Journal of Advanced Research in
Applied Mechanics

Journal homepage:
https://semarakilmu.com.my/journals/index.php/appl_mech/index
ISSN: 2289-7895

Effects of Interface Bonding of NiCrAlY on Sandblasted Laser Modified H13 Tool Steel

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ARTICLE INFO

Article history:
Received 29 October 2024
Received in revised form 30 November 2024
Accepted 7 December 2024
Available online 30 December 2024

Keywords:
Interface bonding; diffusion mechanism; Interfacial Indentation Test (IIT); laser surface modification

ABSTRACT

NiCrAlY (Ni-164/211 Ni22%Cr10%Al1.0%Y) coatings were developed on sandblasted laser modified H13 tool steel surface using atmospheric plasma spray (APS). Bonding strength was investigated between these layers. Constant laser parameters were set on all 5 samples and different sandblasted time of 20s to 60s time were set prior to APS spraying. Its' effect on mechanical properties of H13 tool steel surface were investigated. The coating microstructure and diffusion of atoms along NiCrAlY coating, laser modified and substrate layers were investigated by energy dispersive X-ray spectroscopy (EDXS) using Hitachi Tabletop Microscope TM3030 Plus. Interface bonding of NiCrAlY and Micro Vickers Hardness on each layer was investigated by interfacial indentation test (IIT) method using MMT-X7 Matsuzawa Hardness Tester Machine with vickers indenter. Based from IIT method results, average interfacial toughness, K_{avg} for reference sample was 2.15 MPa $m^{1/2}$ compares to sample 29 range of K_{avg} 2.62 MPa $m^{1/2}$. Hence, according to K_{avg} sample 29 has higher interface bonding and obtained average surface roughness of 4 micrometer prior to coating. The EDXS analysis indicated presence of Fe in the NiCrAlY coating layer and increased of Ni and Cr composition in the laser modified layer. Atomic diffusion occurred in both coating and laser modified layers involved Fe, Ni and Cr elements. These findings introduce enhancement of coating system by substrate surface modification to allow atomic diffusion.

1. Introduction

Surface roughness characteristics is crucial to improve adhesion between substrate and coating. This roughness surface gives mechanical keying effect that grips between both of substrate and coating [1]. Surface modification using laser on the substrate surface increase its' surface characteristics such as average surface roughness, Ra or mean roughness depth, Rz. Both of this surface roughness values are insuffcient enough to evaluate its' effect on coating adhesion. Reason

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https://doi.org/10.37934/aram.130.1.168179

because its' surface profile determines the main characteristics of coating adhesion. In laser modification, a coarse surface profile can be achieved with higher Ra's or Rz's. Due to detail mechanical keying needs to be investigated, it is not sufficient enough for the coating to be adhered onto substrate surface. A surface with fine coarse surface profile pattern needs to be obtained to increase coating adhesion [2]. Lasers with the large spot size is only can obtained a coarse surface profile. But, if this is limited, sandblasted method for the laser modified surface prior to coating is essential for to be adhered between the coating and the substrate. Difference of surface profile between lasered substrate being sandblasted with non-lasered substrate being sandblasted, would showed that lasered substrate have coarse profile and non-lasered have no coarse profile. Hence, the effect of coarse profile between the lasered and non-lasered being investigated.

NiCrAlY alloys are excellent to resist oxidation and hot corrosion. It is frequently being used as bond coats for ceramic coating like Yttria Stabilized Zirconia (YSZ) to resist corrosion up to 980 °C. Coating and its affect on adhesion to the substrate are closely linked to the coating microstructure; specifically, Kirkendall voids, oxides and intermetallic compounds [5-7].

H13 tool steel has been used as die steel material in semi-solid forming processing. This process involves cyclic high temperature metal injection, solidification and rapid quenching by water based lubricants in the die. Rapid heating and cooling cause die surface to compress and tension respectively and lead to crack and fatigue failure. Hence, it is important to maintain die surface from damage and failure. In order to reduce damage at contact surface, many researches indicate surface treatment and coating are the most effective methods for die surface protection [1,2]. To date, many research works have been conducted to enhance/harden metal surface through coatings and surface modification. Many difficulties were encountered to meet coating requirements such as excellent bonding, adequate thickness, suitable mechanical properties, thermal shock resistance and high temperature stability [3,4]. Therefore, laser surface modifications are prior methods that can improve coating characteristics especially thermal barrier coatings. This paper investigates adhesion of bond coat NiCrAlY on laser modified (LSM) AISI H13 tool steel substrate, its effects on fracture toughness and elements diffusion with the differ of sandblasted setup indicated.

2. Methodology

The material investigated in this study was AISI H13 tool steel being used for the experimental substrate. As received, 25.4 mm diameters with 38 mm length H13 tool steel cylindrical samples were cleaned with ethanol prior to processing. Five cylindrical numbered from sample 28 to sample 32 being laser modified on its flat surface sandblasted at certain setup and being coated with nickel based alloy (NiCrAlY). One sample which was the reference sample being coated with nickel based alloy (NiCrAlY) without the laser modification prior to coating. Table 1 shows the chemical composition of AISI H13 tool steel material as the substrate material being coated with bond coat NiCrAlY.

Table 1
Chemical composition of AISI H13 tool steel substrate and NiCrAlY coating powder

Material	C	Mn	Si	Cr	Ni	Mo	V	P	S	Fe
H13	0.32-0.45	0.20-0.50	0.80-1.20	4.75-5.50	0.30	1.10-1.75	0.80-1.20	0.03	0.03	Bal.
NiCrAlY	Ni 67.0	Cr 22.0	Al 10.0	Y 1.0						

An Nd:YAG JK300 HPS laser system with TEM mode and average power of 500 kW was used to modify the sample surface. The laser system was focused to a minimum laser spot diameter of 0.67 mm onto sample surface and setup its' scanning speed according to the desired overlap. Table 2 indicates the parameter settings for laser surface modification. The parameters used were laser peak power, P_p, duty cycle, *DC* and pulse repetition frequency, *PRF*. The duty cycle was based on laser power to result in average power of 100 W. The outcome parameters from the settings were residence time, T_R and irradiance, *I* which were calculated using Eq. (1) and (2).

$$T_R = \frac{DC \text{ x } d}{S} \tag{1}$$

$$I = \frac{F}{T_R} \text{ x } DC \tag{2}$$

where *d* is laser spot diameter, *F* is energy density (pulse energy divided by laser beam spot area) and *S* is traverse speed (laser spot diameter divided by time taken to produce one rotation).

Table 2
Laser surface modification of H13 steel parameter settings

Sample	P_p, kW	DC (%)	PRF (Hz)	Overlap, n %	Residence time, T_R (ms)	Irradiance, I (W/mm²)
L1	1.8	5.6	50	20	1.390	227

Sandblast PanBlast PB500SP Blast Cabinet Assembly with sandblasting medium of white fused alumina with granulation (µm) of 1400/1700. The process was performed at right angles to the substrate surface and at a constant distance of about 100 mm from the nozzle. Sandblast parameter setting indicated in Table 3 below.

Table 3
Sandblast parameter settings

No.	Parameter	Settings	Unit
1.	Disntance	100+-4	mm
2.	Pressure	5	bar
3.	Time	20, 30, 40, 50, 60	second

Atmospheric plasma spray method was used to deposit NiCrAlY coating. Praxair Ni-164/211 Ni22%Cr10%Al1.0%Y alloy powders were deposited on laser modified surface samples at parameters settings shown in Table 4.

Table 4
Parameter settings for APS deposition of NiCrAlY coating

Parameters	Unit	APS setting for NiCrAlY
Feed Rate	g/min	40
Secondary gas (He)	kPa	345
Primary gas (Ar)	kPa	345
Carrier gas (Ar)	kPa	345
Current	A	500
Stand-off distance	mm	110
No. of cycle	no.	5
Torch speed	%	40
Workpiece rotational speed	rpm	250

Surface roughness and profiles were measured by MahrsurfPS1 Mobile Roughness Measuring Instrument. Whereby, metallographic study were measured using IM7000 inverted optical microscope and being analyzed by using image analysis software. Scanning electron microscope (SEM) and elements diffusion were investigated by Energy Dispersive X-ray Spectroscopy (EDXS) using Hitachi Tabletop Microscope TM3030 Plus. Energy Dispersive X-ray Spectroscopy (EDXS) setup was employed with observation condition of 15kV accelerating voltage were applied to the electron probe and spatial resolution setup with standard backscattered observation mode. Interfacial indentation test (IIT) and Microhardness Vickers was conducted using MMT-X7 Matsuzawa hardness test machine at 0.5 kg load. The IIT was carried out at substrate/coating interface of polished cross-section surface. As shown in Figure 1 indentation load was applied for a period of 10 seconds to include delayed cracks. The adhesion was determined by measuring the length of the radial crack caused by the penetration of the Vickers diamond.

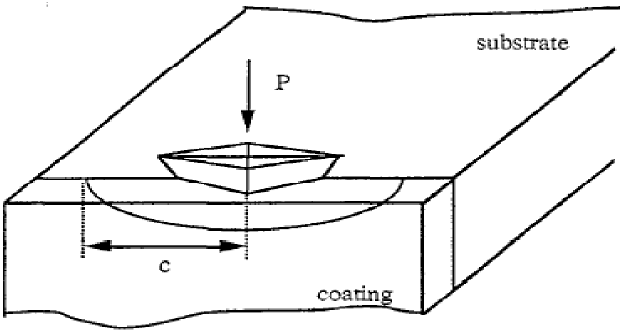


Fig. 1. Principle of Interfacial Indentation Test (IIT)

2.1 Interfacial Toughness, Kc Calculation

The interfacial toughness, K_C was referred to Chicot and Lesage *et al.,* [9], where (P_c, a_c) couple associated to the interfacial crack initiation as given in Eq. (3) and Eq. (4).

$$K_C = 0.015\frac{Pc}{a_c{}^{3/2}}\text{x}\left(\frac{E}{H}\right)^{1/2} \tag{3}$$

where P_c is the applied critical load, and a_c is the interfacial crack length. The $(E/H)_I$ ratio, is defined in Eq. (4), characterises the global behaviour of the coating/substrate system.

$$\left(\frac{E}{H}\right)^{1/2} = \frac{\left(\frac{E}{H}\right)_S^{1/2}}{1+\left(\frac{H_S}{H_C}\right)} + \frac{\left(\frac{E}{H}\right)_C^{1/2}}{1+\left(\frac{H_C}{H_S}\right)} \tag{4}$$

where *E* is the Young modulus, *H* is the hardness and I, S, and C subscripts stand for interface, substrate and coating, respectively.

3. Results

3.1 Laser Modified (LSM) Sandblasted Coated Samples

Samples for laser modified (LSM) sandblasted with NiCrAlY coating showed in Figure 2. Whereby, LSM sandblasted surface profiles prior to coatings for sample 28 until 32 indicated in Figure 3.

Fig. 2. Laser modified sandblasted coated samples

Based on surface profiles from Figure 3 and Figure 4, it was indicated that sample 29 have average roughness of 4.328 µm compare to others.

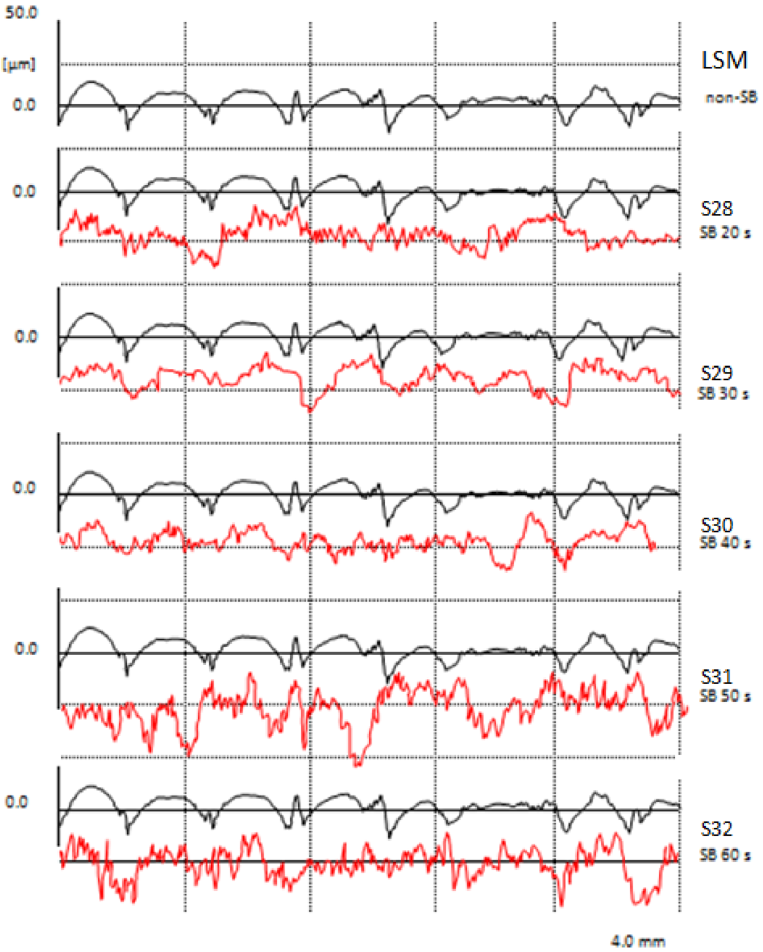


Fig. 3. LSM sandblasted surface profiles prior to coatings

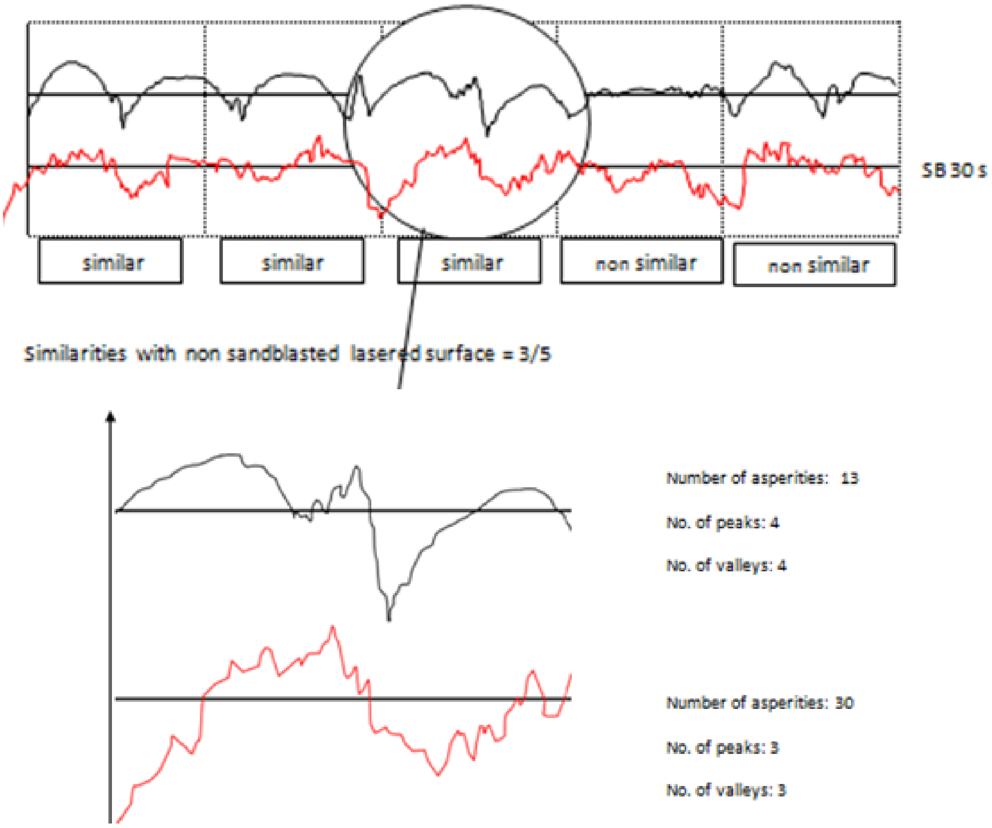


Fig. 4. Surface profile sample 29

In basics of coatings, sufficient substrate surface roughness gives mechanical keying effect on coating interface bonding. Mostly plasma sprayed coatings NiCrAlY adhered on 4 micrometer average roughness using sandblasted process prior to coatings [3]. Average roughness readings, was insufficient enough to state that the coating compromise the roughness preparation prior to coatings. This was proved by the failed coating being plasma sprayed on 4 ~ 5 micrometer average surface roughness without sandblasting take effect. Hence, it was essential for the surface profile to be look detail into on what is the main characteristics that would helped to adhered coating onto substrate's surface [4].

In Figure 5, compare to other samples (28, 30, 31 and 32), between 2 peaks, sample 29 has the lowest peak height and highest amount of asperities. Sample 29 being produced by 30s sandblasted time engaged more materials entrapment, thus better adhesion bonding by physically interlocking the coating-modified substrate interface.

Enhanced interfacial bonding between NiCrAlY coating and laser modified substrate of sample 29 was due to sandblasted surface profile interface geometry as shown by Figure 4. NiCrAlY splats were entrapped within the slopes produce mechanical keying or physically interlocking that allows interface bonding.

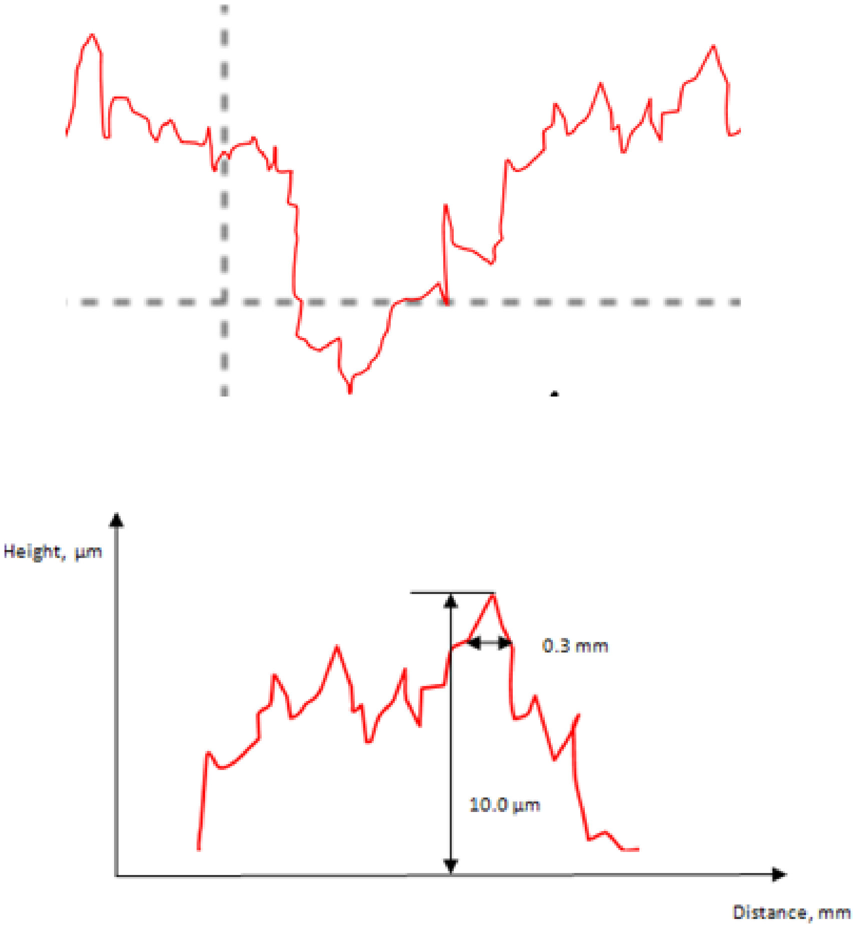


Fig. 5. Asperities between peak to peak and highest height measured for sample 29

3.2 Micro Vickers Hardness Substrate - Bond Coat and Interfacial Toughness, K

Micro Vickers hardness of interlayer from substrate to bond coat (NiCrAlY) was measured and indicated in Figure 6. It shows that hardness in substrate was at average 220 to 280 HV, laser modified layer at average 800 HV decrease to bond coat layer to 600 HV.

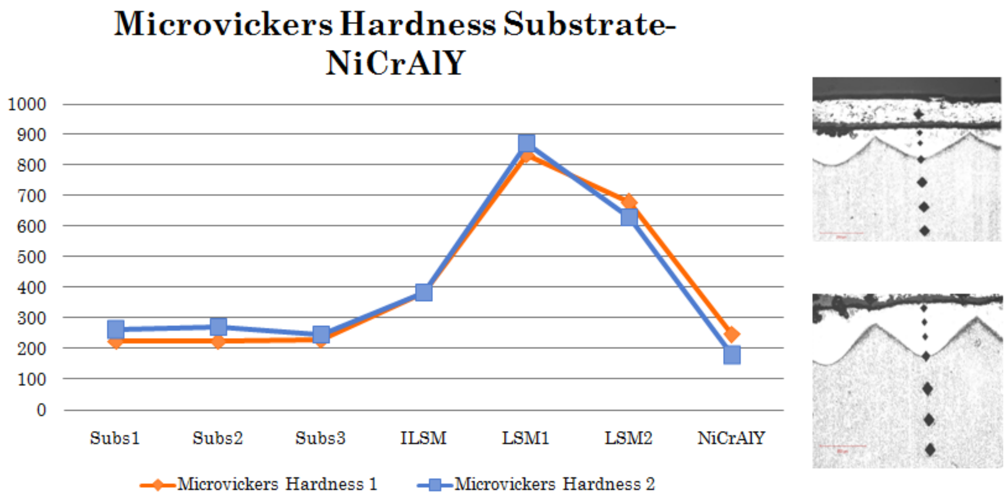


Fig. 6. Micro Vickers Hardness Substrate-Bond coat

Micrograph from interfacial indentation test of reference sample AISI H13 tool steel coated with only NiCrAlY bond coat is shown in Figure 7. A crack length of 69.14 µm and interfacial toughness, K_c = 2.08 MPa $m^{1/2}$was measured at substrate-bond coat interface.

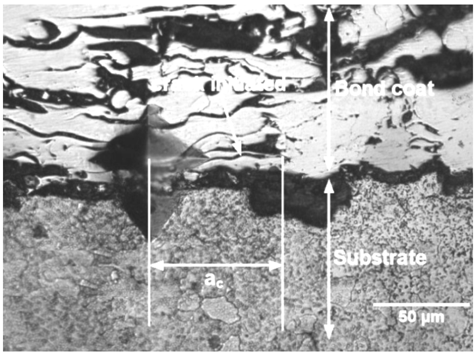


Fig. 7. Micrograph of reference sample cross section with indentation mark at NiCrAlY coating/H13 substrate interface

whereby micrograph for sample 29 from interfacial indentation test of NiCrAlY coating with laser modified layer are shown in Figure 8. The crack length measured for sample 29 was 65.4 µm respectively. Crack initiated from the center of indentation mark and propagated along the NiCrAlY coating/laser modified surface interface. Interfacial toughness, Kc = 2.8 MPa m1/2 was measured.

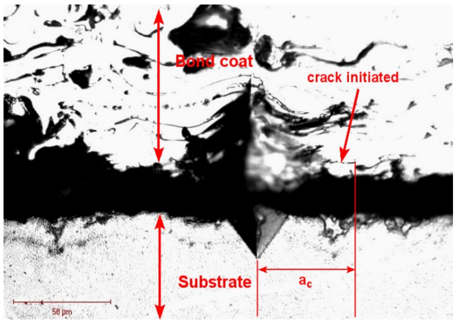
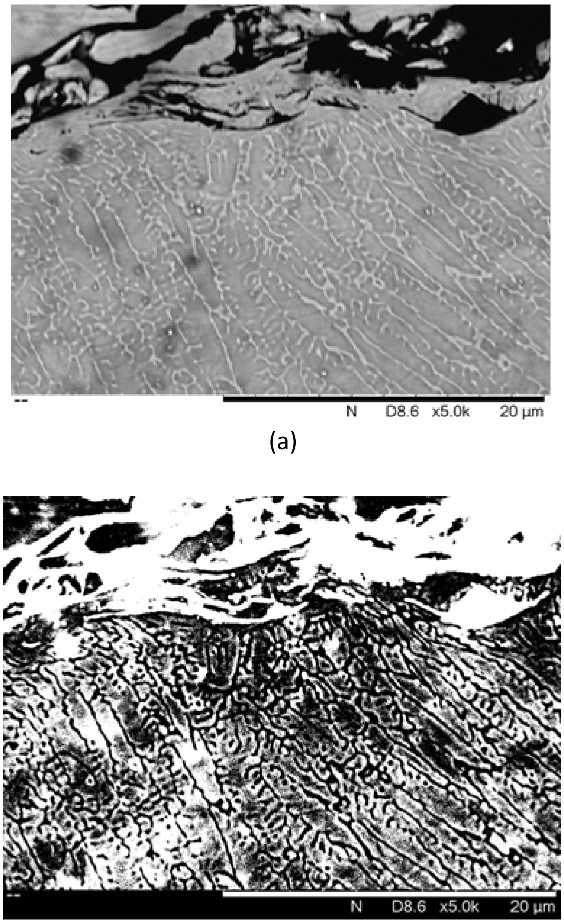


Fig. 8. Micrographs of sample 29 cross section with indentation mark at NiCrAlY coating/laser modified surface interface

Higher K_{avg} value was due to enhanced adhesion bonding between coating and laser modified surface. Fine grains of laser modified substrate are thermally instable when subjected to high temperature NiCrAlY splats. In previous work, the metastable phase formation in laser modified surface varies as a result of laser irradiance and residence time [10]. Lower irradiance at longer residence time or longer exposure time was necessary to allow surface melting to take place. A low heating rate and small undercooling along with longer exposure time to produce martensite phase during processing. The pulse energy and residence time combination determines the suface heating rate and quench rate of undercooled austenite. Such treatment results in the formation of finer grains at the surface which enhances the strength of adhesion bond [11].

3.3 SEM and Energy Dispersive X-ray Spectroscopy (EDXS)

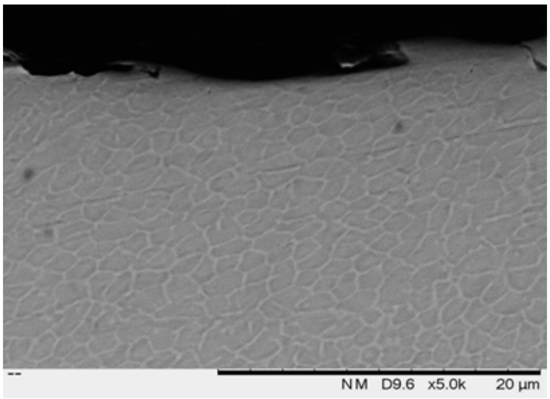
Figure 9(a) and (b) shows SEM image of sample 29 with grains boundaries indicated. Grains pattern near the coated layer has lane pattern compare to sample 30 SEM image in Figure 10(a) and (b).



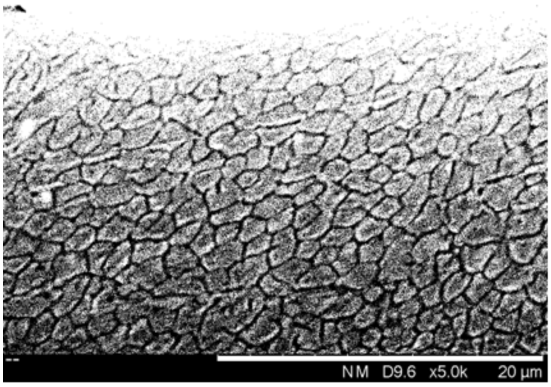
(a)

(b)

Fig. 9. SEM image sample 29



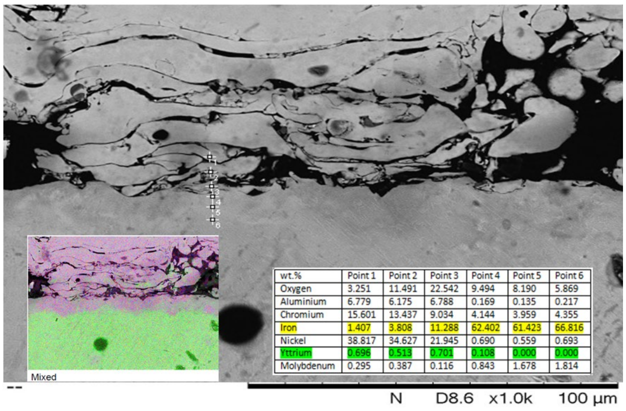
(a)



(b)

Fig. 10. SEM image sample 30

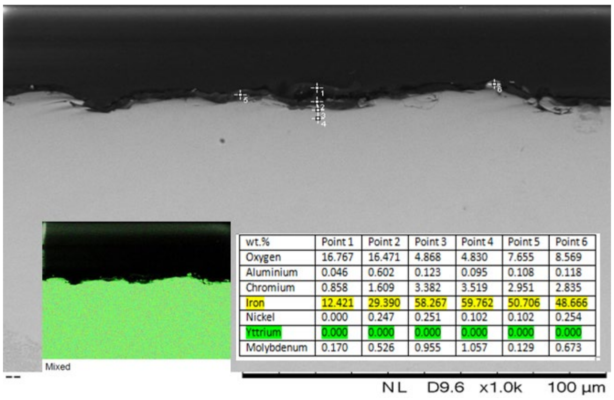
In Figure 11, EDX image of sample 29 shows three layers were observed along the cross sectional depth namely, NiCrAlY coating, laser modified substrate (LSM) and H13 substrate. EDXS analysis detected changes of nickel, chromium, aluminium, oxygen and iron element content along cross sectional depth of bond coat (NiCrAlY) layer and laser modified surface (LSM) layer.



wt.%	Point 1	Point 2	Point 3	Point 4	Point 5	Point 6
Oxygen	3.251	11.491	22.542	9.494	8.190	5.869
Aluminium	6.779	6.175	6.788	0.169	0.135	0.217
Chromium	15.601	13.437	9.034	4.144	3.959	4.355
Iron	1.407	3.808	11.288	62.402	61.423	66.816
Nickel	38.817	34.627	21.945	0.690	0.559	0.693
Yttrium	0.696	0.513	0.701	0.108	0.000	0.000
Molybdenum	0.295	0.387	0.116	0.843	1.678	1.814

Fig. 11. EDX image sample 29

In Figure 12, EDX image of sample 30 shows discoated layers of bond coat (NiCrAlY). Image shows only LSM layer and EDXS analysis detected changes of ferum in LSM layer.



wt.%	Point 1	Point 2	Point 3	Point 4	Point 5	Point 6
Oxygen	16.767	16.471	4.868	4.830	7.655	8.569
Aluminium	0.046	0.602	0.123	0.095	0.108	0.118
Chromium	0.858	1.609	3.382	3.519	2.951	2.835
Iron	12.421	29.390	58.267	59.762	50.706	48.666
Nickel	0.000	0.247	0.251	0.102	0.102	0.254
Yttrium	0.000	0.000	0.000	0.000	0.000	0.000
Molybdenum	0.170	0.526	0.955	1.057	0.129	0.673

Fig. 12. EDX image sample 30

Presence of Fe in NiCrAlY coating indicates diffusion of Fe element from laser modified surface to the coating layer. During plasma spray process, molten NiCrAlY was deposited onto H13 substrate surface which was laser modified beforehand to produce fine grains at metastable phase [13]. The grain refinement in laser modified sample is thermally instable and changes when subjected to high temperature of 650 °C [14]. Therefore, atomic diffusion easily occurred in metastable phase surface as kinetics mechanisms were active during coating process and energy barriers to atomic motion easily overcome [15].

Higher Ni and Cr content found in laser modified surface suggests diffusion of these elements from NiCrAlY coating to the modified surface. During coating deposition, high temperature of NiCrAlY splats energized the modified substrate surface. The energized metastable phase in modified substrate caused grain migration in conjunction with atomic diffusion. Coating layers contained diffused atoms developed strong bonding which is comparable to diffusion coating method.

Coating of NiCrAlY involves high temperature when using plasma spray processing. Thermally grown oxide (TGO) scale starts to form by interaction with oxidizing atmosphere. Formation of alumina, Al_2O_3 in the TGO layer results the changes of Al content in coating layer where Al_2O_3 was formed from the diffusion of Al to TGO layer and finally oxidized with O [16]. The occurrence of rich Ni, Cr outer oxide layer at point D and E in sample 29 associates this process to zero local concentration of aluminum. High content of O implies local activity of oxygen to form other oxides as Ni and Cr oxides. Strawbridge *et al.,* [17] accounted that tensile stress normal to the oxide - metal (Ni and Cr oxides) would develop that would effect in decreasing coating adhesion.

4. Conclusions

Enhanced interfacial bonding effectively in laser modified substrate surface with sandblasted surface profile that produced low peak height and high amount number of asperities between peaks. Interfacial toughness and micro hardness was determined in laser modified substrate of the coated sample. The interfacial bonding toughness was due to metastable phase presence in laser modified and atomic diffusion mechanism between NiCrAlY coating and laser modified surface. The metastable phase in modified substrate induced Fe, Ni and Cr atomic diffusion occurrence in both coating and laser modified layers. These findings introduce enhancement of coating system by substrate surface modification for interfacial adhesion.

Acknowledgement

The authors would like to be obliged to Manufacturing Technology Research Group (MTG), Universiti Malaysia Pahang Al-Sultan Abdullah, Faculty of Mechanical and Manufacturing Engineering, Universiti Tun Hussein Onn, Malaysia and SIRIM Advanced Materials Research Centre, (AMREC), Kulim Kedah for providing laboratory facilities. Not to forget Faculty of Mechanical & Automotive Engineering Technology, (FTKMA), Universiti Malaysia Pahang Al-Sultan Abdullah for providing financial assistance under Fundamental Research Grant Scheme project grant no. RDU120105.

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