Effect of thermal cycles on the microstructure and mechanical properties of cold-resistant steel 09CrNi2MoCu during laser deposition

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Abstract. The formation of microstructure features of cold-resistant bainite-martensite steel 09CrNi2MoCu has been investigated. Thermal cycles during direct laser deposition were studied. The thermal cycles at different points of the deposited samples were investigated. The thermal cycles and CCT diagrams on microstructure formation and mechanical properties have been analyzed. The numerical calculation of the three-dimensional thermal conductivity problem by the finite element method is carried out. The received data of experimentally measured thermal cycles and the calculated data have shown good coincidence of temperature values. On the basis of the obtained data the calculated dependence of inter-layer temperature at depositing the sample with and without a pause is given. The microstructure and mechanical properties of the samples in the initial state and after heat treatment have been studied and compared with traditional hot rolling. The microstructure features at different pauses between passes in different parts of the obtained samples were revealed. The effect on static tensile and impact toughness at -40\textdegree C in the bred and heat-treated state was investigated.

1. Introduction

Additive technologies (AT), including direct laser deposition (DLD) technology, are used to improve the competitiveness of shipyards and the production of parts for shipbuilding industry [1-4]. DLD is characterized by the process of repeated exposure of a laser source to a melt bath with high heating and cooling rates varying in the process of deposition and depending on the size and geometrical parameters of the manufactured part. As a consequence, the obtained microstructure of iron-based alloys during deposition is heterogeneous and anisotropic, which considerably affects the performance characteristics of the finished part. The resulting parts are not inferior in properties to traditional methods of production, whose properties can resist critical loads under extreme conditions [5, 6].

High flexibility of spatial and temporal parameters of laser impact provides an opportunity to obtain a given structure and material properties with a controllable gradient of properties. In contrast to traditional technological processes based on blanks processing. Laser technology combines different
In the DLD process, it is possible to control the structure and mechanical properties by the selection of localized solidification parameters. The space-time parameters of solidification can be estimated from the localized thermal behavior, which can be manipulated by changing the processing parameters, pause to obtain parts with the expected mechanical properties. The analysis and calculation of temperature fields in DLD is of the greatest interest in the global scientific community. At which the formation of operational properties of the final part takes place.

In this paper the influence of technological parameters of laser radiation on the anisotropy of cold-resistant steel 09CrNi2MoCu is considered. Thermal cycles in the deposition process with different pauses between passes are considered. A CCT diagram of the deposited samples using a high-temperature and high-speed quenching dilatometer is constructed. The effect of heat treatment of the obtained samples on the mechanical properties is considered. The microstructure features on the performance properties of the obtained material are investigated. The calculation of temperature fields by the finite element method and the comparison of heating temperatures at different pauses have been carried out. The obtained results are of scientific and practical value for further study of processes occurring in cold-resistant steels under the influence of concentrated energy flows.

2. Materials and Methods

The base material is powder alloy 09CrNi2MoCu produced by «SphereM», bainite-martensite class, the chemical composition of the alloy TU5.961-11571-2006 (RU) and X-ray spectral analysis of the powder are presented in Table 1.

| Steel grade | light element | Cr | Ni | Cu | Mo | Si | Mn | P  | Fe |
|-------------|---------------|----|----|----|----|----|----|----|----|
| TU (RU)     | 0.08-0.11     | 0.01 | ≤0.02 | 0.30-0.70 | 1.80-2.20 | 0.4-0.7 | 0.35 | 0.17-0.37 | 0.3-0.6 | ≤0.015 |
| Powder      | 0.09-0.004    | 0.026 | 0.005 | 0.5 | 1.93 | 0.47 | 0.28 | 0.25 | 0.39 | ≤0.01 |

Samples were deposited using the SPbSMTU experimental unit for direct laser deposition. The complex is equipped with: industrial robot LRM-20iB/25, fiber laser LS-3 Yb, laser sputtering head FLW D30, nozzle SO12, powder feeder. A sealed chamber filled with argon was used for deposition. In the argon-filled chamber, the oxygen content did not exceed 1000 ppm. The samples were produced by the DLD method at power P=2600 W, speed V=25 mm/s, powder flow G=22 g/min as a conveying gas, Δx=1.67 mm, and Δz=0.8 mm were used. A sample of size LxWxH 130 mm x 90 mm x 14 mm was obtained for the experiment figure 1.
Figure 1. Schematic of thermal cycle measurements in the DLD process.

To analyze thermal cycles during deposition, measurements were made at the bottom (point 1) and at the top (point 2) of the sample Figure 1. For the measurements, a K-type chromel-alumel thermocouple with a wire diameter of 1.2 mm was used to measure thermal cycles. The period of temperature measurement by the thermocouple was 20 ms. The thermocouple was mounted on the samples without a pause and with a pause between passes of 30 s.

The CCT diagrams were plotted using a DIL 805A/D high-speed quench-deformation dilatometer and a DIL-402C high-temperature dilatometer. Experiment scheme - heating to a temperature of 900ºC, exposure for 5 min. For experiments with speeds up to 0.1 C/s we used a DIL-402C dilatometer, for speeds above 0.1 C/s we used a DIL 805A/D dilatometer. The positions of critical points of phase transitions during cooling were determined by the tangential method when deviating from the monotonic course of the thermal expansion curve.

Thermal processing of deposited samples was performed in a muffle furnace SNOL 30/1300. Thermal treatment mode: homogenization T=1100°C exposure 6 hours; hardening at T=920°C exposure 3 hours and subsequent cooling in oil; high tempering at T=650°C exposure 5 hours and cooling in air.

To study the microstructure, chemical etching of the Klemm’s (50 ml of concentrated Na$_2$S$_2$O$_3$ solution + 1 g Na$_2$S$_2$O$_5$) was carried out. The deposited samples were examined by optical microscopy on a Leica DMi8 optical microscope using «Axalit» software.

The deposited samples were tensile tested on Zwick Roell Z100 at 20°C, the impact toughness tests were performed on Zwick Roell RKP450 at -40°C. Microhardness was measured on a microhardness meter FM-310, using automated software «Thixomet Pro», with a load of 300 g according to Vickers (HV).

3. Results and discussion

3.1. Thermal cycles of the deposited samples

Figure 2 shows the thermal cycles for measuring the heating temperature of the deposited samples at a given point.
Figure 2. Thermal cycles: (a, c) deposited without a pause; (b, d) deposited with a pause of 30 s.

In DLD there are much more thermal cycles and more energy input than in traditional technologies: welding, rolling, etc. Transitions in additive manufacturing through critical points have 3-4 cycles figure 2, and the rest proceed below the point Ac3. Such temperature cycles significantly affect the formation of the microstructure and, consequently, the mechanical properties. During repeated local temperatures, overlapping thermal cycles occur in some areas, which leads to changes in the structure and properties of the previous layers from repeated heating. The most brittle region is the area of coarse grains undergoing repeated heating between the critical points Ac3-Ac1 [9-11]. The longest effects in such areas are the samples that are obtained with no pause figure 2 (a, c).

In the deposited samples without pause, the temperature in the lower part of the plate is ~500°C figure 2 (c) and in the upper part ~600°C figure 2 (a) in the high-temperature transformation zone. During the sample deposition, the lower part is partially cooled and only at a considerable distance from the deposition region is cooling.

During the deposition process with a pause of 30 sec the temperature in the lower part of the plate is from ~200°C figure 2 (d) and in the upper part from ~300°C figure 2 (b) in the low-temperature transformation zone.

Figure 3 shows the CCT diagram of 09CrNi2MoCu steel after deposition and in the traditional state of rolling.
The behavior of the transformation curves strongly depends on the thermal prehistory, each temperature cycle corresponds to its own transformation. Therefore, the description of austenite decomposition under extreme temperature gradients should be carried out taking into account the kinetics of phase transformations of the CCT diagram.

The transformation of austenite for the initial steel obtained by conventional hot rolling method showed that from 100 to 15 °C/s formed bainite-martensite structure, from 15 to 2.7 °C/s formed bainite structure, which indicates the formation of a more homogeneous bainite structure. The analysis of the constructed CCT diagram of DLD samples showed that in the samples at cooling rates from 40 to 100 °C/s mainly bainite-martensitic structures are formed, from 5 to 40 °C/s bainite, from 5 to 0.1 °C/s bainite-ferritic. The diagram shifts to the left, indicating a nonequilibrium structure.

3.2. Microstructural characteristics

The microstructure after etching with the Klemm’s of samples figure 4 (a-f) represents the upper bainite (blue color), lower bainite (brown color), ferrite (light gray color), martensite (dark brown color), uncolored residual austenite inclusions (white color) [12]. figure 4 (a) shows the upper point of the sample grown without a pause and figure 4 (c) the lower part. Figure 4 (b) shows the upper point of the sample deposited with a pause of 30 s and figure 4 (d) the lower part of the sample.
It has been established that during the deposition of 09CrNi2MoD steel along with technological parameters the process of structure formation and the temperature field significantly influences mechanical properties. High mechanical properties are formed depending on the thermal cycles and the formation of structural components in the process of DLD. The mechanism of formation of microstructure heterogeneity along the height of the sample can be traced by analyzing the thermal cycles of Figure 2 (a-d) during deposition without a pause and with a pause between passes of 30 seconds.

During deposition without a pause figure 4 (a, c) a microstructure is formed of granular bainite grains and residual austenite. Residual austenite is formed because of high-temperature formation of the structure during deposition. The upper part of figure 4 (a) shows directional crystallization because of intense cooling after DLD. In the lower part of the figure 4 (c) sample, an ordered structure is formed with auto-heating and heat transfer from the upper layers.

At a pause of 30 s figure 4 (b, d) a bainite ferrite of batch type is formed with a structure of parallel plates with a small fraction of martensite. In the upper part of figure 4 (b), directional crystallization is observed because of intense cooling after the completion of DLD. In the lower part of figure 4 (d) an ordered structure is formed, which is formed with auto-heating and more prolonged cooling in comparison with the upper samples.

After heat treatment, the microstructure of figure 4 (e) structure consists of irregularly shaped granular bainite grains with evenly distributed grains throughout the volume [13]. The hot-rolled structure consists of granular bainite, with a banded structure, and residual austenite.

*Figure 4. Microstructure of bainite-martensite steel 09CrNi2MoCu: (a) DLD without pause; (b) DLD pause 30 sec; (c) DLD+HT; (d) hot-rolled steel.*
3.3. Results of numerical simulation of the temperature field at DLD

During the deposition process, a small volume of the manufactured DLD specimens is heated to high temperatures in a concentrated and short period of time. As the heat source moves, more and more volumes of metal are heated, and in the previously heated areas, the temperature evens out. Thus an uneven temperature field is formed in the deposited part with a large temperature gradient in the area of local heating. When making samples for the analysis of mechanical properties obtained by DLD, the question often arises - how the parameters of heat deposition and the presence of a time pause between layers affects the initial temperature during cladding of each layer. To solve this problem, a simplified model for determining the temperature field at DLD has been developed. The three-dimensional thermal conductivity problem was solved by the finite element method. The displacement of the heat source during surfacing was not considered. The heat input from all passes of the considered layer was summarized and set as a uniformly distributed source on the elements advising the volume of the considered layer. The computational model makes it possible to quickly predict the temperature change in the sample volume now of cladding each of the layers. It should be noted that with this formulation of the problem, the interpass temperature at the points corresponding to the cladding layer and the HAZ would be lower than that observed in the real experiment.

However, the comparison of the experimentally measured thermal cycles with the calculated ones showed a good agreement of the temperature values corresponding to the beginning of the surfacing of each of the layers. As noted above, the assumptions of the mathematical model allow us to determine the average temperature of the surfacing layer. The model does not accurately determine the effect of each of the passes on the temperature field, which can be clearly seen in Figure 5.

![Figure 5](image_url)
The computational model allowed us to determine the temperature of the sample during cladding each of the layers figure 6. It was seen that when cladding without a pause, the temperature already after the 10th layer is significantly higher than when cladding with a pause of 30s. The absence of a pause leads to the formation of a region in the sample, in which the inter-layer temperature during surfacing of each layer gradually increases. The highest rate of inter-layer temperature rise is observed near the substrate. After layer 40, the temperature reaches 650°C and then practically does not change. The non-uniformity of the inter-layer temperature should lead to the formation of a gradient of microstructure and mechanical properties along the height of the sample. The presence of a pause leads to the fact that the maximum inter-layer temperature is reached already at the 10th layer and then changes little.

![Figure 6. Calculated inter-layer temperature dependence when cladding the sample with a pause of 30s (blue line) and without a pause (red line).](image)

### 3.4. Mechanical characteristics

Table 2 shows the mechanical properties of the samples obtained with and without the pause, as well as with the subsequent heat treatment.

| No. | Direction of specimens | Ultimate strength (MPa) | Yield strength (MPa) | Relative elongation (%) | Impact toughness, (J/cm²) | Microhardness, HV₀.3 |
|-----|------------------------|-------------------------|---------------------|------------------------|--------------------------|----------------------|
| 1. TU | N/A | 690 | 512 | 25.4 | 90-100 | 200-210 |
| 2. DLD no pause | X | 563 | 356 | 27 | 64 | 193 |
| 3. DLD pause 30 s | X | 762 | 732 | 20.9 | 156 | 263 |
| 4. DLD +HT | Z | 665 | 598 | 21 | 120 | 226 |

The mechanical properties of the deposited specimens without pause are characterized by high values of relative elongation and low values of yield strength and ultimate strength. In the samples
obtained without a pause at high-temperature residence, the maximum decrease in impact toughness occurs in the coarse grain section of the HAZ. The main reason for such a decrease in toughness is the intensive growth of former austenite grains during high-temperature heating. For short-term process, heating, as a rule, is characterized by a high rate of heating of the metal and a small time exposure at maximum temperatures, which is not enough for complete homogenization of austenite, so the resulting structural components are enriched with carbon. The impact toughness has low values along with anisotropy of properties in various directions.

High yield strength and tensile strength values were obtained with a pause between passes. The relative elongation in the transverse (Z) direction is 27% lower than in the longitudinal (X) direction. This indicates anisotropy of properties which is inherent in some alloys after AT manufacturing. The impact toughness is higher than that of conventional rolled parts.

After heat treatment, uniform mechanical property values in the longitudinal and transverse directions are observed, which indicates the homogeneity of the structure.

4. Conclusions

It has been established that in the process of deposition from bainite-martensite steel 09CrNi2MoCu along with technological parameters the process of structure formation and mechanical properties is significantly affected by temperature fields. High mechanical properties are formed depending on thermal cycles and the formation of structural components in the process of DLD. The analysis of thermal cycles and thermal history, which directly affect the performance properties of manufactured AT parts, is of the greatest attention. In this work, for the first time, mechanical characteristics were measured at different pauses and thermal cycles were measured during deposition in cold-resistant 09CrNi2MoCu steel with a martensitic-bainitic structure.

In deposition with pauses between passes, this mode is the closest to the production of a real part. However, there are areas where material filling is continuous, so there will be a difference in mechanical properties in some areas, as well as in the height of the part. In this regard, it is necessary to carry out a complete heat treatment of such parts, or to simulate the manufacturing process with a uniformly distributed thermal field to achieve high properties throughout the part.

In continuous deposition without pauses, residual austenite is formed and grain growth takes place, which negatively affects the impact toughness at negative temperatures. In order to increase the level of mechanical properties and to eliminate anisotropy in the whole volume of the obtained material it is necessary to carry out a complete heat treatment.

The DLD process is a promising technology that is not inferior to traditional methods of production, in particular hot rolling, reducing the intermediate technological cycles, increasing the utilization factor of the material. The obtained mechanical properties are at the level of rolled and cast, and sometimes much higher, which indicates the great perspective of additive technologies.

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