

Laser Directed Energy Deposition based additive manufacturing of metallic multi-material: A Review

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Abstract : Laser Directed Energy Deposition (LDED) based additive manufacturing has emerged as one of the advanced manufacturing techniques for fabricating components with multi-materials or functionally graded materials (FGMs). Multi-materials and FGMs are classes of advanced materials designed to achieve intended functionality by varying composition, microstructure, and phase formation across the volume of a component. In this regard, the fabrication of metallic multi-material components or FGMs by LDED is one of the enduring research topics for various engineering applications. This paper provides a brief overview of the current state of art and issues associated with LDED of different combinations of metallic materials. It also includes the research work carried out on LDED of multi-material components using the indigenously developed LDED system at Raja Ramanna Centre for Advanced Technology, Indore. The paper will be a quick start for novices and researchers interested in the LDED of multi-material components.

Keywords : Additive manufacturing; Laser directed energy deposition; Functionally graded material; Multi-material.

1. INTRODUCTION

Multi-material components are designed to meet the prerequisites of extreme environmental duty conditions by combining two or more materials in a single component[1]. These prerequisites can be achieved by varying spatial chemical composition, microstructure and phase formation across the volume of a component. One of the examples is the pressurized heat exchanger, where the stainless steel (SS) tube carrying hot water is expected to transfer heat at a faster rate. However, due to the low thermal conductivity of SS, the desired heat transfer rate cannot be achieved. This limitation can be overcome by deploying a multi-material structure of SS and Cu offering adequate strength to sustain the internal pressure due to the presence of SS along with improved heat transfer due to Cu addition. Thus, by deploying a multi-material component, the intended functionality can be accomplished at extreme duty conditions.

Out of various material combinations, the fabrication of multi-metallic components is one of the enduring research topics for various engineering applications such as power generation units, space research, cryogenic applications, tooling industry, automotive industry, etc.[2][3]. These multi-metallic components are designed by combining materials of desired properties as per intended functionality. The different multi-metallic systems used for various engineering applications are listed below

- Nickel (Ni)-Cu is used in marine environments due to improved strength, conductivity and anti-corrosion properties[4].
- Aluminium (Al)-SS is used in the aviation industry due to its high strength to weight ratio[5].
- Tungsten Carbide (WC)-SS combination offers higher wear resistance, improved corrosion and mechanical strength. This makes it suitable for manufacturing steam turbines, pipelines, valves etc.[6].
- Alumina(Al₂O₃)-tungsten (W) is used for insulating the column in the fusion accelerator [7],
- SS-Ni based multi-material structures offer high-temperature wear and corrosion resistance for applications in nuclear and power generation units, automotive sectors, etc.[8].

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As mentioned above, the intended functionality of multi-material components is achieved by varying spatial compositions or structures. The spatial variation can be a direct/steep change or gradual/diffused change, which are known as bi-metallic material or Functionally Graded Material, respectively (as shown in Figure 1). Bi-metallic multi-material structures follow the sharp variation in compositions or structures. This sharp transition in compositions causes different responses at the interface when operating under different loading/ thermal conditions leading to early or premature failure of components or structures at the interface. Thus, a diffused interface or functionally graded material (FGM) approach is adopted to overcome the issues associated with the sharp interface.

The concept of FGM was first introduced in 1980 by a Japanese researcher to design a thermal barrier for a space research programme [9]. Conventional fabrication techniques to achieve the spatial variations in composition/ microstructure include powder metallurgy, chemical vapour deposition, centrifugal casting and welding[10]. It is observed that these conventional techniques suffer from a slower production rate, lower bond strength, inadequate shape design freedom, and limited material design freedom[11][12].

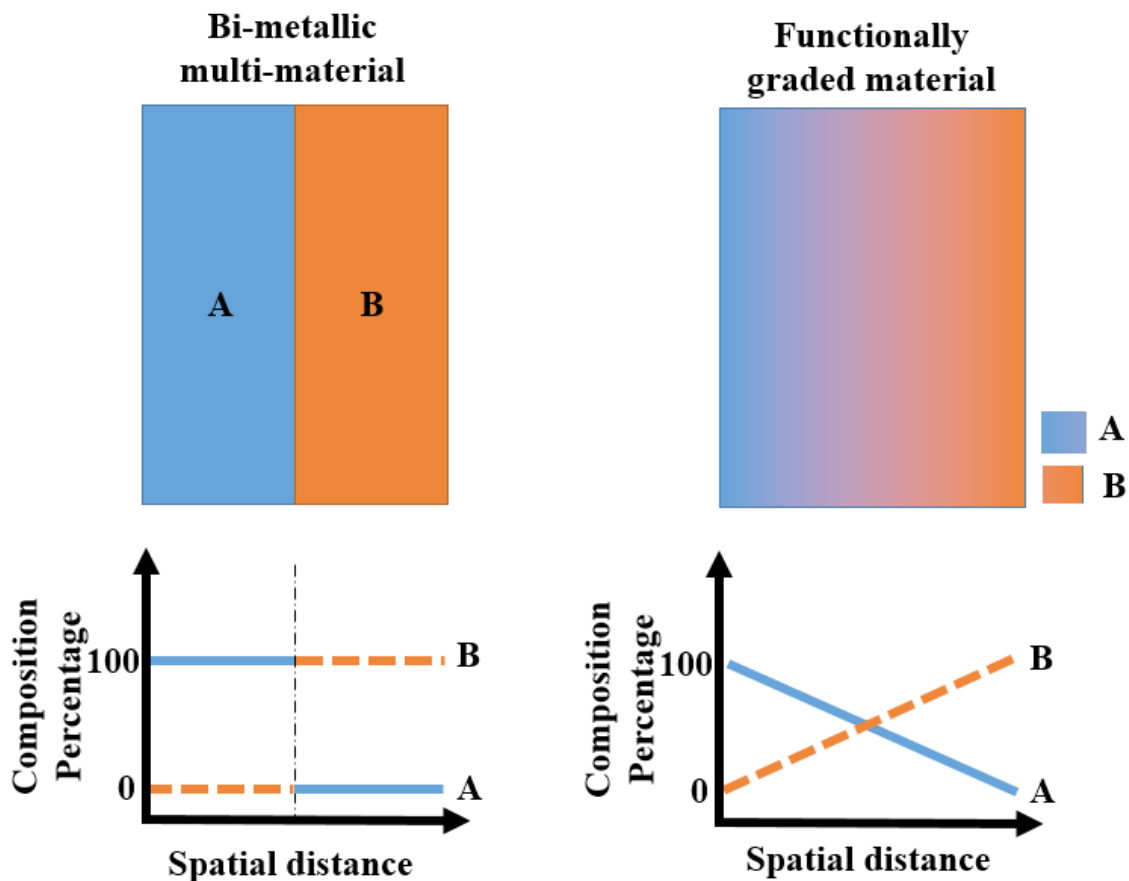
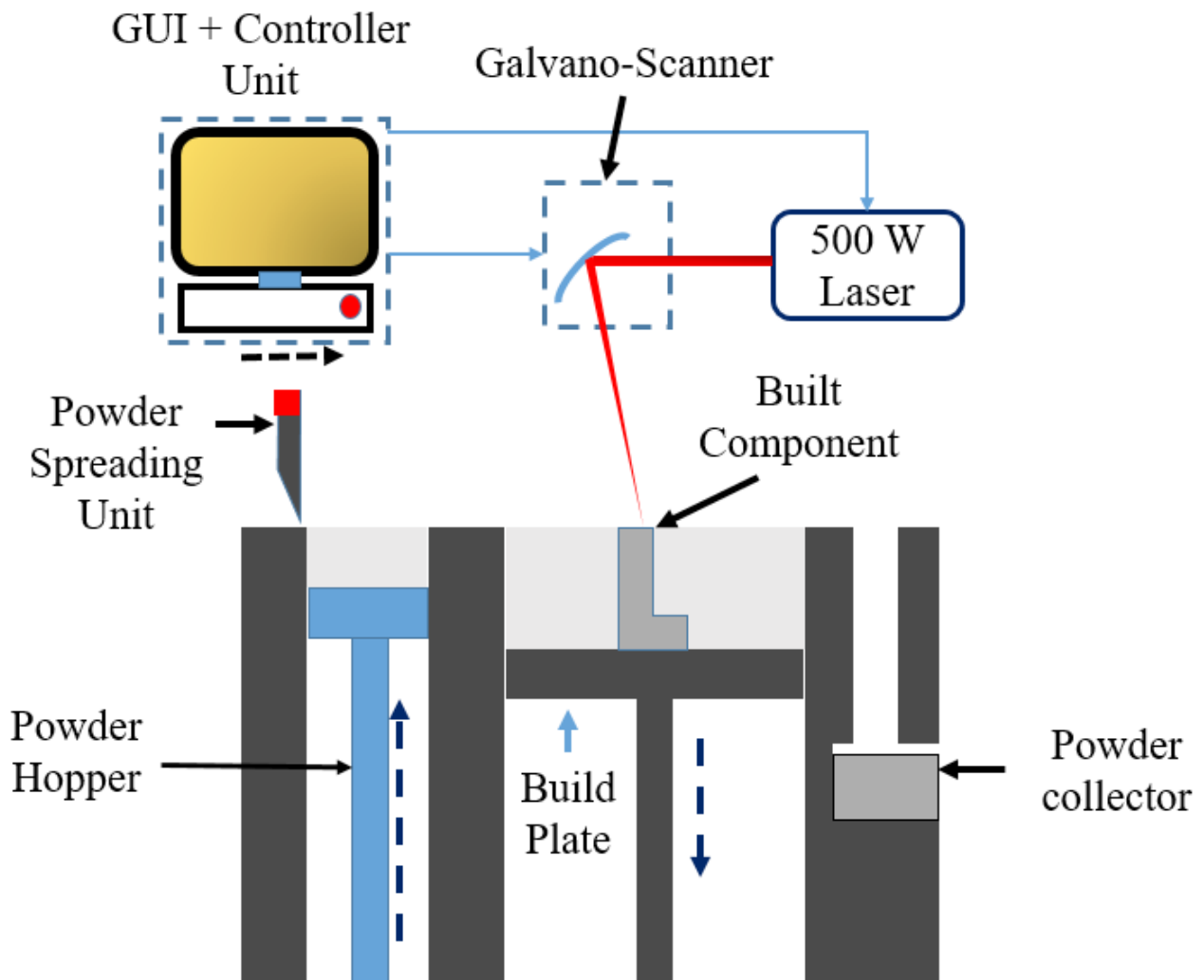


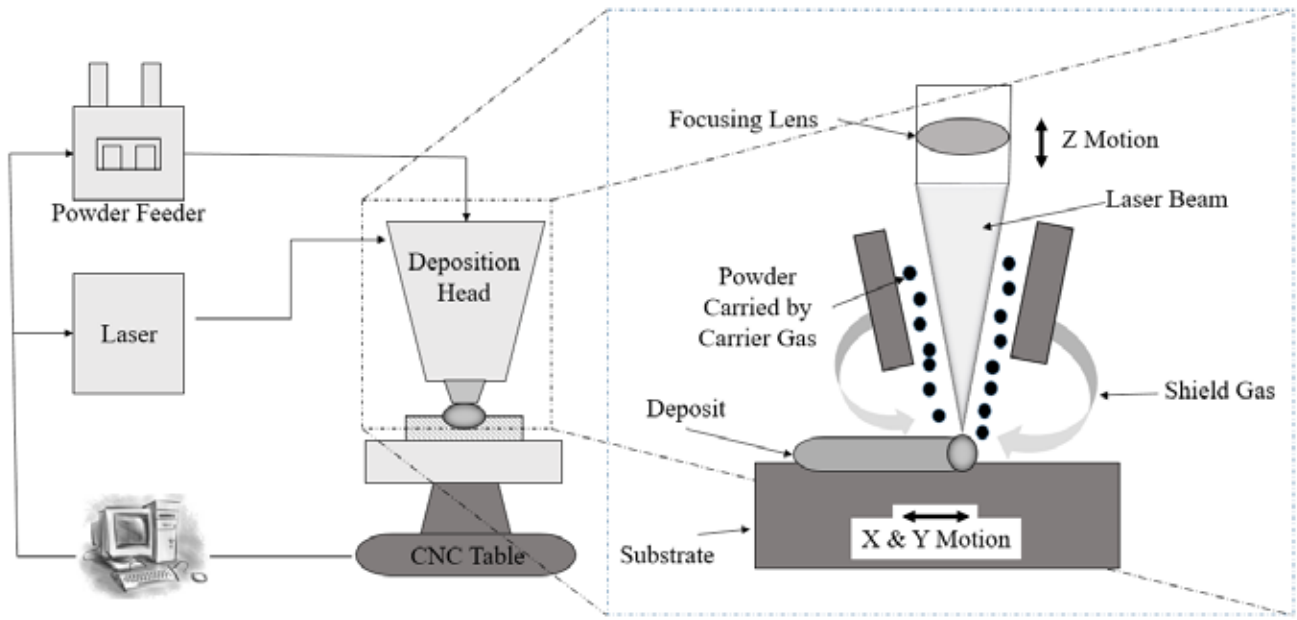
Figure 1: Spatial variation in composition of bimetallic and functionally graded Multi-material

Unlike the conventional fabrication techniques, Additive Manufacturing (AM) possesses capabilities to fabricate fully dense, near net-shaped, complex-shaped and multi-material engineering component in a layer-by-layer fashion. Out of various AM techniques, Laser Additive Manufacturing (LAM) is one of the most widely preferred techniques for fabricating metallic engineering components[13]. LAM consolidates feedstock material into a 3D product in a layer-by-layer fashion using a high power laser beam as the energy source. LAM is broadly classified

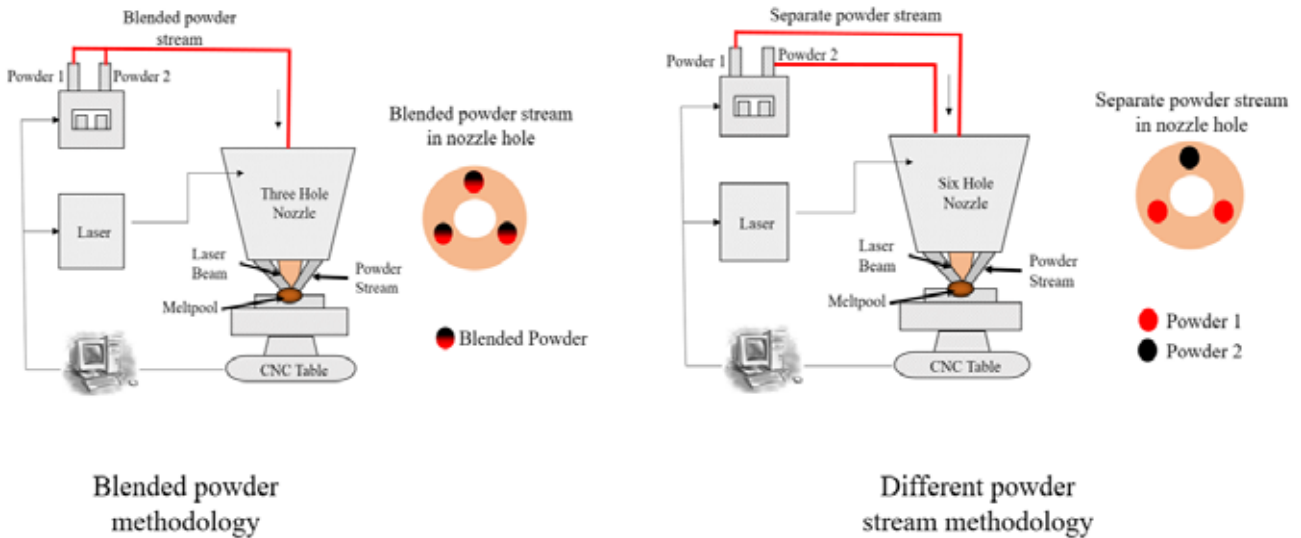
into two processes i.e., Laser powder Bed Fusion (LPBF) and Laser Directed Energy Depositions (LDED). These are classified based on the raw material feeding methods and consolidation technique (refer to Figure 2(a) and (b)). In LPBF, the feedstock powder is spread and melted selectively using a laser beam to consolidate the material as per the Computer-Aided Design (CAD) model input. Further, layer by layer selective melting and consolidation leads to the development of a 3D product[14]. Whereas, in LDED, the raw material (in wire or powder form) is fed into the melt-pool generated on the substrate or previously deposited layer. The subsequent melting and solidification of the fed raw material lead to the formation of a layer as per the design of the first layer. The layer-by-layer consolidation of 2D shape leads to a 3D product[15]. LDED is the most preferred technique for building multi-material or FGM components, as it possesses the capability to feed different power feedstock into the melt-pool. LDED is capable of fabricating both steep gradient and diffuse graded multi-material structures. LDED of multi-material components is carried out by feeding either a blended powder stream or streams of different powder at required flow rates into the melt-pool as described in Figure 2(c). In addition, it offers minimum wastage of blend or mixed powder as it uses a minimum quantity of feedstock as compared to LPBF[16][17]



(a)



(b)



(c)

Figure 1: Schematic of LAM (a) LPBF system (b) LDED system (c) FGM fabrication by LDED

The present article reviews the different combinations of LDED built metallic multi-material components fabricated along with the possible applications. In addition, the challenges associated with the fabrication of multi-materials using LDED are also discussed in detail. Further, the research work carried out on LDED of multi-material components using the indigenously developed LDED system at Raja Ramanna Centre for Advanced Technology (RRCAT), Indore, is also discussed.

2. LDED OF METALLIC MULTI-MATERIALS

2.1 Iron (Fe) and Nickel (Ni) based multi-material:

A combination of Fe and Ni-based multi-material components offer improved strength, high-temperature corrosion, and oxidation resistance. These multi-material components can be deployed in extreme environmental conditions such as nuclear fission, aerospace, and automotive applications [8][18]. Face Centered Cubic (FCC) structure and good mutual solubility of Fe and Ni favors the fabrication of Fe and Ni bi-metallic joint. However, the conventional joining process of Ni and SS suffers from cracking at the Heat Affected Zone (HAZ) due to the formation of brittle intermetallic phases (Chromium (Cr) precipitates and Niobium (Nb) rich phases) in the Ni region [8]. Thus, to avoid early failure of the engineering components, LDED technique is used to fabricate FGM of Fe and Ni-based alloy. Researchers have attempted wide combination of Fe and Ni-based alloys such as IN625-SS304 [18][19], IN625-SS316 [20][21][22], IN718-SS316[23][24][25], Rene88DT-SS316[26], INVAR-SS316 [8], etc. by LDED for various engineering applications.

LDED built Fe and Ni-based graded structures are built by gradually changing the Fe and Ni chemical composition as per the requirement [18][22]. However, localized elemental segregation of C, Cr, Nb, and Mo in the graded region causes the formation of brittle intermetallic phases[8][23].The formation of intermetallic phases in the interdendritic region leads to micro-cracks in the LDED built Fe-Ni based multi-material structures [22][26]. Thus, Thermo-Calc and CALPHAD are used to design the grading percentage for understanding the different possible phase transformations and simulating the solidification behavior to avoid the intermetallic compound formation during LDED[18]. One of the potential solutions to mitigate the formation of micro-cracks is adopting non-linear mixing and deployment of alloy powder with non-linear compositions of Nb and Mo [18]. Microstructural characterization shows the dominance of cellular-dendritic microstructure in the graded region of LDED built Fe and Ni-based multi-material structures. Tensile testing of LDED built Fe and Ni-based multi-material reveals that the strength of FGM bulk is higher than the strength of SS, but lower than the strength of respective Ni-based alloy. The evaluated ultimate strength is 539 MPa and 537 MPa for Direct and graded SS-IN625 LDED built bulk structures, respectively[20]. It was observed that LDED built graded structures yield ductile mode of failure in the SS zone during tensile testing as the strength of SS is lower than the strength of respective Ni-based alloy [20][22].

In summary, a significant amount of work is reported on the particular combination of Ni and Fe based multi-material LDED for various engineering applications. Even though Fe and Ni have the same crystal structure and decent solubility, fabricating FGM of Ni-based super-alloy is challenging due to the formation of brittle intermetallic phases. Thus, there is a need for intensive research on LDED of other combinations of Fe-Ni based alloys with composition-dependent process optimization to avoid the formation of brittle intermetallic phases.

2.2 Titanium (Ti) alloy based multi-material

Ti alloys offer higher strength to weight ratio, high-temperature strength, and excellent corrosion resistance. Thus, a decent number of research efforts have focused on the feasibility of LDED of FGM involving Ti alloys to increase its wear resistance, which aids to improve its applicability in thermal protection systems for aerospace launch vehicles and biomedical implants etc.[11]. A large number of investigations have been carried out to increase the hardness and wear resistance of Ti alloys by adding Titanium Carbide (TiC) using LDED [27][28] [29]. It was observed that an increase in hardness value is obtained by adding hard particles of TiC in the Ti alloy matrix by LDED. However, increased TiC content increases the chances for crack formations due to the large difference in thermal expansion coefficient and ductility between Ti and TiC[27]. To mitigate this issue, researchers have reported methodology for defect-free LDED of Ti-TiC graded bulk structures up to 95% volume of TiC in Ti matrix [29].

Another multi-material combination involving Ti is the Ni-Ti based joints, due to its extensive applications. One of the important applications of graded Ti alloy and Ni-based alloy is a thermal protection system for aerospace launch vehicles [30]. LDED of Ni-Tialloy is challenging due to the phase transformation, the large difference in the thermal expansion coefficient, and the inclusion of Al, Cr and Ti[31]. Complex phase transformation of Ni-Tialloy leads to the intermetallic formation (Ti₂Ni, TiNi, Ti₂Ni and TiNi₃) at different compositions of Ni and Ti during LDED [17][32]. Thus, selection of appropriate composition-dependent process window and grading percentage leads to crack-free graded bulk of Ni-Tialloy [32].

One of the important applications of Ti-based materials is in the biomedical sector. Titanium Oxide (TiO₂) is used for fabricating hip implants due to its bio-compatible nature [33]. Graded bulk structures of Ti and TiO₂ are fabricated using LDED by researchers and subjected to metallography and cell culture tests for qualification[33]. LDED built Ti-TiO₂ graded bulk structures at 50% grading yield desirable features such as enhanced wettability, cell-material interactions, and lubrication ability[34][35].

Ti-6Al-4V is one of the most explored Ti alloys for biomedical applications due to its higher strength to weight ratio, high-temperature strength and corrosion resistance and biocompatibility [36]. However, Al and V are not recommended for long term implants due to toxicity and associated health issues[37]. Therefore, researchers are working on overcoming the above limitations by deploying Ti-6Al-4V-Mo or Ti-Mo alloys [38][39][40].

2.3 Fe and Copper (Cu) based multi-material

The multi-material component of Fe and Cu based alloy is designed to take advantage of the higher thermal conductivity of Cu and improved strength of Fe in a single component. Cu-SS FGM is one of the emerging research fields for power generation, heat transfer, tooling, and cryogenic applications [41][42]. There are pieces of literature available on LDED of Fe based alloy (tool steel and stainless steel) and Cu for the tooling applications [43][44][45][46]. FGM of steel and Cu is intended to increase the cooling rate during the last stage of the molding cycle by improving the thermal conductivity of molds through Cu addition. However, LDED of Cu-SS is challenging due to the large difference in thermo-physical properties of Cu and SS leading to crack and porosity formation at the Cu-SS interface. Literature indicates that crack susceptibility of Cu-SS joint primarily depends on composition during LDED processing[43][44][46]. Lower Cu content leads to trapping of Cu in the form of thin-film leading to higher crack susceptibility during LDED. Thus, LDED of Cu-SS has higher crack susceptibility at a lower Cu percentage (<49%)[46]. This issue of crack formation can be resolved by adding a buffer layer of Ni-based alloy between Cu and SS [43]. However, there is no study on LDED process parameter optimization or composition-dependent investigation for defect-free deposition of Cu-SS multi-material structures.

2.4 Steel based multi-material

A dissimilar joint of ferritic and austenitic steel is one of the commonly used joints in the heat exchanger of fossil-fired and nuclear power plants[47][48][49]. A ferritic and austenitic steel joint is needed to connect the light water reactor side and pressurized steam side of power plants[50]. A large difference in Chromium and Carbon content in ferritic and austenitic steel leads to the migration of Carbon and Chromium causing cracking and early failure of the welded joint [50]. FGM is one of the methods to mitigate the issues associated with the sharp interface of ferritic and austenitic steel joints. Theoretical investigation using the finite element method and Thermo-Calc reveals that the difference in thermal expansion coefficient and carbon diffusion can be minimized by adopting 10% grading [51]. Further, LDED of various graded ferritic and austenitic steels is carried out using a combination of SS316 to 1085 steel [52], S316 alloy to magnetic ferritic SS430[53], 2.25Cr-1Mo steel to Alloy 800H[54], SS316 to P21 [55], etc. It is observed that smooth transition in the chemical composition helps to overcome the issue of carbon migration and prevent the early failure of multi-material components[56].

3. ISSUES ASSOCIATED WITH LDED OF MULTI-MATERIAL

The advances in LDED has widened its application and it is becoming a sustainable option for fabricating multi-material components. However, there are several issues associated with LDED of multi-material components that need to be addressed for the technical viability and wide acceptability of the technique. Some of the issues associated with LDED of multi-material are described below.

3.1 Lack of standard practices

The acceptability of LDED at the industrial level is delayed by the lack of standardization in terms of process and material qualification [57]. Similarly, there is a dearth of standard practices to understand the relation between process-structural-material properties during LDED multi-material processing. There is a lack of literature on selection criteria that defines grading or composition percentage and post-process treatment of LDED for different combinations of material. Thus, standardization is needed in terms of selection of material combination, multi-material process window, and material qualification as per the requirement for complete acceptability of the technique.

3.2 Metallurgical issues associated with multi-material combination

Multi-material fabrication using LDED is preferred to overcome the issues associated with a sharp interface at the dissimilar material joints. However, dissimilar material offers lower solubility causing the crack, porosity, and non-homogenous properties in LDED built multi-material. Fabrication of functionally graded components is one of the possible solutions to mitigate the above issues associated with dissimilar materials. However, even after adopting the grading approach, complete elimination of defects in LDED built part is challenging due to the large difference in thermo-physical properties, limited solubility, and the tendency of intermetallic phase formation. Formations of intermetallic phases lead to cracking due to their inability to accommodate the thermal strain generated during LDED processing. Thus, a complete metallurgical and thermodynamically compatible solution is needed to overcome the issue of poor solubility and intermetallic formation for LDED of different combinations of materials.

3.3 Limitations on material processing capabilities

LDED is successfully deployed for fabricating several multi-material combinations for various engineering applications. However, several material combinations are unexplored or not attempted due to metallurgical issues. There is a need for advancements in LDED to process material combinations having a large difference in melting point and thermo-physical properties such as Cu-SS, Ti-Ni, Al-SS, etc. It is observed that processing of difficult to process materials, like - Cu, Al, Ceramics, etc. requires special processing conditions (i.e., higher laser power, preheating, control atmosphere, controlled thermal cycle) and an improved feedback system for defect-free fabrication of multi-material engineering components.

3.4 Lack of multi-material modelling data

LDED is a complex process that involves energy transfer, mass transfer and phase transition. Thus, modelling of process and material transformation has become a significant tool to understand and control the LDED process. However, LDED modelling is challenging due to the simultaneous involvement of various transport phenomena and the unavailability of temperature-dependent thermo-physical properties for several alloy systems. Further, there is a lack of literature on modelling approaches at the non-equilibrium conditions to evaluate local composition distribution and elemental segregation in a melt pool at various processing conditions during LDED of multi-material.

4. LDED OF MULTI-MATERIAL STRUCTURES AT RRCAT

Laser Additive Manufacturing Laboratory (LAML) at RRCAT, Indore, has indigenously developed LPBF and LDED systems. Indigenously developed LDED system, equipped with 2 kW CW fibre laser and five-axis CNC manipulator, possesses a build volume capacity of 250 mm× 250 mm×250 mm (refer to Figure 3). More details about system specification and architecture are published in one of our previous works[13]. This section presents a brief overview of the LDED of multi-material components built at LAML, RRCAT.

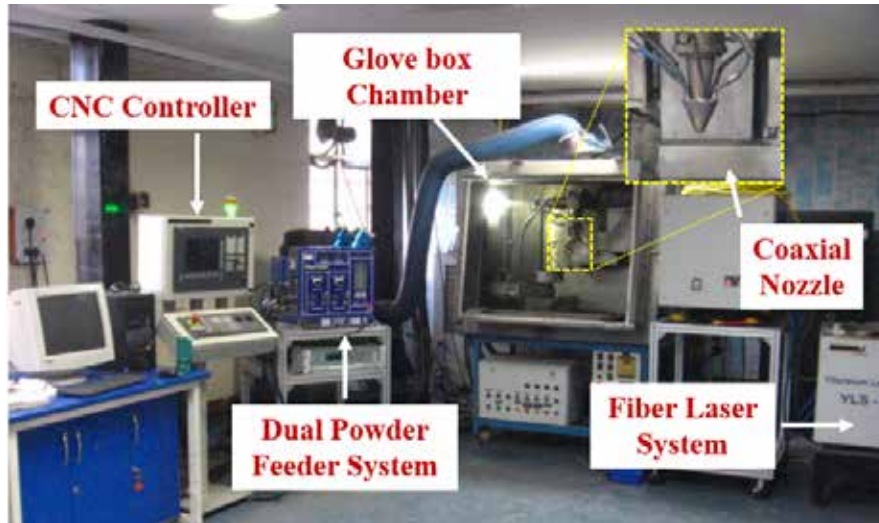


Figure 3. Indigenously developed LDED system at RRCAT

4.1 Functionally graded Ni-Cr-B-Si and SS316L bulk structure

LDED of functionally graded Ni-Cr-B-Si and SS316L bulk structures was carried out for nuclear and power generation applications due to improved corrosion and wear resistance at high temperatures. A parametric investigation was performed to evaluate the process window for fabricating the functionally graded bulk structure of Ni-Cr-B-Si and SS316L. The identified process parameters for LDED of functionally graded Ni-Cr-B-Si and SS316L bulk structures are laser power of 1000 W, scan speed of 0.5 m/min, and powder feed rate of 4 g/min. LDED of graded bulk was carried out at 25% compositional grading using the pre-alloyed or blended powder technique. It was observed that LDED built functionally graded bulk yields defect-free deposition at identified parameters [58].

4.2 Functionally graded Cu and Ni bulk structures

Cu-Ni FGM is designed to use the higher thermal conductivity of Cu and high strength and corrosion resistance of Ni in a single engineering component for marine applications. However, LDED of Cu-Ni is challenging due to the higher thermal conductivity and oxide formation tendency of Cu. In addition, Cu offers minimum absorption to infrared lasers leading to the unavailability of sufficient laser energy during LDED. A full factorial experimental design was used to identify the process window for depositing blended and virgin compositions of Cu_xNi_{100-x} , where x varies as 0,25,50,75, and100. LDED built Cu-Ni graded bulk at identified parameters yields relative density > 99.99% with pure Cu and Ni phase [59].

4.3 Bi-metallic structures of Cu and SS

Cu-SS joint is required for tooling, power generation, and heat transfer applications. However, LDED of Cu-SS is challenging due to the large difference in thermo-physical properties and poor solubility of Cu and Fe, which leads to cracking and porosity generation at the interface of Cu-SS. Thus, a detailed investigation was performed on the LDED of Cu bulk structures on SS304L substrate. LDED of Cu-SS joint was carried out using three different

inter-layer processing strategies in terms of Laser power per unit Feed (LEPF): Constant LEPF, Decreasing and increasing LEPF. It was observed that LDED built Cu-SS structures at increasing LEPF strategy yields defect-free bulk deposition of Cu with metallurgically sound Cu-SS interface (refer Figure 4)[15][60].

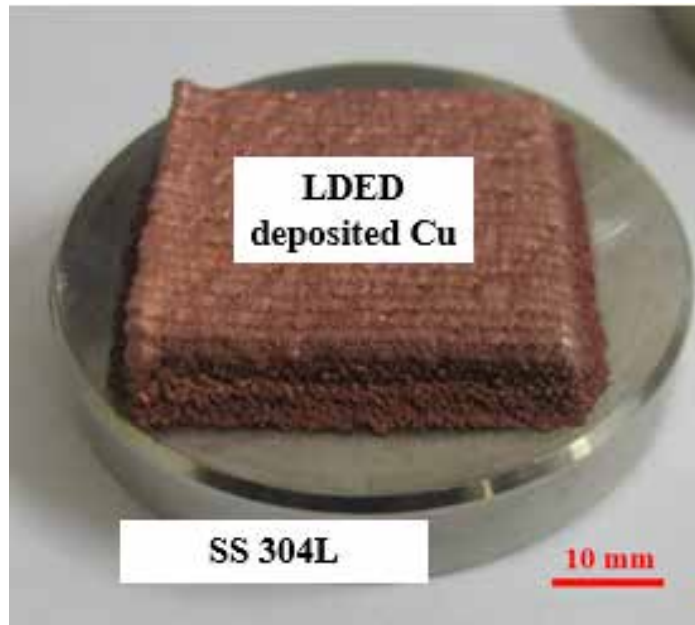


Figure 4: LDED of Cu on SS304L substrate

4.4 LDED of SiC on Zircaloy-4 tubes

Zircaloy-4 (Zry-4) is one of the commonly used fuel clad materials in nuclear power plants. It suffers from oxide formation and hydrogenation at operating conditions leading to brittle failure of the clad layer. One of the commonly used techniques to protect the fuel-clad layer is developing an “accident tolerant fuels” (ATF) clad layer of SiC on Zry-4. Thus, a detailed investigation is carried out by varying process parameters to deposit a 0.2mm thick coating of SiC on a 0.6mm thick Zry-4 tube. The process parameters were identified for achieving uniform thickness and controlled deposition of SiC on Zry-4 using LDED [61].

4.5 LDED of Molybdenum-CuCrZr

LDED of Molybdenum (Mo) is carried out on CuCrZr for International Thermonuclear Experimental Reactor (ITER) project to endure the component for power loads. LDED of Mo on Oxygen-Free Copper (OFC) is carried out by adopting three different approaches: Direct Mo deposition on Cu substrate without preheating, direct Mo deposition on Cu substrate with preheating, and Mo deposition on Cu substrate by using a buffer layer of Ni. It was observed that Mo deposition on Cu at preheat temperature of 250°C yields a significant reduction in interfacial porosity. A buffer layer of Ni between Mo and Cu yields defect-free and desired deposition quality at the identified parameters.

4.6 LDED of WC on SS

LDED of tungsten carbide (WC) is carried out on Stainless steel to avoid solid particle erosion (SPE) in steam and jet turbines, pipeline carrying slurry, valves, pipes etc. LDED of WC on SS was challenging due to the large difference in thermal expansion coefficient, which leads to the formation of crack, porous and brittle deposits. Thus, WC is mixed with Ni matrix at different weight percentages (5, 10 and 15 percentage) to prepare WC-Ni metal matrix composite (MMC). LDED built WC-Ni MMC on SS yields crack free and dense deposition at identified parameters[6]

5. SUMMARY

Multi-material processing using LDED is one of the emerging sectors and it offers a paradigm change in the technique of fabricating engineering components by offering material and design freedom. This review briefly describes the current state of the art and issues associated with the LDED of various combinations of multi-material metallic components. Though the advancements in LDED has boosted its applications in various sectors, there is a need for systematic research to qualify LDED for fabricating multi-material engineering components. Thus, improvements are required in terms of the powder quality control, process control, metallurgical compatibility of multi-material, and modelling of multi-material LDED process for its acceptance and viability as a manufacturing technique.

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[1] J. Singh, A. Kumar: Investigation of structural, morphological and electrochemical properties of mesoporous $\text{La}_2\text{CuCoO}_6$ rods fabricated by facile hydrothermal route. *Int. J. Miner. Metall. Mater.* 27: 987–995, 2020
 - ❖ **Book:**

[2] J.D. Verhoeven: Fundamentals of physical metallurgy, 1st Edition, New York: Wiley, New York, pp. 55-61, 1975
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[3] Cory Mitchell: Fractal Indicator Definition and Applications. Technical analysis basic education. Available via <https://www.investopedia.com/terms/f/fractal.asp>, Retrieved 3 August 2020
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 - ❖ **Input** : Time domain data X(t)
 - ❖ **Output** : P
 - Read the signal X(t)
 - Crop the signal and get the length N
 - Supply sampling frequency, f_s
 - Do fast Fourier transformation of the signal
$$X_k = \text{fft}(X(1:N), f_s);$$
 - Compute power spectral density and corresponding frequency by
$$P = X_k \cdot \text{conj}(X_k) / N;$$
$$f = f_s * (1:N) / N;$$