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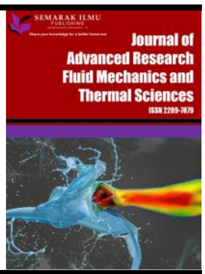
Journal of Advanced Research in Fluid Mechanics and Thermal Sciences

Journal homepage:
https://semarakilmu.com.my/journals/index.php/fluid_mechanics_thermal_sciences/index
ISSN: 2289-7879



Influence of Heat Treatment on Microhardness and Surface Roughness of Electroless Ni-YSZ Composite Coating

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ARTICLE INFO

Article history:
Received 10 April 2022
Received in revised form 25 July 2022
Accepted 5 August 2022
Available online 1 September 2022

Keywords:
Electroless nickel; composite coating; microhardness; surface roughness; heat treatment

ABSTRACT

The application of heat treatment was introduced in Ni-YSZ composite coating as the incorporation of ceramic YSZ in electroless nickel deposition is new and worth investigating. In this paper, the influence of heat treatment by varying the heating temperature and time on the electroless Ni-YSZ composite coating is investigated. The Ni-YSZ composite is deposited onto a high-speed steel substrate via electroless nickel co-deposition. YSZ powder of mixed sizes of micro- and nano-sized of the ratio of 1:1 is incorporated in the electroless deposits. The electroless Ni-YSZ composites coating was heated up to 400^{o}C for a maximum of 2 hours. The microhardness measurements were carried out using a Vickers microhardness tester (Shimadzu) according to ISO 6507-4. The surface roughness of the coating was measured using Mitutoyo surface roughness tester SJ-301. The surface characterisation was analysed using Cambridge Stereoscan 90 Scanning Electron Microscope (SEM) coupled with Energy Dispersive X-ray Analysis (EDX). The crystallographic structure of materials was analysed by X-ray diffraction (XRD) Bruker D8 Advance instrument. The microhardness and surface roughness of the coating both increase with time. The microhardness is directly proportional to the heating temperature and time and these observations are supported by the XRD analysis. The surface roughness observation is explained by the SEM micrographs.

1. Introduction

Heat treatment is a method of heating a material to a certain temperature for a specific period to improve its mechanical properties. Heat treatment on electroless Ni-P shows that the phosphorus content plays the most important factor in influencing the microhardness properties as mentioned by Parkinson [1]. There are also studies investigating the effect of heat treatment on electroless nickel composites.

Composites are formulated via a combination of two or more materials in a body. The applications of this composite are found in various industries, especially for high operating

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https://doi.org/10.37934/arfmts.99.1.114122

temperature, high wear, and corrosion resistance environments. Electroless nickel composites are produced by incorporating inert particles in the nickel metal matrix. The electroless deposition has a variety of metal matrices, i.e. aluminium, nickel, copper, silver, and iron as reviewed by Abdel Hamid [2]. The most common electroless technique is electroless nickel since nickel has interesting properties as superalloy as described by Glotka [3] and it can be improved by incorporating carbide as stated by Ahmadkaniha *et al.,* [4] or other ceramics.

A study by Sudagar *et al.,* [5] found that electroless nickel deposited gives excellent corrosion, wear, and abrasion resistance as well as good ductility, lubricity, and electrical conductivity. The deposits usually contained 2-14% of phosphorus and it does vary the structure of the deposit from crystalline to amorphous. The incorporation of ceramic particles into the Ni-P deposit produces a cermet such as silicon carbide work done by Ahmadkaniha *et al.*,[4], nano-diamond and diamond work done by Mirhosseini *et al.,* [6] and Trzaska *et al.,* [7], and cubic boron nitride by Norsilawati *et al.,* [8]. The influence of electroless coating parameters play an important effect on the deposit quality and the incorporation of particle quantity can be enhanced by using smaller size particle, stirring agitation, and blasting surface treatment as reported by Baba *et al.,* [9]. It was also found by Ashtiani *et al.,* [10] that different types of complexing agents in the electroless nickel bath also give affect the deposit properties.

Extensive study has been done to improve the properties of electroless nickel or its cermet by giving heat treatment. A study by Buchtík *et al.,* [11] shows that heat treatment improves the hardness and wear resistance of the electroless co-deposition. It was found by Khuram Shahzad *et al.,* [12] that the microhardness of electroless nickel-phosphorus deposit decreases as the number of phosphorus increases due to the structure change from crystalline to amorphous in electroless nickel-titanium nanocomposite coating. In another study by Winowlin Jappes *et al.,* [13], heating the electroless nickel between 300-400^{o}C improves the phase transformation of the deposit from amorphous to crystalline. The amount of nickel composition in electroless nickel-activated carbon was found by Abioye *et al.,* [14] to increase as the treating time increases from 1 to 2 hours.

The hardness of the electroless nickel boron nitride was found to be increased by 40% after being heat-treated at 300^{o}C however it reduces the wear rates as reported by Kiran *et al.,* [15]. The Ni-P deposit under heat treatment between 400-700^{o}C and water-quenched shows an increase in hardness as well as surface adhesion in work done by Arulvel *et al.,* [16]. The microhardness and wear of electroless nickel-phosphorus increase as the temperature increases from 300 to 600^{o}C, however above 600^{o}C for 4 hours show no effect on both due to the formation of Ni_3P crystalline as mentioned by Biswas *et al.,* [17].

There is a finding by Sharma *et al.,* [18] shows the effect of surface roughness after the coating and concluded that there are higher values of surface roughness after the coatings. The effect of the surface roughness on the nano composition can make the particle concentration the most significant factor in wear rate followed by current density and temperature. The surface roughness is affected substantially by the wear behaviour and thickness of the coating as Raghavendra *et al.,* reported [19].

In this paper, the electroless nickel (Ni) and yttria-stabilised zirconia (YSZ) is investigated on the influence of heat treatment on the coating microhardness and surface roughness.

2. Methodology

2.1 Materials Preparation

The substrate is a base material for cermet coating to be deposited on it. The substrate used is high-speed steel (HSS) from Bohler-Bleche GmbH manufacturer with composition in Table 1. The substrate was cut using a wire-cut electrical discharge machine into dimensions illustrated in Figure 1 with a thickness of 1.25 mm and diameter of 25 mm.

Table 1
Chemical composition of HSS substrate

Element	C	Si	Mn	P	S	Cr	Mo	V	W
Composition (wt.%)	0.890	0.200	0.280	0.025	0.0008	3.930	4.720	1.700	6.130

Fig. 1. Sample preparation of HSS substrate by wire-cutting electrical discharge machine

Reinforcement particles of yttria-stabilised zirconia (YSZ) with 8 mol% by Tosoh Japan are used. There are two sizes of 8YSZ used, micro-sized with of nominal diameter of 2 μm and nano-sized particles ranging between 100-500 nm. The optimum particle loading is in the range of 5-10 g/L as used by Baba *et al.,* [9]. The particle loading for both sizes used is 10 g/L and the mixed particle size is by a ratio of 1:1.

Electroless nickel solutions are prepared by AR grade chemicals and high purity ionised water as described in previous research done by Baba *et al.,* [20]. HSS substrate was pre-treated in 4 different chemicals namely cuprolite, pre-catalyst, catalyst and niplast for 15 minutes each at different temperatures. Then the substrate was placed in Slotonip electroless nickel solution together with 8%YSZ powders for the co-deposition process to occur.

2.2 Equipment and Testing

The electroless Ni-YSZ coating undergoes a heat treatment process to improve its mechanical properties. The coating was heated in a Protherm electric furnace under a controlled environment of nitrogen gas flow at a constant pressure of 1 atm. The temperature and time were varied in a range of 300-400^{o}C and 0-2 hours respectively as conducted by previous studies by Sarkar *et al.,* [21] and Abishek *et al.,* [22]. After the heat treatment process, 2 batches of samples underwent microhardness testing, surface roughness testing and materials characterisation.

The microhardness measurements were carried out using a Vickers microhardness tester (Shimadzu) under the microhardness range; HV0.025 (25 gf) at a 0.245 N force for 10 s according to ISO 6507-4. There are a total of 9 samples and five measurements of microhardness are taken for each sample.

The coating surface was measured using Mitutoyo surface roughness tester SJ-301. There are a total of 9 samples and five surface roughness measurements are taken in each section along the radial direction and averaged.

3. Results
3.1 Microhardness and Surface Roughness

Electroless Ni-YSZ composite coating was successfully deposited onto HSS substrate with an average of 30 µm thickness measured by Mitutoyo digital micrometre. The heat-treated electroless Ni-YSZ composite coatings were tested for Vickers microhardness and surface roughness. Upon heat treatment up to 400^{o}C for a maximum of 2 hours, the composite coatings are generally hardened as shown in Figure 2. The increment of microhardness Hv heated at 300^{o}C for 2 hours is only 33% compared to when heated at 400^{o}C for 2 hours, the increment is up to 70%. The effect of varying heating temperatures from 300^{o}C to 400^{o}C has a great influent on the electroless Ni-YSZ composite coating microhardness as the increment is almost doubled. A similar finding was also mentioned by Kiran *et al.,* [15] which the hardness increased by 40% as the electroless nickel boron nitride was heated to 300^{o}C. In most previous studies, this behaviour is observed due to the phase transformation of the nickel from amorphous to crystalline structure [11,12,13].

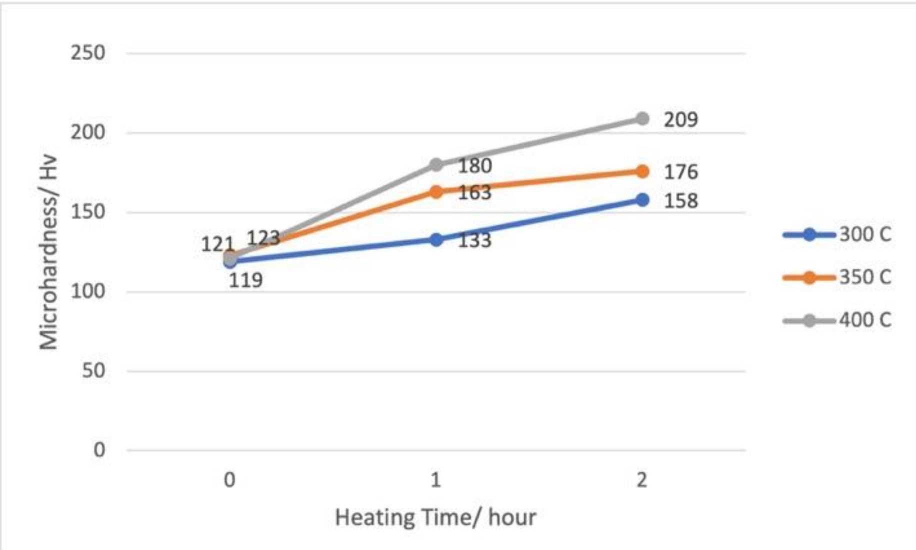


Fig. 2. Vickers microhardness against heating time of electroless Ni-YSZ composite coating

The electroless Ni-YSZ composite coating was also tested for its surface roughness. It is found that the surface roughness of the heat-treated electroless Ni-YSZ composite coating is not directly proportional to the heating temperature or heating time as shown in Figure 3. Electroless Ni-YSZ composite coating samples heated at 300^{o}C for 2 hours does not affect the surface roughness which the variation of surface roughness of only 3.5%. On the other hand, electroless Ni-YSZ composite coating heated at 350^{o}C and 400^{o}C both show rougher surfaces with surface roughness increases by 23% and 11% respectively. A study by Sarkar *et al.,* [21] shows the surface roughness of electroless Ni-P coating in optimised conditions by controlling the electroless nickel bath parameters is 0.32 µm. Compared to the data obtained by electroless Ni-YSZ composite heat treated at 350^{o}C, the surface roughness is still lower than 0.32 µm.

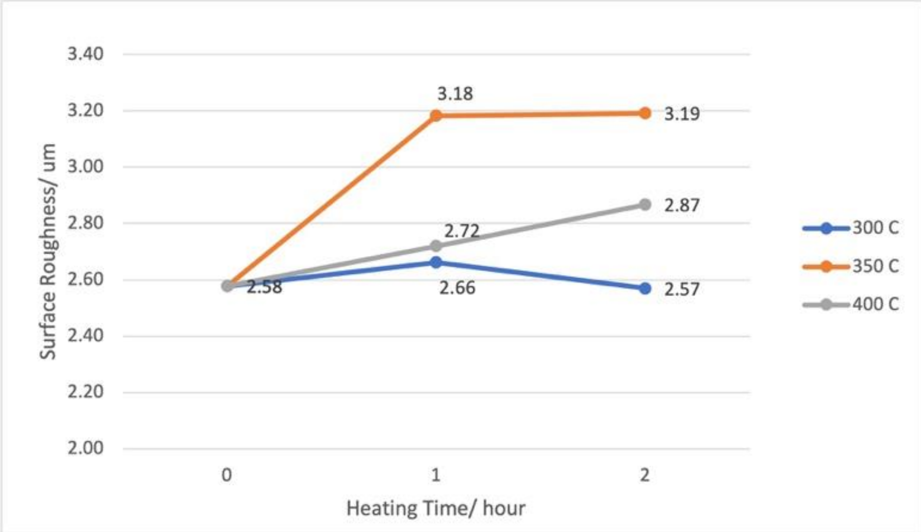


Fig. 3. Surface roughness against heating time of electroless Ni-YSZ composite coating

It can be drawn from these observations that microhardness and surface roughness have some correlation with each other. Generally, both microhardness and surface roughness increase as the heating time increases.

3.1.1 Materials characterisation

Materials characterization of electroless Ni-YSZ composite coating was done by SEM micrograph and XRD analysis. Figure 4 shows the comparison of electroless Ni-YSZ composite coating surface morphology heat-treated at 300^{o}C to 400^{o}C for 1 and 2 hours. The surface morphology of the composite coatings heated at 300^{o}C for 1 hour and 2 hours in Figure 4(a) and Figure 4(b) respectively are having similar cauliflower patterns. This explains the surface roughness differed only by 0.09 µm between the 2 surfaces. On contrary, Figure 4(c) and Figure 4(d) show the electroless Ni-YSZ composite coating heated at 350^{o}C for 1 hour and 2 hours respectively. The surface morphology electroless Ni-YSZ heated for 1 hour in Figure 4(c) shows rough ridges across half of the surface. Whereas the surface morphology for electroless Ni-YSZ heated for 2 hours (Figure 4(d)) has some parts of rough ridges and fine cauliflower pattern compared to Figure 4(a) and (b). These fine cauliflower patterns also contributed to the high surface roughness measurement obtained. Upon heating to 400^{o}C, the surface morphology of electroless Ni-YSZ composite coating heated for 1 hour (Figure 4(c)) and 2 hours (Figure 4(d)) show similar medium cauliflower patterns. It is found that the surface morphology in Figure 4d is slightly rougher with few low ridges. On both coating surfaces, there are few significant pores found. According to Abhishek *et al.,* this observation is caused by improper surface activation before the plating process as mentioned by Abishek *et al.,* [22].

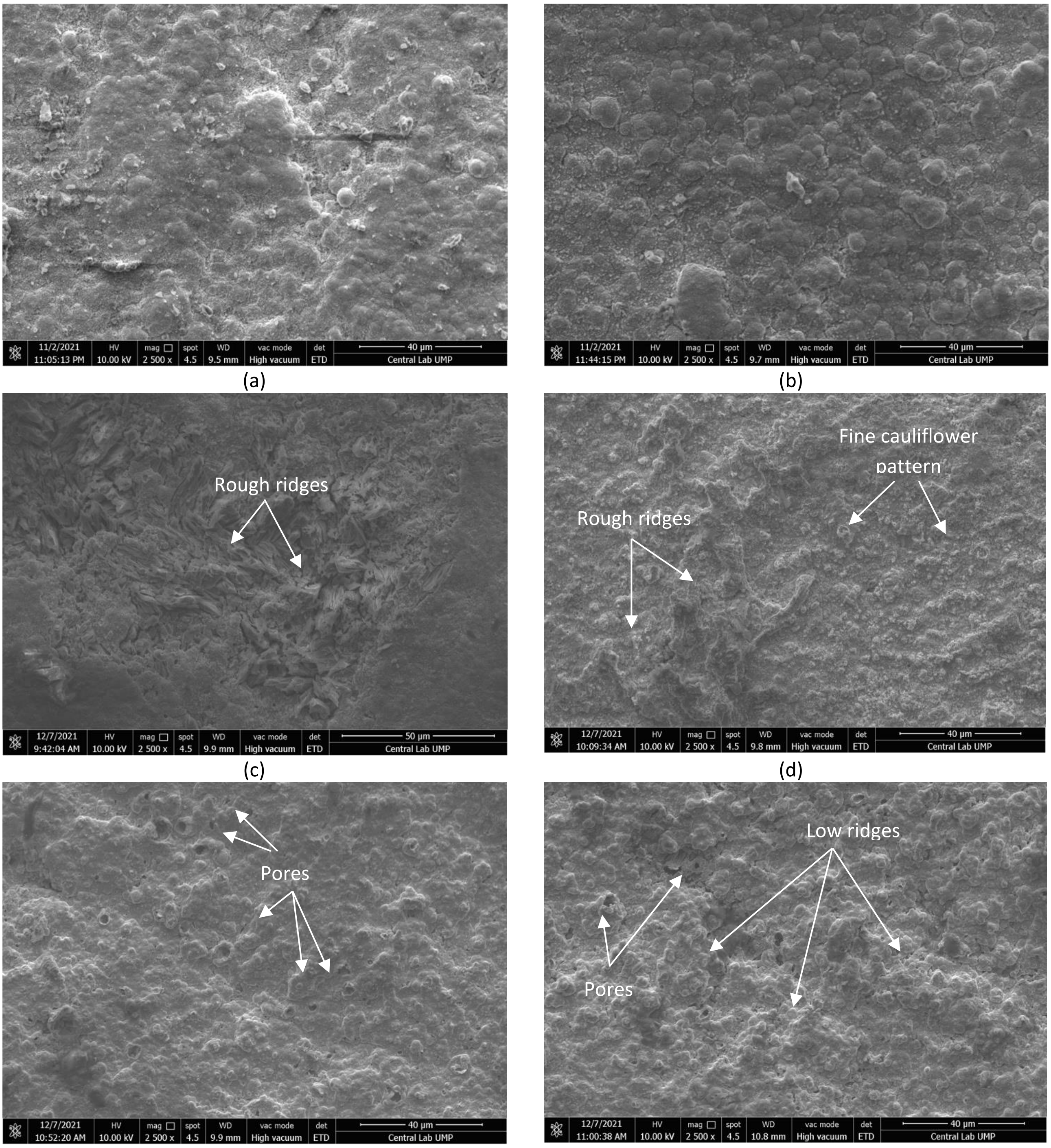
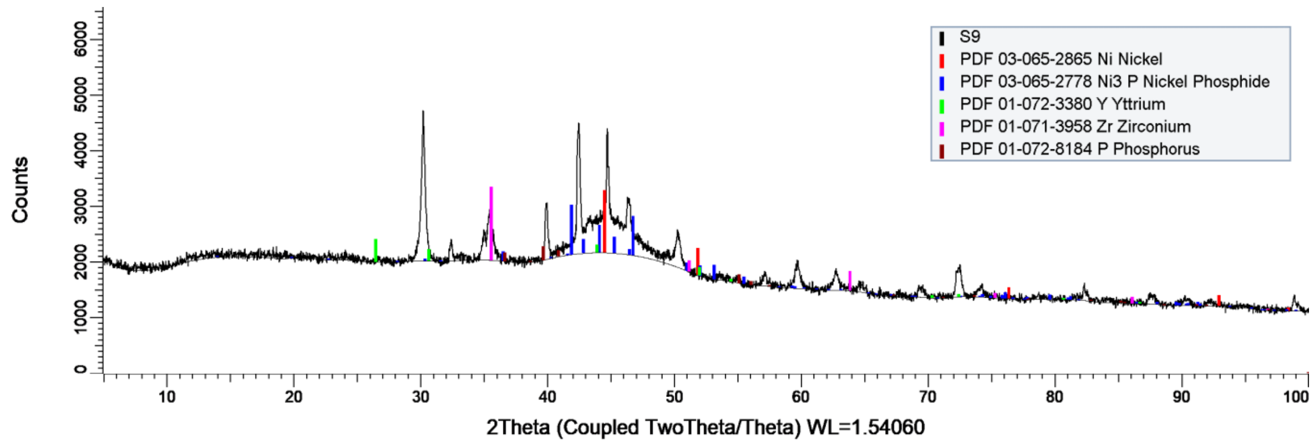
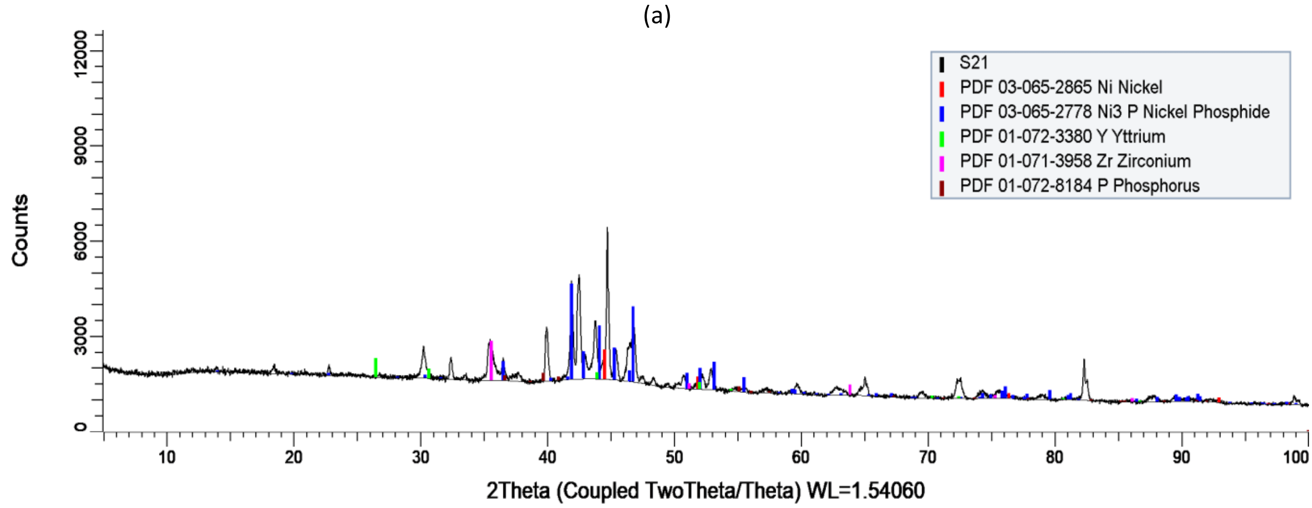


Fig. 4. SEM micrographs for electroless Ni-YSZ composite coating (a) Heat treated at 300°C for 1 hour (b) Heat treated at 300°C for 2 hours (c) Heat treated at 350°C for 1 hour (d) Heat treated at 350°C for 2 hours (e) Heat treated at 400°C for 1 hour (f) Heat treated at 400°C for 2 hours

The crystallographic analysis was carried out by XRD giving the XRD patterns as shown in Figure 5. The blue peaks of nickel phosphite Ni_3P crystals are more pronounced in Figure 5(b) which was heat treated at 400°C for 2 hours. The peaks of sample heat treated at 300°C show broad-spectrum indicating that it is an amorphous structure whereas the sample heat treated at 400°C shows sharp peaks indicating that the structure is crystalline. These observations support the findings of electroless Ni-YSZ composite coating's hardness heated at 400°C is higher than 300°C.



(a)



(b)

Fig. 5. XRD crystallographic analysis (a) S9 Heat treated at 300°C for 2 hours (b) S21 Heat treated at 400°C for 2 hours

4. Conclusions

Electroless Ni-YSZ composite coating was uniformly deposited onto the HSS substrate. Heat treatment with heating temperatures 300°C to 400°C and heating time from 0 to 2 hours show a directly proportional influence on the microhardness. This behaviour is supported by the XRD analysis that shows at 400°C the crystal Ni_3P is found to exist that hardened the coating. Besides that, the XRD spectrum for 400°C shows sharp peaks compare to the 300°C broad spectrum indicating at 400°C, the structure is crystalline and at 300°C is amorphous. On the other hand, the surface roughness is not directly proportional to both heating temperature and time. The surface morphology using SEM shows the variation of cauliflower pattern and ridges observed indicating variation of surface roughness.

Acknowledgement
The author acknowledges the financial support of the Ministry of Higher Education (MOHE) Malaysia for the fund under the Fundamental Research Grant Scheme FRGS1/2018/STG07/TATI/01/1.

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