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RESEARCH STATUS AND APPLICATION OF THE HIGH-ENTROPY AND TRADITIONAL ALLOYS FABRICATED VIA THE LASER CLADDING

The objective of this paper is presenting a review of high-entropy alloys and traditional alloys fabricated by laser cladding. In this paper, recent developments of different material system are summarized, and the developments in laser cladding for functional coatings with high wear resistance, good corrosion and oxidation resistances, and better medical biocompatibility are reviewed. By summarizing the analysis of microstructure, mechanical properties, corrosion resistance of high-entropy alloys and traditional alloys' coating fabricated by laser cladding, it stated that laser cladding treatment can improve corrosion resistance, homogenize grain size, and increase microhardness and other properties. Laser cladding is considered as the potential method to ameliorate mechanical properties, improve microstructure and repair broken parts. Therefore, laser cladding has the successful applications in automobile and aerospace productions, and shipbuilding due to those advantages.

Keywords: high-entropy alloys, traditional alloys, laser cladding, microstructure, mechanical properties, application.

1. Introduction

High-entropy alloy (HEA), as an emerging material, is developed in one of the most promising choice as surface modification material. The HEA coatings have good advantages such as superior corrosion resistance, excellent thermal stability, high hardness, and better wear performance

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[1-4]. Other metals such as magnesium alloys, Ni-based alloys, and titanium alloys can be also applied in a surface modification. Titanium and its alloys possess high specific strength, excellent corrosion and oxidation resistances, and good biocompatibility [5]. Magnesium alloys have good castability, machinability, high strength-to-weight ratio, and some advantages in price due to the abundant reserves on the earth and in the sea [6]. The aerospace and ship industries are equipped with Ni-based alloys due to those good advantages. Nowadays, relevant researches are needed in alloy processing and surface treatment techniques, and various surface-modification techniques have been adopted to improve the mechanical properties of alloys, such as chemical conversation, physical vapour deposition, diffusion treatment, corrosion resistance, and broken-parts' repairing [7].

In recent decades, laser alloying, laser re-melting, laser cladding, laser shot peening and so on have gone through rapid developments owing to the high energy density [8-12]. Especially, laser cladding owns many potential advantages, such us low dilution and limited heat effects on base metal, integral metallurgical bond, minimal warpage and distortion of clad components, reduced cracking susceptibility, suitability for full automation and possibility of developing coatings with nonequilibrium microstructures and superior physical properties [13]. Laser cladding synthesizing HEA coatings is a potential method in manufacturing industry. Compared to the traditional methods like magnetron sputtering, electrospark process, plasma arc cladding and casting method, laser cladding shows greater advantages: (i) larger thickness coating, (ii) reduced density, (iii) lower cost, (iv) optimized structure [14]. The laser cladding considered as a repair method has enough potential to improve the performance of remanufactured parts. Therefore, compared with traditional repair methods, laser cladding has big potential development in broken-parts' repairing [15]. The process of laser cladding can be carried out in three ways: pre-placed powder, wire feeding and powder injection, which is talked about in details in the following.

At present, laser cladding has developed into a promising method to improve some properties or repair complex geometric components. This paper reviews the research of HEAs and traditional alloys coating manufactured by laser cladding, including analysis and comparisons of microstructure and mechanical properties, and the successful application of laser cladding for automobile, aerospace and ship. The current problems were put forward, and the prospect to the future development tendency is made. By summarized the development status, this paper is expected to play a guiding role for related studies about laser cladding.

2. Laser Cladding

Laser cladding combining the laser technology, computer aided manufacturing (CAM) and the control system together is a rapid prototyping technology [16]. Laser cladding is a hard facing process that uses a high-powered laser beam to melt the coating material and a thin layer of the substrate to form a pore- and crack-free coating (of 2–50 mm thick) with low dilution that is perfectly bonded to the substrate [17]. The process may be used for improving hardness and surface-dependent properties such as wear, corrosion, oxidation, and, to some extent, fatigue resistance [17]. The principle of laser cladding with coaxial powder injection was shown in Fig. 1 [10]. Based on the material feed stock, laser cladding systems are divided into three categories: (i) laser cladding with pre-placed powder, (ii) laser cladding by wire feeding [17], and (iii) laser cladding with powders injection.

2.1. Laser Cladding with Pre-Placed Powder

The powder is preplaced in the surface of substrates and then the laser beam based on the established procedures selectively scan powder which can be seen from Fig. 2 [18]. Before laser cladding, powder and substrates prefer to being dried for a while in order to get good shape and properties.

2.2. Laser Cladding via the Wire Feeding

Feeding wire from nozzle into the deposition melt pool can be applied in laser cladding technology. The schematic diagram of laser cladding by wire feeding is shown in Fig. 3 [13]. Some research about the effect of wire feeding angle and deposition efficiency on surface quality shows that $40^{\circ}\pm15^{\circ}$ in the vertical plane, low values of feeding angle in hori-



Fig. 1. The illustration of laser cladding with coaxial powder injection [10]





zontal plane and high deposition efficiency can produce better surface quality [19]. Compared with powder injection, less porosity was found in the wire feeding samples [19].

2.3. Laser Cladding with Powder Injection

A defocused laser beam is employed in laser cladding, under the function of which, preplaced or synchronous feeding powders as well as a thin layer of the substrate will melt and solidify rapidly, forming a metallurgical bond with the substrate [20]. In this process, laser power, laser beam size and laser scanning velocity or specimen motion speed all affect the quality of laser cladding coating. The schematic diagram of laser



Fig. 4. Schematic diagram of laser cladding with powder injection [21]

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cladding with powder injection can be observed in Fig. 4 [21]. When we apply this technique, the following issues must be noticed [22]: (i) the size and order of laser beam should be adjusted properly to ensure good quality and low divergence; (ii) the speed of powder feed is important factor; (iii) the base material or substrate should have a proper fixed mode.

2.4. Factors Affecting the Laser Cladding

In order to achieve the desired properties, a good control of the process parameters is indispensable. There are numerous factors affecting the quality of laser cladding, such as the laser power (P), laser beam size (beam diameter D) and laser scanning velocity (V) [23]. Authors of Ref. [24] pointed that the laser parameters are related to the level of dilution by the substrate and a minimum amount of dilution can promote a partially solid melt pool and maintain a high volume fraction of TiC in the clad. As found in Ref. [25], with the scanning velocity increased to 0.065 m/s, the wear resistance performance began to increase and the wear resistance with high scanning velocity or low scanning velocity is very poor. However, other authors [26] pointed out that the composite coatings fabricated in higher laser scanning velocity exhibited finer microstructure, higher microhardness and thinner thickness. The effect of laser parameters on different chemical composition and temperature is not the same. A series of experiments were carried out in constant

Laser cladding systems	Alloys	Materials
pre-placed powder	high-entropy alloy	6FeNiCoSiCrAlTi [27] TiZrNbWMo [28] 6FeNiCoCrAlTiSi [29] FeCoCrNiCu _x [30] NiCrBSi/WC [31] FeCrCoNiTiAl [32]
	others	Co42 + TiN [33]
wire feeding	high-entropy alloy	—
	others	Inconel 625 [34] Ti-6Al-4V [35]
powder injection	high-entropy alloy	AlCoCrCuFeNi [36] Ni-Cr-Co-Ti-V [37] FeNiCoAlCu [38] AlxCoCrFeNi [39]
	others	WC/W ₂ C [35] Cr-Ni-based stainless steel [40] Ti-6Al-4V [41]

Table. Materials and corresponding laser cladding systems

laser power, constant laser scan speed, constant effective energy and constant powder deposition density. The microstructure of clad could be widely varied by using different laser parameters. Even the same combined laser parameters still produce different microstructures if the laser power were different. Therefore, the optimization of process parameters is significant for the laser cladding.

3. Materials

After intense development and exploration, laser cladding has become potential technology to perfect microstructure and mechanical properties. In its early development stage, laser cladding was applied to melt normal alloy powders (wire) on substrate and many new alloys especially highentropy alloys have been manufactured by laser cladding. Table lists some materials that can be processed by laser cladding.

3.1. High-Entropy Alloys

Laser cladding in high-entropy alloys (HEAs) manufacturing industry is considered as a new treatment technology. Choosing laser cladding technology with high energy density, fast heating speed and small thermal effect on substrate to produce HEA can obtain excellent properties and full dense metallurgically bonded layer as well as less microscopic defects [42]. Due to those advantages, manufacturing industry will be more inclined to laser cladding processing HEA. In addition, there are lots experiments studying different HEA microstructures, mechanical properties and the effects on them.





Fig. 6. Microstructure distribution of a coating [27]

3.1.1. Microstructure and Mechanical Properties

The HEA cladding layer traditionally consists of cladding zone, bounding zone and heat affected zone, which can be seen from Fig. 5 [43]. The microstructure of each zone is as follows. (1) Cladding zone is composed of equiaxed grains and some columnar grains (Fig. 5, *b*, *c*). (2) Bounding zone is the transition portion between cladding zone and heat affected zone, which mainly compose by equiaxed grains and nanocrystallites (Fig. 5, *d*). (3) Heat affected zone is close to the bounding zone whose microstructure is similar to substrate (Fig. 5, *d*) [42, 43]. The microhardness of coating is much higher than the substrate, such as 6FeNiCoSiCrAl-Ti prepared by laser cladding as shown in Fig. 6 [27]. That is to say cladding layer effectively enhanced the surface hardness of components to some extent.

Although high-entropy alloy coating can improve surface hardness of components, some problems still exist in cladding layer. Some experiments prove that after annealing at proper temperature more secondary phase formed in the interdendritic regions leading to increase of microhardness [28]. The structure of high-entropy AlCoCrCuFeNi alloys is characterized by the existence of an interdendritic Cu-rich phase [44].

Mo replacing Cu element was intended to overcome the segregation of Cu and to increase microhardness and wear resistance [45]. The microstructure of CoCrBFeNiSi coating shows that (1) black nanosize phase was embedded into the grey matrix

Fig. 7. The microstructure of CoCrBFeNiSi coating by laser cladding [46]





Fig. 8. Potentiodynamic polarization curves for high-entropy alloys AlCrFeCoCu (prepared by laser cladding) in 1 mole/l NaCl solution [1]

in the upper layer and (2) the columnar dendrites were about 10 μ m in length and strip-shaped intergranular precipitates were uniformly distributed in the bottom layer, which can be observed from Fig. 7 [46]. High-temperature wear mechanism of the amorphous layer and the crystallized layer were mainly abrasive wear and mainly adhesive wear, respectively, which is caused by different microstructure component in different layer [47]. Columnar grain also can be observed at the bonding zone of the Al₂CrFeNiMo_x coating and equiaxed grains can be found in the cladding zone, which is similar to other high-entropy alloy coatings [48]. Meanwhile, the experiment shows that with the addition of Mo element, the diffusion of Mo atoms to the grain surface and grain growth are promoted, and the hardness is decreasing from the surface to substrate [47]. To some extent, the wear resistance of high-entropy Al₂CrFeNiMo_x alloy coatings increases greatly [47].

In the work [29], authors found that, after annealing with temperature less than 750 °C, the 6FeNiCoCrAlTiSi coating has high thermal stability and the slightly decreased resistivity, but the microhardness almost remains unchanged, after annealing more than 750 °C, the microhardness of the coating slowly decreases with the increasing decomposition rate of bcc solid solution [29]. The microstructure morphology is related to the value of G/R (G is a temperature gradient, R is a growth rate). While as the value of G/R gradually decreases from the solid-liquid interface to the centre of the molten pool, the microstruc-



Fig. 9. SEM micrographs showing: (a) cross-section of NiCrBSi with 50% angular WC; (b) cross-section of Stellite 21 with 50% spherical tungsten carbides; (c) cross-section of M2 tool steel matrix material with 50% TiC; (d) cross-section of M2 matrix material with 30% VC; (e) cross-section of M2 matrix with 50% VC; (f) cross-section of Inconel 625 with 50 vol.% of chromium carbides [50]

tural morphology changes from planar to columnar grains, which are nearly perpendicular to the interface. As for the cladding zones, the microstructure consists of cellular dendritic grains because of faster nucleation in the top surface layer of the HEA coatings [48]. Wear of the high-entropy alloy coatings has improved greatly attributing to the combination of the hard Fe_nNb-type laves phase and ductile f.c.c. solid matrix. It also reveals that with the higher laser power, it raises the temperature of the molten pool leading to the serious dilution effect of Fe atoms [48]. Some experiments about high-entropy alloy coating with laser remelting shows that the hardness and wear resistance of NiCrCoTiV HEA coatings prepared by combining with the technologies of laser cladding and laser remelting are improved [37]. With Cu element adding into high-entropy FeCoCrNi alloy coating, it enhanced the Gibbs free energy of the cladding layers and the Cu element is easier to segregate in the grain boundaries [30]. The synthesized high-entropy FeNiCoALCu alloy coating is composed of f.c.c. and b.c.c. solid solution phases with a typical uniform dendrite microstructure [38]. After different annealing process, the results show that there is no phase transformation existing from room temperature to 780 °C, proving the good thermal stability [38]. The Al element has a big effect on properties and microstructure of high-entropy coating. As revealed in Ref. [39], the increasing Al content in Al_cCoCrFeNi results in a transformation from f.c.c. to b.c.c. solid-solution crystal structures, and the microhardness is also improved by increasing the content of Al element [39]. Many heat treatments relieve residual stress, increase the fracture toughness, and significantly



Fig. 10. Microstructure of laser cladding layer of W particles with different sizes: $75 \mu m$ (a) and $23 \mu m$ (b) [51]

reduce the cracking susceptibility of the coatings [31]. Reductions in cracking susceptibility were closely related to the microstructural variations of the coatings. With adding B and C elements, the microhardness has been reduced, but the cracks have been eliminated successfully in the coating [32].

3.1.2. Corrosion

The corrosion resistance of laser cladding layer was investigated in Ref. [1], where the high-entropy AlCrFeCuCo alloy has exhibited an excellent corrosion resistance performance. As shown in Fig. 8, with the scanning speed increasing, the corrosion resistance firstly increased and then reduced [1]. The corrosion resistance of the FeCoCrNiCu_x cladding layers was deteriorated and the high-temperature oxidation resistance of the cladding layers reduced with the increase of the content of Cu element [30]. The formation of oxide film may be the main reason for the good high-temperature wear performance. The surface oxidation film of laser-cladded high-entropy FeNiCoALCu alloy coating at 800 °C mainly contains Al_2O_3 , Fe_2O_3 , Fe_3O_4 and CuO [38].

3.2. Other (Traditional) Alloys

3.2.1. Microstructure

When laser beam melt alloys into a melt pool on the surface of the substrate, the reinforcement occurring in coating. For example, Ni-based composite coatings manufactured by laser cladding with pre-placed B_4C and NiCrBSi powders was investigated that interface occurs between coating and substrate and reinforcements of TiB₂ (C1), TiC (C2) and CrB (C3) leading to high microhardness [49]. On metal matrix composites (MMC) manufacturing, metal matrix powder with adding different percent (or shape) of carbide powders was melted by laser cladding on



Fig. 11. Microhardness profile of the typical single laser clad bead of Inconel 625 wire [34]



Fig. 12. Polarisation curves of Inconel 625 laser clad layers and 304L stainless steel substrate in de-aerated 3.5 wt.% NaCl solution at room temperature [34]

substrate. As Figure 9 shows, the percent, shape and variety of carbide affect dissolution in metal matrix, as well as the melting point the matrix [50]. In order to investigate the effect of adding alloys powder size on microstructure, two-size W particles were added into powders and

then melting on Q235 steel [51]. The W particles of smaller sizes generate more WC, which can be observed in Fig. 10 [52]. This research group also found that powder with small particle size has refined grain with lots dislocation defects [53].

The microstructure of stainless steel coating made by laser cladding is mainly composed of dendritic structures growing up under the high solidification and cooling rate [40]. The experiments also reveal that uneven distribution of heat delivered by the moving laser beam promotes the formation of micropores [41]. Compared to the conventional bulk forming techniques (such us forging, welding and extruding), laser cladding can obtain the dense, homogeneous and ultrafine structures with a minimum distortion and dilution [40, 53–55].

3.2.2. Mechanical Properties

Mechanical properties of cladding layer can be improved by laser cladding melting alloys on substrates. The low temperature phase change alloy powders with LTT1, LTT2 and 17-4PH stainless alloy powders were adapt to investigate the residual stress and mechanical properties of the cladding layer after laser-cladding fusion, and the results found that cladding layer with high density has better wear resistance and higher microhardness than substrates [52]. For example, the results of the Inconel 625 cladded-layer microhardness testing show that the cladder layer has higher hardness than the substrate, which can be observed from Fig. 11 [34]. Almost all microhardness tests have similar tendency, namely, cladding layer has higher hardness than the substrate [40]. The value of microhardness is not related to microstructure but also the feeding material. A comparative study of Inconel 625 laser cladding by wire and powder feedstock demonstrated that the typical powder laser





Fig. 13. Remanufacturing of cylinder head laser cladding [62]: crack (a), laser cladding (b), the sample after laser cladding (c), and the sample after repair (d)



Fig. 14. (a) Laser cladding Inconel 625 alloy powder onto Inconel 713 turbine blade; (b) the built-up layer of clad material; (c) turbine blade whose tip was formed by the laser cladding process courtesy of Sifco Turbine Components (Cork) Ltd. [22]

track possessed higher hardness (245 $HV_{0.3}$) compared with the corresponding wire laser track (224 $HV_{0.3}$) due to finer dendritic microstructure and higher number density of interdendritic precipitates [56].

3.2.3. Corrosion

The corrosion resistance is related to the effect of Fe dilution on the laser deposited coatings. Authors of Ref. [34] investigated that the corrosion performance of the Inconel 625 wire laser coating at 4.5% Fe dilution is similar to that of the wrought Inconel 625 alloy, but the corrosion performance was decreased when the Fe dilution increased to 12% [34], as shown in Fig. 12. The facts that the laser cladding can provide good corrosion resistance have been reconfirmed in Ref. [40].

4. Applications

The development of innovative, new non-traditional process has progressed greatly in recent years, yielding broader and broader industry applications. Laser cladding owing unique advantages is particularly suitable for producing or repairing special parts. The following review of laser cladding in the automobile, aerospace, shipping and others.

4.1. Automobile

In the early stage, laser was used in welding and cutting for welding repairing, as a traditional remanufacturing method, seriously affect the substrate leading to higher residual stresses and the weld seam position been susceptible to a secondary crack. However, laser cladding has higher forming accuracy, lower substrate thermal effect zone, and outstanding mechanical properties [57]. Due to those advantages of laser cladding, it is considered as the promising remanufacturing technique of the volume damage of failure parts, such as cylinder heads, camshafts and turbine blades [58–61]. Some institutes employed laser cladding on cylinder head repairing and obtained good performance as shown in Fig. 13 [62]. Puguang natural gas purification plant (in China) successfully applied laser cladding to repair the cylinder joint surface deformation, and the repair surface of the bonding layer is not defective [63].

4.2. Aerospace

Aerospace broken components often have complex geometries and high strength, which are difficult, costly and time consuming to repair. During laser cladding process, molten allow is feely to spread and freeze over a workpiece surface due to suitable hard-facing alloys. The injected powder laser cladding technique is used for repairing a broken aerospace turbine blade and a tip of high-pressure turbine blade, which can be seen from Fig. 14 [22]. Those parts of turbine blade after repaired by laser cladding can prolong working time and improve mechanical properties. In the turbine industry, laser cladding can produce fine microstructures with low dilution, high hardness comparing to traditional methods, hence, it is considered as the promising cladding technology [64]. As pointed out in Ref. [65], the laser cladding process could be used to repair damaged 7xxx series aluminium alloy components and a certification process should be developed in order for repaired components to be re-installed with confidence in aircraft. High-strength steels (such as AISI 4340 with wide application in aircraft) can be also repaired through the laser cladding [66].

4.3. Shipping

The unremitting working conditions and intensive wear of marine diesel engine crankshaft main and crankpin journal surfaces, along with potential lubrication failures, easily cause ridges, cuts, grooves, tears, marks and formation of a built up edge [67]. In-situ crankshaft laser cladding is a promising technology with good financial prospects and remediation effect for ship-repair enterprises [67]. To increase the lifetime of marine propellers made of HBsC1 alloy, the laser cladding with automatic wire feeding has been done in Ref. [68], where authors obtained a good clad layer without cracks and with low dilution with the optimum processing parameters.

5. Conclusion

The high-entropy alloy and traditional alloys coating of laser cladding have high hardness and good heat resistance, corrosion resistance and wear resistance. The research about alloy coating manufactured by laser

cladding has also been well studied. However, due to the extremely fast heating and cooling rate of the laser cladding, it is bound to cause a difference in temperature gradient and thermal expansion coefficient between the cladding layer and the base material, which may cause various defects in the cladding layer. This paper reviews the research of HEA and other alloys coating manufactured by laser cladding, including analysis and comparisons of microstructure and mechanical properties, and the successful application of laser cladding for automobile, aerospace and ship. The current problems were put forward and the prospect to the future development tendency is made. By summarizing the development status, this paper is expected to play a guiding role for related studies about laser cladding.

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СТАН ДОСЛІДЖЕНЬ ТА ЗАСТОСУВАННЯ ВИСОКОЕНТРОПІЙНИХ І ТРАДИЦІЙНИХ СТОПІВ, ВИГОТОВЛЕНИХ СПОСОБОМ ЛАЗЕРНОГО ПЛАКУВАННЯ

Метою даної статті є представлення огляду стопів з високою ентропією та традиційних стопів, виготовлених із використанням методи лазерного плакування. У статті коротко викладено останні розробки систем різних матеріялів, а також розглянуто розробки в області лазерного плакування для функціональних покриттів з високою зносостійкістю, хорошою стійкістю до корозії, окиснення та кращою медичною біосумісністю. Підводячи підсумки аналізи мікроструктури, механічних властивостей, корозійної стійкости стопів з високою ентропією та покриттів із традиційних стопів, виготовлених за допомогою лазерного плакування, показано, що воно може поліпшити корозійну стійкість, гомогенізувати розмір зерна, збільшити мікротвердість, а також поліпшити деякі інші властивості. Продемонстровано, що лазерне плакування є ефективною методою поліпшення механічних властивостей, мікроструктури та відновлення пошкоджених деталів. Таким чином, завдяки цим перевагам лазерне плакування успішно застосовується в автомобільній і аерокосмічній промисловостях і суднобудуванні.

Ключові слова: високоентропійні стопи, традиційні стопи, лазерне плакування, мікроструктура, механічні властивості, застосування.

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СОСТОЯНИЕ ИССЛЕДОВАНИЙ И ПРИМЕНЕНИЯ ВЫСОКОЭНТРОПИЙНЫХ И ТРАДИЦИОННЫХ СПЛАВОВ, ИЗГОТОВЛЕННЫХ ПОСРЕДСТВОМ ЛАЗЕРНОГО ПЛАКИРОВАНИЯ

Целью данной статьи является представление обозрения сплавов с высокой энтропией и традиционных сплавов, изготовленных с использованием метода лазерного плакирования. В статье кратко излагаются последние разработки систем различных материалов, а также рассматриваются разработки в области лазерного плакирования для функциональных покрытий с высокой износостойкостью, хорошей устойчивостью к коррозии, окислению и лучшей медицинской биосовместимостью. Подводя итоги анализа микроструктуры, механических свойств, коррозионной стойкости сплавов с высокой энтропией и покрытий из традиционных сплавов, изготовленных с помощью лазерного плакирования, показано, что оно может улучшить коррозионную стойкость, гомогенизировать размер зерна, увеличить микротвёрдость, а также улучшить некоторые другие свойства. Продемонстрировано, что лазерная наплавка является эффективным методом улучшения механических свойств, микроструктуры и восстановления повреждённых деталей. Таким образом, благодаря этим преимуществам лазерное плакирование успешно применяется в автомобильной и аэрокосмической промышленностях и кораблестроении.

Ключевые слова: высокоэнтропийные сплавы, традиционные сплавы, лазерное плакирование, микроструктура, механические свойства, применение.