Investigation on the cold rolling and structuring of cold sprayed copper-coated steel sheets

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Abstract. A current driving force of research is lightweight design. One of the approaches to reduce the weight of a component without causing an overall stiffness decrease is the use of multi-material components. One of the main challenges of this approach is the low bonding strength between different materials. Focusing on steel-aluminum multi-material components, thermally sprayed copper coatings can come into use as a bonding agent between steel sheets and high pressure die cast aluminum to improve the bonding strength. This paper presents a combination of cold gas spraying of copper coatings and their subsequent structuring by rolling as surface pretreatment method of the steel inserts. Therefore, flat rolling experiments are performed with samples in “as sprayed” and heat treated conditions to determine the influence of the rolling process on the bond strength and the formability of the coating. Furthermore, the influence of the rolling on the roughness and the hardness of the coating was examined. In the next step, the coated surface was structured, to create a surface topology suited for a form closure connection in a subsequent high-pressure die casting process. No cracks were observed after the cold rolling process with a thickness reduction of up to $\varepsilon = 14\%$ for heat treated samples. Structuring of heat treated samples could be realized without delamination and cracking.

1. Introduction

A current research project at the RWTH Aachen deals with the joining of molten aluminum and a steel sheet by high-pressure die casting. Due to the challenge that no sufficient metallurgical bonding between steel and aluminum occurs, thermal spraying processes shall be used as a pretreatment of steel sheets to overcome the issues occurring during the joining of steel and aluminum. Thermally sprayed coatings can come into use to reduce the formation of brittle intermetallic phases and to combine the advantages of metallurgical bonding with mechanical interlocking. In order to achieve higher bond strength, the thermally sprayed coating shall be structured. This requires a high formability of the coating.

Cold spraying as no other thermal spraying process allows the deposition of coatings with properties similar to those of bulk materials. As the particles of the deposited material remain solid during the spraying process, they are less susceptible to oxidation [1]. Coatings of ductile materials such as copper or aluminum applied by cold gas spraying exhibit a low porosity and oxide content. However, cold gas sprayed coatings have low ductility due to a strong work hardening effect occurring during deposition and are therefore difficult to deform plastically. Recently some studies have investigated the mechanical behavior of cold sprayed coatings indicating the possibility to improve the microstructure and mechanical properties by means of heat treatment. Renzhong et al. have
investigated the mechanical properties of detached cold sprayed copper coatings under tensile loading in as sprayed condition and after heat treatment at different temperatures [2]. It was shown that the elongation of the copper coatings can be increased from $A = 0.5\%$ up to $A = 35\%$ after heat treatment at $T = 500\, ^\circ\text{C}$ for $t = 5\, \text{h}$ at the expense of the tensile strength which decreased from $\sigma_M = 290\, \text{N/mm}^2$ to $\sigma_M = 240\, \text{N/mm}^2$. Gärtner et al. compared in their study mechanical properties of cold sprayed, arc sprayed and high velocity oxygen fuel (HVOF) sprayed coatings in as sprayed and annealed conditions [3]. Taking hardness as an indicator for work hardening, the amount of deformation for cold sprayed copper corresponded to that of cold rolling of bulk material with thickness reduction of about $\varepsilon = 90\%$. It was also shown that the formability of cold sprayed coatings can be significantly improved by annealing. Despite the interest in mechanical performance of thermal sprayed coatings, the formability of the coated components by means of cold rolling has not yet been investigated.

Copper coatings often serve as bonding agent for multi-material components. During high pressure casting of aluminum onto steel sheets, copper interlayers can be used as a barrier to prevent the formation of brittle iron aluminides. The binary system of copper and aluminum has a low melting eutectic with a melting temperature of $T_m = 548.2\, ^\circ\text{C}$ implying the possibility of a better metallurgical bonding between these materials in comparison to that between steel and aluminum. Copper possesses a further benefit, as it has a coefficient of thermal expansion in the range between steel and aluminum and thus can reduce the thermal stresses occurring during the solidification process after high pressure die casting. In addition, the bond strength can be optimized by a form closure connection. This can be achieved by forming the coated steel sheet to increase the contact area and to create a suitable surface structure for mechanical interlocking between aluminum cast and steel inserts.

Surface structuring by rolling is an established research area at the Institute of Metal Forming (IBF) of RWTH Aachen University. Some work in the past concentrated on the rolling of channel-like grooves into steel sheets made of steel DC04 using a modular roll-setup. Grooves with a height of up to $h = 0.5\, \text{mm}$ and aspect ratios, thus the ratio of structures height to structures width, between 0.5 and 1.0 could be realized using different roll setups. Figure 1a) shows a top view of such a structure and the profile measured with a digital microscope. For all investigated structures, the same trend could be determined regarding the resulting size of the structure $h_s$ in dependence of the thickness reduction. For the structuring of steel DC04 by solid roll with a structure height of a roll $h_{s,\text{roll}} = 0.5\, \text{mm}$ it was shown that the thickness reduction of $\varepsilon = 28\%$ was required, to achieve a relative size of 1 ($h_{s,\text{roll}} = h_s$), Figure 1b) [4].

**Figure 1.** a) channel-like structure in steel sheet and b) the resulting size of the structure in dependence of the thickness reduction [4].

In the current study the process limitations of cold rolling and surface structuring of cold sprayed copper coated steel sheets are investigated. In order to increase the formability of the copper coatings a heat treatment is performed prior to cold rolling. Moreover, the influence of mechanical loads in a range comparable to those occurring during roll bonding on the bond strength of the coating is investigated. For steel sheets, a thickness reduction of at least $\varepsilon = 50\%$ is needed to realize an
adhesive bond by roll bonding [5], as the oxide layers at the metal surface can only be broken by a high degree of deformation. For coated components, the adhesion between coating and substrate is achieved through the high velocity impact of the particles during the deposition process but an additional influence of the rolling process on the bond strength can be expected.

2. Experimental Setup
The experimental procedure includes cold spraying process, heat treatment, cold rolling and sample analysis. The principle scheme of the experimental setup is shown in Figure 2.

2.1 Cold Gas Spraying
In the present study, the conventional steel DC04 was selected as a substrate material. Prior to spraying process, the steel plates were grit blasted using alumina with the grit size of F20 and cut to rectangular plates with the dimensions of V = 200 × 30 × 2 mm³. For the deposition, a 99 % pure copper powder with a size distribution of -45 +15 µm, delivered by ECKA Granules GmbH (Fürth, Germany), was employed. The spray parameters are shown in Table 1. The deposition of the coatings was performed at Surface Engineering Institute of RWTH Aachen University using the Kinetics 8000 Cold spray system from Oerlikon Metco AG (Switzerland) with nitrogen as process and carrier gas.

| Parameter                | Value       |
|--------------------------|-------------|
| Gas temperature [°C]     | 500         |
| Process gas flow [m³/h]  | 66          |
| Carrier gas flow [m³/h]  | 2.4         |
| Spray distance [mm]      | 30          |
| Transverse movement velocity [mm/s] | 300         |

In order to improve the formability of the sprayed coating, some samples were heat treated in vacuum at a temperature of T = 500 °C for t = 5 h. The choice of these parameters is based on the analysis of the results presented in the study of Renzhong et al. [2]. The cold sprayed samples were heat treated using a PVA MOV 553 vacuum furnace. The non-heat-treated samples were also cold rolled as a reference for the investigation of the effect of annealing.

2.2 Cold Rolling Process
All rolling experiments were performed with a rolling mill at IBF. The two principles for flat rolling and rolling of a channel like structure are shown in Figure 3. In order to avoid difficulties regarding the insufficient stiffness of the tooling in case of a modular roll, observed by Senge et al. in [4], a solid
roll with a higher stiffness was used for these experiments. During the experiments, the force and the global thickness of the compound, \( h_1 \) for flat rolling and \( h_{c\text{-peak}} \) for structure rolling, are measured. The dimensions of the structure such as the sheet thickness measured in the valley of a channel \( h_{c\text{-val}} \) and the depth of a channel \( h_{c\text{-depth}} \), are determined in metallographic cross sections, Figure 3.

![Figure 3. Principle scheme of the two rolling procedures, where \( h_{c\text{-peak}} \) is the thickness in the peak of the structure, \( h_{c\text{-val}} \) is the thickness in the valley of the structure.](image)

The global thickness reduction performed in these experiments was about \( \varepsilon = 8\% \) in one rolling pass and about \( \varepsilon = 14\% \) in two subsequent passes. Table 2 gives an overview of the cold rolled and structured samples from both processing routes with and without heat treatment step. The thickness reduction of the structured samples is defined as the difference between initial thickness \( h_0 \) and final thickness measured in the valley of the structure \( h_{c\text{-val}} \).

**Table 2.** Overview of the performed experiments and the thickness of the coated steel sheets (CR - cold rolled, HT - heat treated, S - structured).

| Sample     | initial thickness, \( h_0 \) [mm] | final thickness, \( h_1 \) [mm] | thickness reduction, \( \varepsilon \) [%] | heat treatment | rolling process |
|------------|----------------------------------|---------------------------------|------------------------------------------|----------------|----------------|
| CR8        | 2.07                             | 1.91                            | 8.0                                      | no             | flat           |
| CR14       | 2.07                             | 1.78                            | 14.2                                     | no             | flat           |
| HT+CR8     | 2.07                             | 1.89                            | 8.5                                      | yes            | flat           |
| HT+CR14    | 2.08                             | 1.78                            | 14.3                                     | yes            | flat           |
| HT+S       | 2.26                             | 2.26                            | 2.06                                     | 0.0            | 8.7            | yes            | channel |

### 2.3 Testing

In order to characterize the coating, the cold sprayed samples were metallographically prepared and examined in terms of their microstructure. The coating thickness was measured on the cross sections using a light microscope by Carl Zeiss with Axio Vision image analysis tool. The roughness of the surfaces in as sprayed and cold rolled condition was measured by a confocal laser microscope (VKX 210, Keyence, Osaka, Japan). The acquired topography images were used to evaluate the roughness parameters according to the evaluation procedure defined in DIN EN ISO 4288:1997 [6]. Three measurements were carried out on each sample at different random positions. The bonding strength of the deposited coatings was determined using the PATHandy tester (PATHandy, DFD Instruments, Kristiansand, Norway). The measurement principle is described in [8]. Even though this method is primarily standardized for lacquer coatings according to DIN EN ISO 4624 [7], it is industrially...
deployed for thermally sprayed coatings due to its simple handling and robustness. As it was shown by Schlaefer [9], the measurements by PATHandy are in good agreement with the adhesion tests conducted according to DIN 582 [10]. A microhardness tester Fisherscope HM2000 by Helmut Fischer GmbH (Sindelfingen-Maichingen, Germany) and Bühler Micromet 1 by ITW Test & Measurement GmbH (Esslingen am Neckar, Germany) were used for the examination of the Martens and Vickers hardness of the coatings, respectively. In order to identify a possible formation of cracks in the coating during the cold rolling process, a SEM analysis was performed by means of the scanning electron microscope Zeiss Leo 1530.

3. Results

The cold spraying experiments revealed that no adhesion of copper particles to the untreated steel surface could be achieved in the process temperature range of $T = 450 \, ^\circ\mathrm{C}$ to $T = 500 \, ^\circ\mathrm{C}$, as the particles bounced off the substrate. After grit blasting the steel surface with alumina F20, a coating with a thickness of about $s = 120 \, \mu\mathrm{m}$ could be deposited at a temperature of $T = 500 \, ^\circ\mathrm{C}$, yielding a bonding strength of $13.3 \pm 0.9 \, \mathrm{MPa}$. A further increase of the process temperature did not improve the coating properties significantly and led to particles clogging on the nozzle. The process parameters shown in Table 1 were adopted for further investigations.

3.1 Microstructure

The porosity of the copper coatings after deposition was about 0.5 %, Figure 4a). Copper feedstock exhibits a good plasticity resulting in a tight bonding between particles. The deposition quality was uniform through the whole thickness. Only the upper layer of the coating exhibits a lower density resulting in higher porosity close to the surface. This phenomenon is well known in cold spraying as a tamping effect, describing the process of additional deformation of deposited particles by impacting ones [11].

![Figure 4](image)

**Figure 4.** Cross section in plane of normal direction (ND)/rolling direction (RD) of the a) sprayed, b) cold rolled with $\varepsilon = 8 \, \%$, c) cold rolled with $\varepsilon = 14 \, \%$, d) annealed, e) annealed and cold rolled with $\varepsilon = 8 \, \%$, f) annealed and cold rolled $\varepsilon = 14 \, \%$ copper coated samples.

When the coated samples were flat rolled without previous annealing, some cracks were observed in normal direction/rolling direction (ND/RD) cross sections. After the first rolling pass, the adhesion of the copper coatings was still maintained. With a higher thickness reduction of $\varepsilon = 14 \, \%$, cracks propagation through the whole coating thickness towards the substrate surface and the coating
delamination occurred at different positions, Figure 4c). The SEM images show that the crack propagation occurs mainly at the particles boundaries, Figure 5.

![SEM images showing crack propagation](image)

**Figure 5.** SEM/RBSD images of the coatings cross section, showing the crack propagation after the flat rolling process with a thickness reduction of $\varepsilon = 8\%$ without annealing.

After annealing, the interfaces between the particles almost disappear. The EDS analysis showed that mutual diffusion of copper from the coating into the steel and iron from the steel into the copper coating occurs at the interface coating/steel during the annealing. The EDS measurement revealed at point P1 within the steel substrate the presence of copper in an amount of about 1.4 wt.%, and at the measurement point P2 within the copper coating the presence of about 1.8 wt.% iron, Figure 6. No cracks were observed in the annealed coating subsequent to flat rolling with a thickness reduction of $\varepsilon = 8\%$ and $\varepsilon = 14\%$, Figure 4f).

![EDS analysis and iron and copper content](image)

**Figure 6.** EDS analysis and iron and copper content at the interface steel/coating after annealing.

In the next step, the annealed samples were successfully structured by channel rolling without delamination or cracking. The mean structure height was about $h_{\text{c-depth}} = 120 \, \mu\text{m}$ at a thickness reduction of nearly $\varepsilon_{\text{val}} = 9\%$. Cross section of the structure is shown in Figure 7. For the structuring of bare DC04 sheet the same thickness reduction of about $\varepsilon = 9\%$ resulted in a structure height of nearly $h_{\text{c-depth}} = 170 \, \mu\text{m} [4]$. 
3.2 Surface roughness

The surface roughness after deposition and after each flat rolling pass was determined to observe the evolution of surface topography, Figure 8. The surface roughness is of interest as it can affect the wetting behavior of the aluminum melt during the high pressure die casting. Directly after deposition, the mean surface roughness was $Ra = 13.9 \pm 0.3 \mu m$. After subsequent flat rolling with the total cross section reduction ratio of $\varepsilon = 8 \%$ and $\varepsilon = 14 \%$, the roughness decreased to $Ra = 3.7 \pm 0.3 \mu m$ and $Ra = 1.9 \pm 0.1 \mu m$, respectively.

![Surface topography](image)

Figure 8. Surface topography of copper coatings after a) deposition, b) flat rolling with cross section reduction of $\varepsilon = 8 \%$, c) flat rolling with cross section reduction of $\varepsilon = 14 \%$.

The same trend can be seen for the surface roughness depth $R_z$ which decreased from $R_z = 100.8 \pm 4.2 \mu m$ in the as sprayed condition to $R_z = 44.1 \pm 3.2 \mu m$ after the first cold rolling pass and to $R_z = 26.1 \pm 1.0 \mu m$ after the second pass. No significant difference was observed when comparing non-treated and heat treated samples, Figure 9.
3.3 Hardness of the coating

The cold spraying as well as the cold rolling process causes a work hardening effect and a respective reduction of formability of the coatings. The hardness measurement provides a simple way to estimate the extent of strengthening after cold spraying and cold rolling. In Figure 10a), the evolution of the Vickers hardness of the copper coating after each process step is depicted. After the coating deposition, the hardness was about 140 HV0.05. The subsequent flat rolling does not contribute to the further work hardening of the copper coating. This can be related to the high degree of the particle deformation during the spraying process where they possibly reached their strain limit. This assumption is in accordance with the coating behavior observed metallographically, indicating the crack propagation already after the flat rolling with a cross section reduction of ε = 8 %. After annealing of the cold sprayed samples, the hardness of the copper coatings recovers to the level comparable with that of bulk copper of about 70 HV0.05. A subsequent cold rolling process leads to an increase of the hardness up to 100 HV0.05. The initial hardness of the copper powder couldn’t be determined by Vickers hardness measurement, as the particles were too small for indentation. To observe the evolution of the mechanical properties from initial state through cold spraying process and subsequent annealing, the Martens hardness were measured by means of Fisherscope. As can be seen in Figure 10b), the hardness after the heat treatment has almost reached the level of the bulk material. These results indicate the near-complete recovery of mechanical properties after annealing.

**Figure 9.** The mean surface roughness Ra and surface roughness depth Rz of the copper coating after deposition and subsequent heat treatment and flat rolling with ε = 8 %, and ε = 14 %.
Figure 10. a) Vickers hardness of the copper coatings after deposition, heat treatment and flat rolling and b) Martens hardness of the copper powder, after deposition and subsequent annealing.

For the pure copper coating, the strain hardening is the only mechanism contributing to the hardness increase as the other common mechanisms like precipitation hardening and solid solution hardening can be excluded due to the absence of alloying elements. For this reason, the hardness evolution directly indicates the deformed state of the microstructure. The comparison of the Martens hardness of the copper powder with the hardness in the as sprayed and annealed condition shows that the heat treatment at a temperature of $T = 500 \, ^\circ\text{C}$ for $t = 5 \, \text{h}$ is sufficient for reaching almost complete recovery of the mechanical properties.

3.4 Adhesion strength
To determine the influence of the annealing and the cold rolling processes on the adhesion of the coating to the steel substrate, the bond strengths were measured after corresponding process. Recently, some studies have pointed out, that a heat treatment can improve the bonding strength of the coating. Li et al. have observed an almost 100 % increase in the adhesive strength of a cold sprayed titanium coating after annealing at $T = 850 \, ^\circ\text{C}$ for $t = 4 \, \text{h}$ [12]. Although the EDS measurement revealed mutual diffusion processes at the interface Cu coating/steel during the annealing, no significant changes of the bonding strength were observed in the current study after the heat treatment process compared to the as sprayed condition, Figure 11. The flat rolling process without heat pretreatment influenced the adhesion of the copper coatings negatively. After the first rolling pass with thickness reduction of about $\varepsilon = 8 \, \%$, the bond strength decreased slightly. However, a number of cracks was observed by light microscopy in the normal direction. After the second rolling pass with the thickness reduction of about $\varepsilon = 14 \, \%$, the bonding strength decreased by approx. 50 %. For annealed samples, the flat rolling process did not lead to deterioration of the adhesive strength even after a thickness reduction of $\varepsilon = 14 \, \%$. 
The bonding strength of the copper coatings after deposition and flat rolling.

The substrate material, surface preparation as well as spraying condition are known to have an influence on the tensile adhesion strength of the coatings. For the material combination steel substrate/copper coating applied by cold gas spraying the typical values of adhesion strength are reported to be in the range of 9 to 30 MPa depending on parameters described above [13,14]. In the current study, it was shown that the annealing treatment is an efficient tool to increase the formability of the Cu coating without influencing its adhesion strength negatively.

4. Conclusions
In the present study the formability of cold sprayed copper coating on steel substrates was investigated in as sprayed and annealed conditions. The effect of the heat treatment and cold rolling on the adhesion strength of the coating and its mechanical properties was the main focus of the research. The results have demonstrated the high extent of the work hardening effect during the deposition. Thus, the Martens hardness of the bulk copper has increased by the factor of two after coating application. The subsequent annealing at a temperature of $T = 500 \, ^\circ C$ for $t = 5 \, h$ resulted in the recovery of the initial hardness values of the feedstock material. The microstructure of the coating also experiences a modification during the heat treatment. Due to the diffusion processes, the boundaries between particles almost disappear, leading to a homogenous microstructure.

Following observations were made about the coating behavior during the cold rolling process. The non-heat-treated samples are prone to crack initiation and propagation in the copper coating due to cold rolling process. For a thickness reduction of $\varepsilon = 14 \, %$, the bonding strength decreased drastically by a factor of 2. If the sample was annealed prior to cold rolling, no cracking and no coating delamination was observed after rolling. The bonding strength is not influenced negatively. The heat treated samples were structured by the rolling of channel-like grooves in the coating with a maximal structure height of $h_{\text{depth}} = 120 \, \mu m$. Concluding, it can be stated that an annealing treatment can drastically increase the formability of cold sprayed coatings by retaining the initial bond strength. The current study showed that metal forming processes like cold rolling are applicable for cold gas sprayed coatings in the annealed condition. Moreover, coatings can be structured to increase the bonding surface and to create a suitable surface topology for high pressure die casting process.

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