Optimisation of Electroless Ni-YSZ Composite Coating Process on High Ceramic YSZ Particle and Porosity Content

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ABSTRACT

The study discusses on the optimisation of the ceramic yttria-stabilised zirconia (YSZ) particle and porosity content in the electroless Ni-YSZ composite coating. There were two sets of design of experiment (DoE) were designed and conducted to optimise each yield. Both DoEs are using Taguchi orthogonal array L9 with three varying parameters at three levels. DoE optimisation of ceramic YSZ particle was varying electroless nickel process parameters namely agitation, substrate orientation and particle loading. Another set of DoE to optimise porosity content was designed by varying the types of poreformers, bath agitation and poreformer volume. These L9 Taguchi DoE was analysed using statistical tool ANOVA in Minitab 15 software with YSZ particle yield was determined by energy dispersive X-ray analysis (EDXA) and porosity yields by Archimedes buoyancy method. Discussion of the optimum process for both DoE Taguchi based on the signal to noise ratio with both DoEs were set at larger the better characteristic. The discussion was supported by the Ni-YSZ composite coating microstructure and characterisation using scanning electron microscope (SEM) coupled with energy dispersive X-ray (EDX).

Key words: Electroless Ni-YSZ; composite coating; Taguchi; poreformer; porosity

INTRODUCTION

Electroless nickel co-deposition process is an electrocatalytic process where the metallic nickel deposited together with the inert ceramic particle onto the substrate. The inert ceramic particle that have previously been investigated including the co-deposition of diamond, silicon carbide, silicon nitride, silicon oxide, alumina, ceria, yttria and zirconia. The composition of metallic nickel and ceramic particle in the coating is controlled to give desired physical and mechanical properties. Typical co-deposition consists of particulates in the size range of 0.1-10 μm and the maximum particle loading obtained was 40 vol.% of the total matrix [6].

The electroless nickel co-deposition process is highly affected by several factors such as bath composition, bath temperature, bath pH, particle inertness, shape and size, bath agitation, and substrate surface condition and others [1,2]. The nickel-ceramic particle coating is widely applied in corrosion [3], wear [4], thermal [5] resistance and has become highly important in renewable energy, solid oxide fuel cell [15,11]. The incorporation of YSZ enhances the corrosion, wear and thermal resistance to the metallic nickel. However in the application in thermal resistance, the thermal expansion coefficient difference between metallic nickel (16.5 x 10^-6 K^-1) and YSZ (10.7 x 10^-6 K^-1) ceramic are high. Thus, it is important to ensure the composite coating is having a uniformly distributed YSZ to avoid cracking within the coating.

Investigation of porosity level in the deposition is critical as the amount of porosity enhances thermal insulation for thermal barrier coatings and gas circulation in fuel cell anode [8] applications. The amount of porosity should not be more than 40 vol % as greater amount of porosity will reduce the mechanical properties of the deposit. It is known that electroless nickel coating has an excellent uniformity and dense deposition with thickness less than 10 μm [9]. The amount of porosity in electroless nickel
deposition could be induced by varying the agitation methods, deposition rate, bath pH, substrate surface condition as well adding poreformer.

Objectives:
The objectives of the studies are to optimise fabrication process of electroless Ni-YSZ composite coating on two yields; the ceramic YSZ particle content and porosity content in the composite coating. The optimisation was designed using a DoE Taguchi orthogonal array L9.

Materials and Methods
Two sets of DoE Taguchi L9 were designed to evaluate the optimum ceramic YSZ particle and porosity content. The DoE Taguchi for optimising YSZ particle content in the coating is given in Table 1. The parameters and levels were selected based on previous studies conducted by authors as well as by others.

The DoE Taguchi for optimising the porosity content in the coating is given in Table 2. It was expected that the addition of pore former in the coating will increase the amount of porosity. Previous study showed that type of agitaion (Baba, Davidson & Muneer, 2011) did contribute to the increament of porosity level in the coating. Logically, adding more pore former will increase the porosity.

Results and Discussion
Ceramic YSZ particle content optimisation
Electroless nickel co-deposition is an autocatalytic process. The addition of ceramic YSZ particles in the bath will cause a co-deposition of metallic nickel and ceramic YSZ onto the substrate. In order to enhance the amount of ceramic YSZ in the composite coating, several bath parameters were chosen that are the agitation, substrate orientation and volume of YSZ particles. Previous studies found that particle sizes and type of agitation were the dominant parameters affected the amount of ceramic YSZ particl-deposited into the coating [11]. Thus in this study, the type of agitation is maintained with two others parameters namely the substrate orientation and particle loading are introduced.

A DoE Taguchi L9 was developed to determine the optimum condition for increasing the amount of YSZ particle in the composite coating. The signal to noise ratio of the DoE Taguchi was analysed using statistical tool ANOVA in Minitab software. The target was set at 95% level of significance and the larger the better optimisation.

Table 3 shows the ANOVA for ceramic YSZ particle optimisation. ANOVA calculates the F-ratio, which is the ratio between the regression mean square to the mean square error. This ratio is used to determine the significance of the parameters under investigation with respect to the variance of all the terms included in the error term at the desires significance level. The percent of contribution, P was calculated based on the F-ratio value.

It was found that parameter A is the most significance parameters as it showed the highest percent of contribution for optimising the ceramic YSZ particle content in the composite coating. The other two parameters, B and C are not much affected.

The variation of signal to noise ratio for optimising ceramic YSZ content was given in Fig. 1. It is clearly shown that the signal to noise ratio variation is large for A but not so much for B and C. As the target set larger the better, parameter A at level 2 give the optimum condition followed by B and C at level 3. Since the percent of contribution for parameter B is very the smallest, the optimum
condition for high ceramic YSZ particle content in the composite coating is A2C3B3.

This concluded that to achieve high ceramic YSZ particle in the EN composite coating, the optimum condition is to set the agitation air bubble at 90° substrate orientation and highest particle loading at 45 g. This was contradicted with the previous study where it was found mechanical stirring was the optimum condition. This can be explained due to the bubbling effect that push the particle upwards and the position substrate at 90° (flat) with the deposition surface facing downwards induced more ceramic YSZ particle to co-deposited on to the substrate.

The surface of the Ni-YSZ composite coating was investigated under the SEM copule with EDX. The corresponding EDX spectrum (Figure 2) shows the presence of major peak of nickel (Ni), zirconium (Zr), sodium (Na), carbon (C), oxygen (O) and phosphorous (P). These confirmed that the composites are composed of the combination of metallic nickel and ceramic YSZ. The existence of phosphorus (P) element in the composite coating is due to the fact that phosphorus is one of the major elements in the EN hypophosphite-base bath solution.

The microstructure of Ni-YSZ composite coating is commonly divided into three region. White area represented the ceramic YSZ particle, gray area represented the metallic nickel and the dark area represented the porosity as described in the previous study [11]. Figure 3 compares the SEM microstructures of Ni-YSZ composite coating varying types of agitation which are stirring, bubble and fan respectively at varying particle loading of 15g, 30g and 45g.

![Main effect plot for SN ratio in ceramic particle optimisation](image1)

**Fig. 1: Main effect plot for SN ratio in ceramic particle optimisation**

![EDXA spectrum of electroless Ni-YSZ composite coating](image2)

**Fig. 2: EDXA spectrum of electroless Ni-YSZ composite coating**
Comparing the variation of bath agitation, air bubbling give the most uniform ceramic YSZ particle distribution for all variation of particle loading. Uniformly distributed ceramic particle ensure constant coefficient of thermal expansion within the coating to avoid cracking or delamination due to thermal gradient. Stirring and fan showed at lowest particle loading showed agglomeration of ceramic YSZ particle where some areas are fully gray indicating no ceramic YSZ particle co-deposited.

In general as the particle loading increases, the amount of ceramic YSZ particle incorporated in the composite coating also increases. This is very obvious for the fan bath agitation and proportional to Braudrand (1994). Air bubbling bath agitation at 45 g showed more dark area which represent the porosity. The air bubbling caused higher volume of ceramic YSZ particle and thickness, thus it is induced more porosity as the thickness increases (das & chin).

Porosity content optimisation:

The second objective of the study is to enhance the porosity incorporation in the composite coating. In order to induce more porosity in the composite coating, previous studies show that incorporation of poreformer increases the amount of porosity. The poreformer usually is organic materials which will be burnt at low temperature leaving an empty space i.e. porosity. Thus the major parameters chosen for this study are type and volume of poreformer as well as bath agitation.

A DoE Taguchi L9 was developed to determine the optimum condition for increasing the amount of porosity in the composite coating. Table 3 shows the ANOVA for porosity optimisation. The F-ratio value determine the significance of the parameters A and B are worth to further investigated. The calculated percent of contribution, P showed the highest for A (24.22%) followed by B (17.60%) and lastly C (6.58%).

It was found that parameter A is the most significance parameters as it showed the highest percent of contribution for optimising the porosity content in the composite coating together with parameter B. Parameter C was not very much affected the increment of porosity in the coating.

The variation of signal to noise ratio for optimising porosity content was given in Fig. 4. It is clearly shown that the signal to noise ratio variation is large for A as well as B but not so for C. As the target set larger the better, parameter A and B at level 2, and parameter C at level 3 give the optimum condition. As a conclusions, the optimum condition for high porosity content in the composite coating is A2B2C3.

In order to achieve high porosity content in the EN composite coating, the PTFE poreformer with air bubbling agitation at 15 g poreformer volume is the
optimum condition. It was found contradicted with the previous study [12] where the mechanical stirring was the optimum condition. In the previous study, the agitation was mainly to keep the particle in suspension but in the current study the bath agitation is to keep both particle and poreformer in suspension. The scenario was the ceramic YSZ particle tends to stay at the bottom of the bath whereas the poreformer tend to stay floating at the top of the bath. This explained why bubbling is the most optimum because it is more random. The mechanical stirring acts at the bottom of the bath and give vortex at the centre in the other hand, fan acts in the centre where turbulence across horizontally in one direction.

Poreformer is expected to give higher porosity content thus it is proportional to the amount of poreformer incorporated. Comparing the three types of poreformer, PTFE shows the best followed closely by PE.

Figure 5 shows the SEM microstructure of Ni-YSZ composite coating varying types of poreformer at different volume of 5g, 10g and 15g. Generally the resulting pores are numerous and subtle. SEM microstructure for PTFE poreformer in general showed the most uneven and accumulate lots pores by the dark area compared the graphite poreformer (Figure 5c). The SEM microstructure for graphite poreformer generally show the most flat and almost no porosity, however the highest volume poreformer did show some subtle pores on the surface of the composite coating, e.g. white circle in Figure 5c(iii).

Fig. 4: SEM microstructure for Ni-YSZ by (a) Polyethylene poreformer at (i) 5g (ii) 10g (iii) 15g; (b) Polytetrafluoroethylene poreformer (i) 5g (ii) 10g (iii) 15g; and (b) Graphite poreformer (i) 5g (ii) 10g (iii) 15g; where the bath agitation was kept constant.
Conclusions:
Optimisation studies on ceramic YSZ particle and porosity content in electroless Ni-YSZ composite coating using DoE Taguchi L9. The ceramic YSZ particle content optimisation was varying bath parameters, i.e. bath agitation, substrate orientation and particle loading at three levels. The optimum condition for high YSZ particle content in the composite coating is A2C3B3. This indicates the air bubbling bath agitation at maximum particle loading of 45 g and 90° substrate orientation resulted in high ceramic YSZ particle in the composite coating. The DoE for porosity content optimisation was developed with varying parameters such as types of poreformer, bath agitation and volume of poreformer at three levels. The optimum condition for high porosity content in the composite coating is A2B2C3. This showed that to achieved optimum porosity content in the composite coating, the PTFE poreformer with air bubbling agitation at 15 g poreformer volume should be used. The microstructures of both DoE do support and justify the findings. The thermal and mechanical behaviour of the composite coating will be discussed in the future publication.

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