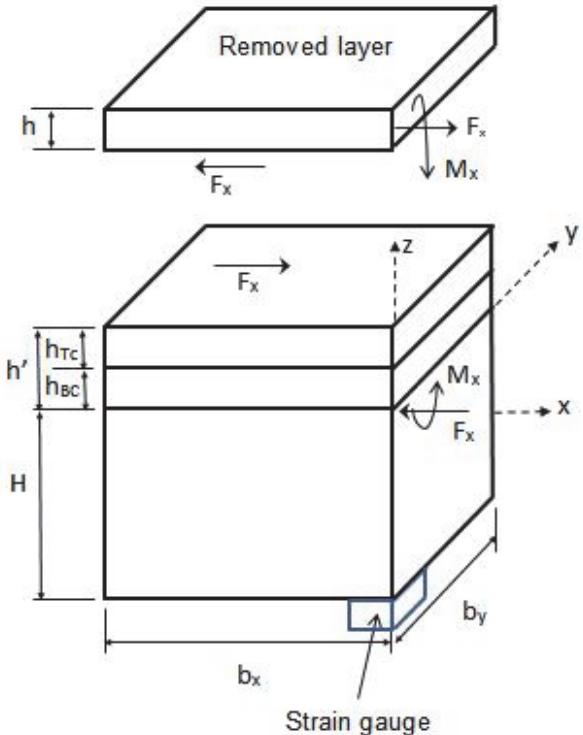


Figure 1. Schematic representation of the TBC indicating the removed layer.  
Source: The authors

Where  $\sum h$  is the sum of the thickness of removed layers.

The resulting forces and momenta along and around  $x$  and  $y$  axes are  $F'_x$  and  $F'_y$ , and  $M'_x$  and  $M'_y$ , respectively, which are related with the stresses by means of eq. 5:

$$\begin{Bmatrix} F'_x \\ F'_y \end{Bmatrix} = \int_{-H}^0 \begin{Bmatrix} \sigma_x \\ \sigma_y \end{Bmatrix} dz + \int_0^{h'} \begin{Bmatrix} \sigma_x \\ \sigma_y \end{Bmatrix} dz \quad \text{and} \quad \begin{Bmatrix} M'_x \\ M'_y \end{Bmatrix} = \int_{-H}^0 \begin{Bmatrix} \sigma_x \\ \sigma_y \end{Bmatrix} z dz + \int_0^{h'} \begin{Bmatrix} \sigma_x \\ \sigma_y \end{Bmatrix} z dz \quad (5)$$

Substituting equations (1) and (2) in (5) results:

$$\begin{Bmatrix} F'_x \\ F'_x \\ M'_x \\ M'_x \end{Bmatrix} = \begin{bmatrix} A_{11} & A_{12} & B_{11} & B_{12} \\ A_{12} & A_{22} & B_{12} & B_{22} \\ B_{11} & B_{12} & D_{11} & D_{12} \\ B_{12} & B_{22} & D_{12} & D_{22} \end{bmatrix} \begin{Bmatrix} \varepsilon_{x0} \\ \varepsilon_{y0} \\ k_x \\ k_y \end{Bmatrix} \quad (6)$$

Where:

$$\begin{aligned} A_{11} &= A_{22} = E_s H + E_t h'; \quad A_{12} \\ &= v_s E_s H + (v_{BC} + v_{TC}) E_t h' \\ B_{11} &= B_{22} = ((E_t - E_s) H h')/2; \quad B_{12} \\ &= (((v_{BC} + v_{TC}) E_t - v_s E_s) H h')/2 \\ D_{11} &= D_{22} = ((E_s H/12) * (H^2 + 3h'^2)) \\ &+ ((E_t h'/12) * (h'^2 + 3H^2)) \\ D_{12} &= ((v_s E_s H/12) * (H^2 + 3h'^2)) \\ &+ (((v_{BC} + v_{TC}) E_t h'/12) * (h'^2 \\ &+ 3H^2)) \end{aligned}$$

Using eq. (1) and the deformations measured by the strain gauges, equations for  $\varepsilon_{x0}$  and  $\varepsilon_{y0}$  can be derived:

$$\varepsilon_{x0} = \Delta\varepsilon_{xG} + k_x \left( \frac{h'-H}{2} \right) \quad \text{and} \quad \varepsilon_{y0} = \Delta\varepsilon_{yG} + k_y \left( \frac{h'-H}{2} \right) \quad (7)$$

Substituting eq. (7) in (6) to eliminate  $\varepsilon_{x0}$  and  $\varepsilon_{y0}$ ; thus eq. (8) is obtained:

$$\begin{Bmatrix} F'_x \\ F'_x \\ M'_x \\ M'_x \end{Bmatrix} = - \begin{Bmatrix} \sigma_{xL}h \\ \sigma_{yL}h \\ \sigma_{xL}h \left( \frac{h+h'-H}{2} \right) \\ \sigma_{yL}h \left( \frac{h+h'-H}{2} \right) \end{Bmatrix} \quad (8)$$

Where  $\sigma_{xL}$  and  $\sigma_{yL}$  are the stresses of the removed layer.

Equations (6) and (8) are substituted in (7) in order to obtain 4 equations with 4 variables, 2 stresses and 2 curvatures. This is undertaken in terms of deformation changes and sample properties. Thus, the following is obtained:

$$\begin{Bmatrix} h & 0 & A_{11} \left( \frac{h'-H}{2} \right) + B_{11} & A_{12} \left( \frac{h'-H}{2} \right) + B_{12} \\ 0 & h & A_{12} \left( \frac{h'-H}{2} \right) + B_{12} & A_{22} \left( \frac{h'-H}{2} \right) + B_{22} \\ \left( \frac{h+h'-H}{2} \right) h & 0 & B_{11} \left( \frac{h'-H}{2} \right) + D_{11} & B_{12} \left( \frac{h'-H}{2} \right) + D_{12} \\ 0 & \left( \frac{h+h'-H}{2} \right) h & B_{12} \left( \frac{h'-H}{2} \right) + D_{12} & B_{22} \left( \frac{h'-H}{2} \right) + D_{22} \end{Bmatrix} \begin{Bmatrix} \sigma_{xL} \\ \sigma_{yL} \\ k_x \\ k_y \end{Bmatrix} = \begin{Bmatrix} A_{11} & A_{12} \\ A_{12} & A_{22} \\ B_{11} & B_{12} \\ B_{12} & B_{22} \end{Bmatrix} \begin{Bmatrix} \Delta\varepsilon_{xG} \\ \Delta\varepsilon_{yG} \end{Bmatrix} \quad (9)$$

Equation (9) shows the equation system to be solved for the stresses of the removed layer,  $\sigma_{xL}$  and  $\sigma_{yL}$ , and changes in the curvatures,  $k_x$  and  $k_y$ . Equation (7) is used to calculate  $\varepsilon_{x0}$  and  $\varepsilon_{y0}$ . Then, the residual stresses on the remaining part of the sample can be calculated by eq. (2).

### 3. Results and Discussion

Different residual stresses are developed during the fabrication of TBCs by thermal spraying. A compressive stress state is first developed on the substrate front face when its surface is prepared by grit blasting. As the BC is deposited, a tensile residual stress is developed by particle quenching and restricted contraction on the substrate [5], and finally, when the TC is deposited, a mixed stress state is developed due to the significant difference in thermal expansion coefficients between both layers. Compressive residual stresses are developed in the TC, whereas the BC develops tensile residual stresses [28].

The residual stress profiles that were calculated by the procedure described in section 2.4 are presented in Figs. 2 and 3. Fig. 2 shows the residual stress profiles, obtained by MLRMDC, for the samples that were heat treated at 1200 °C for 8 and 16 h, and for that without heat treatment. The plots show how the residual stress varies from the TC surface to the substrate.

For the sample without heat treatment (WT-DC), an almost zero residual stress was measured on the TC surface, which agrees with what was reported by Lima et al. [21] and Weyant et al. [6]. This stress state resulted from the balance between the compressive and tensile stresses in the coating. The former stress is promoted by the large difference of thermal expansion coefficients of YSZ and CoNiCrAlY, whereas the latter is produced by rapid quenching of flying particles when impacting the substrate and previously deposited layers. At an approximate depth of 250 µm, within the BC, a residual tensile stress of 20 MPa was measured. Such behavior has also previously been reported by other authors [21, 29]. In this case, a dominant effect of quenching stress in the bond coat was evident. As the measurements approach the substrate, the stress state tended, once again, to be zero, and the residual stress measured within the substrate was compressive. Although the results obtained by the MLRMDC method in this work showed a similar behavior to those reported in the literature employing the MLRM procedure [21], the magnitude of the measured stresses was different for each technique. This behavior could be due to the different processing conditions employed in each case (substrate, fuel and spraying distance).

The residual stress profile of the TBC that was heat-treated at 1200 °C for 8 h (8H-DC, Fig. 2) also showed a near zero stress at the TC surface. From 50 µm only compressive stresses were measured. The highest residual stress (-19 MPa) in the coating was located within the BC, whereas a further increase, up to -40 MPa, was measured in the substrate. The residual stress profile of the TBC that was heat-treated at 1200 °C for 16h (16H-DC) revealed a stress of -18 MPa on the TC surface, which increased as the depth was increased. The profile of this sample showed the same trend as that of the sample treated for 8 h, but the magnitude of the stresses was higher; this behavior has also been reported by Khan et al. [3]. These authors reported an increase in compressive residual stresses of a TBC as the exposure time at high temperature was increased. The TBC that was heat-treated for 16 h also showed a maximum compressive residual stress (-58 MPa) within the BC at a depth of 250 µm. The substrate showed a larger compressive residual stress of -78 MPa. A BC having a large residual compressive stress promotes good adherence of the TBC to the substrate; therefore, an increased adherence is expected in heat-treated samples compared to that of the as sprayed counterpart. Also, a TBC having a compressive residual stress state shows higher resistance to cracking and delamination in service [15]. The operating temperature of aeronautic and land turbines are higher than the testing temperature employed in this work (1200 °C). This testing temperature was selected as in a future investigation the residual stress profiles of TBCs, affected by chemical attack of calcium magnesium alumino-silicate (CMAS), will be calculated by MLRMDC. CMAS attack normally occurs between 1100 and 1200 °C on TBCs in aeronautic turbines exposed to volcanic flying ashes or SiO<sub>2</sub>-based sand.

Fig.3 shows the evolution of residual stress calculated by MLRM and MLRMDC as a function of depth. A similar behavior of the evolution of residual stress was observed for both methods for the substrate and BC. However, the stress

magnitude for the TC was slightly different. Fig. 4 shows a magnified view of the residual stress profile calculated for the TC. The residual stresses on the surface of the samples without heat treatment and heat-treated for 8 h showed a difference smaller than 1 MPa, whereas for the sample heat-treated for 16 h the difference was about 2 MPa, which represents a dissimilarity of 15 % between both methods. The difference of the residual stress profiles, calculated by both methods for the TC, can be explained by the fact that the MLRMDC procedure takes into account the physical properties of the substrate, BC, and TC, whereas the MLRM only considers the physical properties of the substrate and TC.

This work should be regarded as an exploratory study aiming to provide an initial insight into the results obtained by MLRM and MLRMDC. This is why only one sample per condition was studied. A larger number of samples need to be studied in order to assess the error involved in each measurement and unambiguously determine if there is a statistically valid difference between both methods when calculating the residual stress profile through the TC. Nonetheless, it seems that the MLRM underestimates the magnitude of the stresses within the TC.

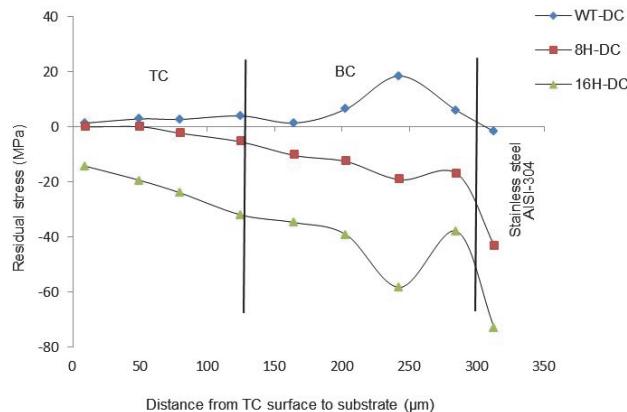


Figure 2. Evolution of residual stress, obtained by MLRMDC as a function of depth for samples with and without heat treatment.  
Source: The authors

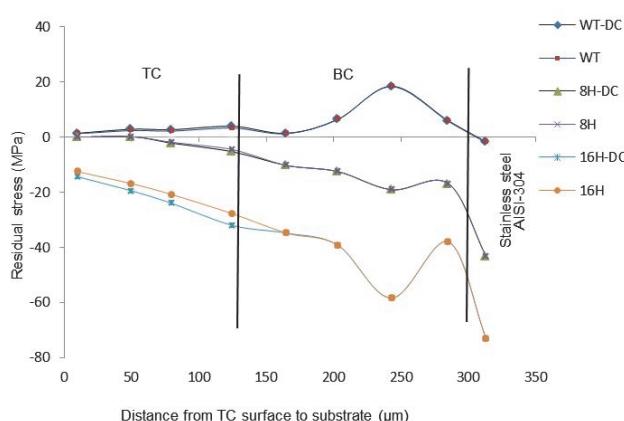


Figure 3. Residual stress profiles for the TBC obtained by MLRM and MLRMDC.  
Source: The authors

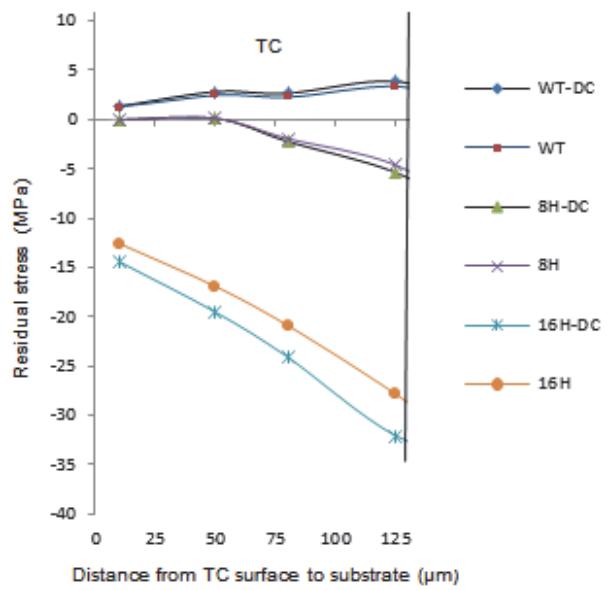


Figure 4. Comparison of the residual stress profiles in the TC calculated by MLRM and MLRMDC.  
Source: The authors

#### 4. Conclusions

The evolution of residual stress state in the TBCs (YSZ/CoNiCrAlY), fabricated by thermal spraying, was obtained by employing the modified layer removal method for duplex coatings (MLRMDC). This method resulted in residual stress profiles similar to those obtained by MLRM, which is a procedure regularly reported in the literature to determine the residual stresses of coatings manufactured by thermal spraying. The results obtained employing both methods are in good agreement with those reported in the literature. The effect of heat treatment at 1200 °C on the evolution of the residual stress profiles was also studied here. It was also demonstrated that an increase in soaking time at high temperature eliminates all tensile stresses in the TBCs, which results in compressive stresses after heat treatment. This, in turn, may enhance the adherence and delamination resistance of the coating in service. Until this point, it seems that the MLRM may be underestimating the magnitude of the compressive stresses within the TC.

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