Investigation of the interfacial reactions between steel and aluminum coatings for hybrid casting

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Abstract. Coating of AA7075 was applied by means of cold gas spraying on steel substrates of 22MnB5 and DC04 as an interlayer for high pressure die casting of aluminum/steel hybrid components. The morphology and growth kinetics of intermetallic compounds formed at the interface between coating and steel has been investigated. Furthermore, the effect of alloying elements on the formation of the intermetallic phases was analyzed. The coated samples were heat treated by means of induction heating at the temperature T = 550 °C with different dwell times in the range of 10 s < t < 5 min. The reaction layer growth was examined by means of scanning electron microscope (SEM) with energy dispersive spectroscopy (EDS). Additionally, the intermetallic compounds were characterized by means of nanoindentation. Intermetallic compounds of AlFe phases occurred as the major constituent in the reaction zone for different combinations of coating and substrates.

1. Introduction

Trends in the modern automotive industry such as lightweight constructions give rise to the use of hybrid structures and therefore the demand for new joining techniques of dissimilar materials arises, in particular aluminum and steel. The hybrid structure of steel sheet and aluminum cast provides a favorable combination of good mechanical properties and reduced weight. For decades, aluminium and steel have remained the most significant materials in automotive industry. Joining of these materials, however, is still a challenging task due to dissimilar thermophysical and metallurgical properties and the formation of brittle intermetallic phases. Besides welding and brazing, the high pressure casting represents another technique for joining dissimilar metals. Casting of grey cast iron liners with aluminum [1] or Al inserts with Mg alloy [2] in the engine crankcase is already state of the art. Pressure casting of aluminum onto a steel insert has been also at the focus of research at RWTH Aachen University in recent years [3]. The cooperation between the foundry institute and industrial partners resulted in a "VarioStruct" prototype of the roof cross-beam of the vehicle [4]. Despite these successes, further improvement of the compound properties is required, as the problem of the brittle intermetallic phases and the gap formation at the aluminum/steel interface has not been completely solved. Due to enormous cooling rates after pressure casting and insufficient wetting of steel by aluminum, it is difficult to achieve a metallurgical bonding at the interface. In order to enable a gap-free material bonding between the steel and the cast aluminum alloy, different coatings for steel inserts are investigated in the current study. The main challenge is the application of coatings with a reliable adhesion on the steel substrate which facilitate metallurgical bonding with the aluminum melt during the pressure casting. The control of the reaction zone at the interface between aluminum and steel is of special interest regarding the mechanical properties of the joint, in particular its strength. The majority of the studies analyse the growth behaviour of intermetallic phases during hot dip aluminizing of steel [5–8]. Some of them were focused on the effects of alloying elements on the morphology and growth kinetics of the intermetallic compounds [5,7]. During the high pressure die casting process the surface of the steel insert is heated up to a temperature of T = 400 – 520 °C, while the temperature of the molten aluminum is about T = 700 °C [3]. A coating of AA7075 with a low solidus temperature was selected to enable a metallurgical bonding with the cast alloy during high pressure die casting. First die pressure casting trials showed that a thin surficial layer of the coating could be partially molten, while residual coating remains solid nearby the interface of the substrate material. Thus, the diffusion processes and the formation of the intermetallic phases at the interface between coating and steel take place in the solid state. In the current study two different steel inserts of DC04 and 22MnB5 which are commonly used in the automotive industry were selected. In order to investigate the influence of the alloying elements of the steel on the morphology and growth kinetics of the intermetallic compounds, coated samples have been heat treated at temperatures of T = 550 °C with different dwell times in the range of 10 s < t < 5 min. In order to reproduce the high heating rates typical for high pressure die casting, induction heating has been chosen in the current study.

2. Experimental

The experimental procedure consisted of a cold spraying process, followed by heat treatment and the analysis of the coated samples. The principle scheme of the experimental setup is shown in Figure 1.

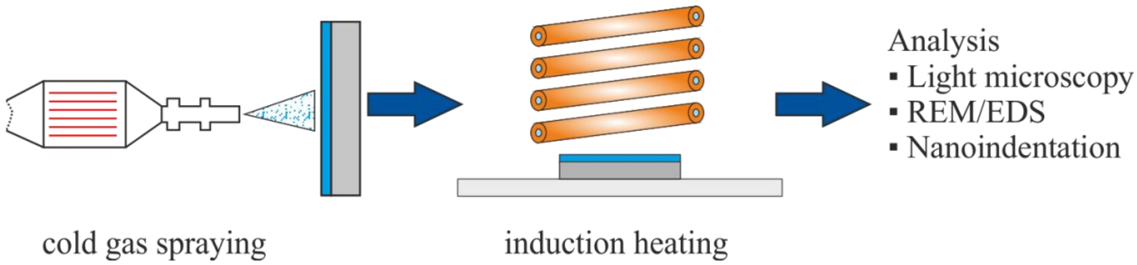

Figure 1. Principle scheme of the experimental procedure

2.1. Cold spraying

In the present study, the conventional cold rolled deep-draw steel DC04 and steel 22MnB5 were selected as substrate materials. Alumina with the grit size of F150 was used as blasting abrasive for surface pre-treatment of the steel prior to cold spraying. The steel sheet was punched to rectangular plates with the dimensions of $l \times b \times d = 200 \times 30 \times 2\ mm^3$. Due to the low solidus temperature of the alloy AA7075, it was chosen as feedstock material for coating application to achieve a metallurgical bonding with the aluminum melt during the high pressure die casting by partial melting of the coating. The compositions of the materials are given in Table 1. AA7075 powder with a particle size distribution of -63 + 20 µm was delivered by TLS Technik GmbH (Bitterfeld, Germany). The Kinetics 8000 Cold spray system from Oerlikon Metco (Pfäffikon, Switzerland) was used for the deposition process with nitrogen as process and carrier gas.

Table 1. Chemical composition of the substrate and coating materials (wt.-%)

	Si	Mn	Mg	C	S	P	Cu	Al	Fe	Zn	Cr
DC04		0.40		0.08	0.03	0.03			bal.		
22MnB5	0.30	1.26		0.23	0.01				bal.		0.12
AA7075	0.40	0.30	2.50				1.60	bal.		5.50	

2.2. Heat treatment

The coated samples were heat treated at the temperature of T = 550 ± 5 °C by using an induction heating system from iew Induktive Erwärmungsanlagen GmbH (Gumpoldskirchen, Austria) with five different dwell times varying between t = 10 s and t = 5 min. The temperature of the samples was controlled by a pyrometer.

2.3. Analytical methods

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 (Oberkochen, Germany) with Axio Vision image analysis tool. Universal hardness HU were determined using a calibrated TI 950 TriboIndenter, Bruker Corporation (Billerica, Massachusetts, USA) by applying a Berkovich tip (indenter code TI-0083) with a nominal edge radius of approximately r = 20 nm using a loading force of F = 8 mN. The indentation was performed at five different positions with the same distance from the interface between steel and reaction zone to calculate the average value. In order to identify the reaction layers at the interface between steel and aluminium alloy after heat treatment, an energy dispersive spectroscopy analysis was performed by means of the scanning electron microscope Zeiss Leo 1530 with the EDS device Bruker Quantax 200.

3. Results and discussion

Figure 2 shows cross sections of the AA7075 coatings on two different substrate materials. Due to a good plasticity of the feedstock material the coatings of AA7075 exhibit a tight bonding and low porosity regardless of substrate material. The mean thickness of the coatings was approximately d = 140 µm.

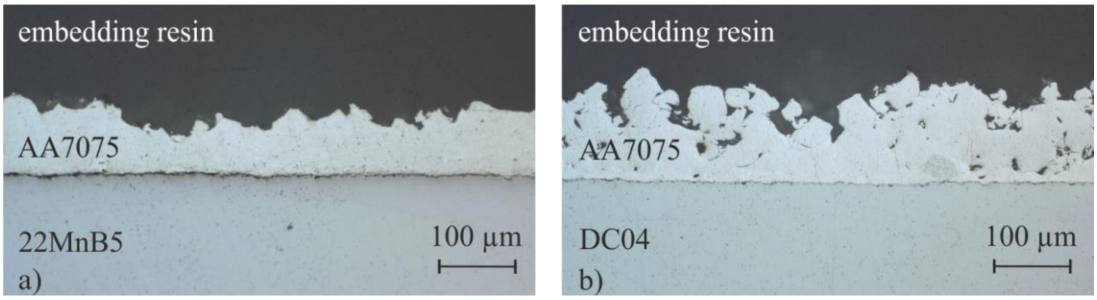

Figure 2. Cold sprayed coatings of AA7075 on a) steel 22MnB5 and b) steel DC04

3.1. Microstructural analysis of the coatings after heat treatment

After the heat treatment at the temperature of T = 550 C for t = 10 s, a thin intermetallic layer could be observed at the interface between AA7075 coating and steel DC04 as well as at the interface between AA7075 coating and steel 22MnB5, Figure 3a, d. The morphology of the intermetallic compounds was similar for both substrate materials. When the heat treatment time reaches t = 60 s, some pores could be observed at the interface between intermetallic layer and the coating. This effect became more pronounced with increased dwell time of the heat treatment, Figure 3c, f. It can be attributed to the Kirkendall effect due to the higher mobility of Al atoms compared to Fe resulting in a predominated diffusion flux from the AA7075 coating towards the reaction zone. A similar behaviour was described by Springer et al. for friction stir welded compounds at the interface between DC01 and AlSi5 [9].

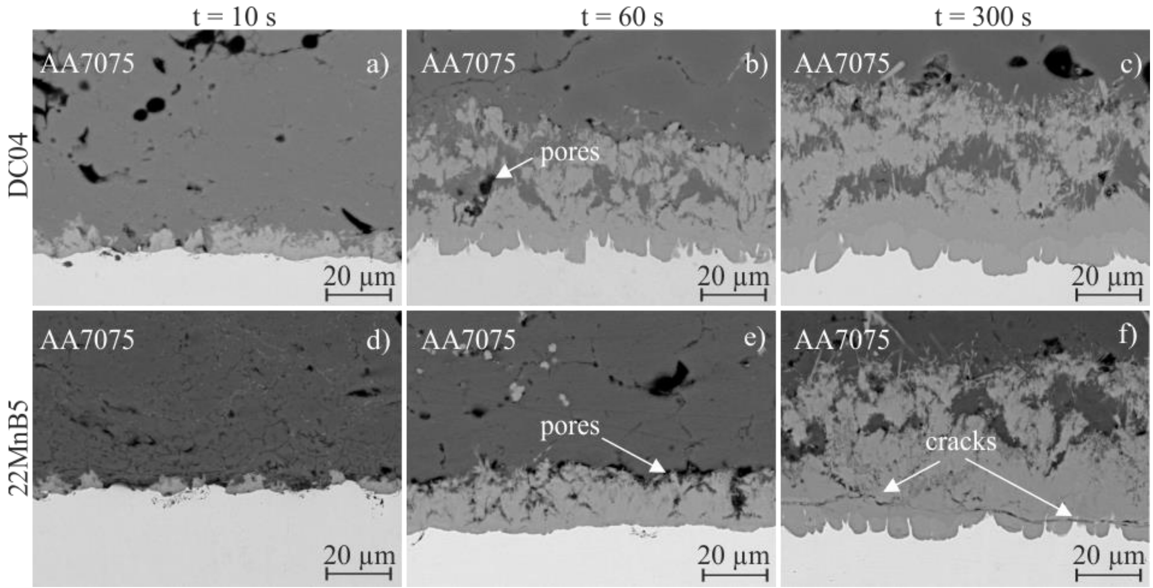

Figure 3. SEM images of the interface between AA7075 coating and steel DC04 after heat treatment at T = 550 °C for a) t = 10 c, b) t = 60 s, c) t = 300 s and between AA7075 coating and steel 22MnB5 after heat treatment at T = 550 °C for d) t = 10 s, e) t = 60 s, f) t = 300 s

Furthermore, some cracks occurred within the reaction zone between intermetallic layers. This phenomenon was considerably stronger for steel 22MnB5, Figure 3f. Similar crack propagation behaviour was described by Springer et al. during immersion of steel DC01 into aluminium melt AlSi5, accompanied by intermetallic phase formation [9]. Furthermore, Windmann et al. described crack formation in the reaction zone of Al-coated steel 22MnB5 during heat treatment at T = 920 - 1,000 °C, pointing out a preferred crack propagation along Al_5Fe_2 intermetallic phase [10]. The thermal stresses occurring during the cooling process could be a possible explanation for the crack formation. In Figure 4 thermal expansion coefficients of both steel grades and AA7075 coating is presented for a relevant temperature range. The occurrence of the cracks in the reaction zone can be explained by low fracture toughness of the AlFe intermetallic phases. Indeed, the difference between coefficients of thermal expansion between coating and steel 22MnB5 is slightly higher as in the case of steel DC04, however, it is too low to explain clearly such a different behaviour regarding the crack initiation.

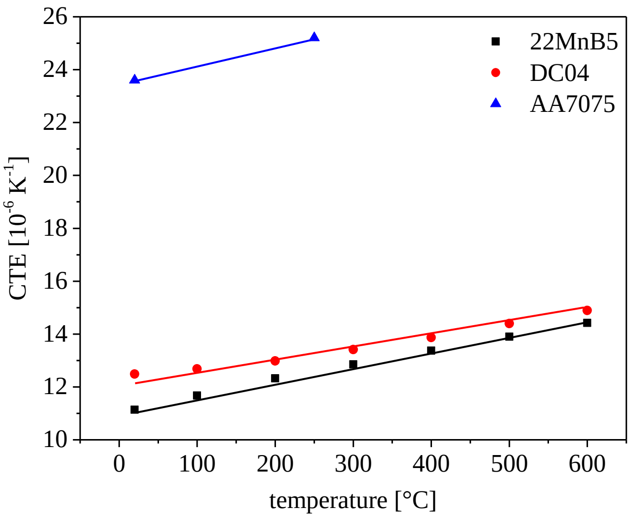

Figure 4. Coefficient of thermal expansion (CTE) for steel substrates and AA7075 alloy [13]

3.2. Analysis of the intermetallic compounds and their growth kinetics

The morphology as well as chemical composition of the intermetallic compounds was investigated by means of SEM/EDS measurements. Figure 5 shows the line scan of the atomic fraction profile at the interface between the steel and the coating. A special characteristic of the reaction zone is that after longer heat treatment times, it could be subdivided into two regions with a low-iron region between them. Furthermore, there are two intermetallic layers with different compositions to be found in the reaction zone adjacent to the steel substrate.

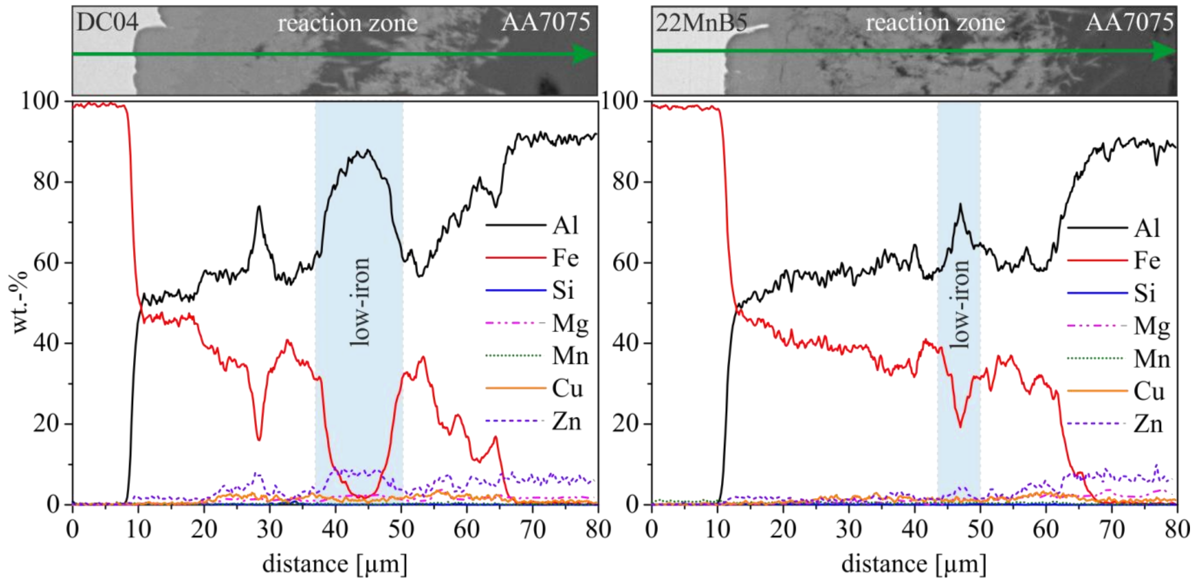

Figure 5. SEM/EDS analysis of the intermetallic compounds after heat treatment time of t = 5 min at the interface between AA7075 coating and a) DC04 steel and b) 22MnB5 steel

According to EDS analysis, a thin layer with a thickness of about s_{IMP} = 8 µm at the steel interface has a composition corresponding to that of $FeAl_2$ or Fe_2Al_5, while the composition of the thicker one with a thickness of about s_{IMP} = 20 µm indicates the formation of Fe_4Al_{13} or $FeAl_3$. These results are in accordance with the data reported in the literature [6,10,11]. Some Zn content of about m(Zn) = 1,4 wt.-% could be observed in the reaction zone, however, the content of Zn and other key alloying elements such as Cu and Mg is significantly higher in the aluminum matrix. Thus, only aluminum and iron play an essential role in the formation of the intermetallic compounds. The main differences in chemical composition of both steel grades are related to the Mn and Si content. In Figure 5b it can be seen that the Mn content in the steel 22MnB5 remains at the same level as in delivery state and no significant diffusion into the reaction zone could be detected.

The growth kinetics of the intermetallic compounds was determined from the measurements of the reaction zone thickness. In Figure 6 the average thickness of the intermetallic phase after the heat treatment is plotted as a function of the square root of time. The linear regression has been used to describe the relationship between the thickness of the reaction zone and the square root of the heat treatment time. Commonly, this relation can be described by the parabolic law as for example in [5,7,12]

$$d = k \cdot t^{0.5} \tag{1}$$

In accordance with equation (1), the slope of the lines corresponds to the parabolic coefficient k. The growth kinetics of the intermetallic compounds for steel DC04 with k = 2.31 µm/$s^{0.5}$ is slightly faster than for the steel 22MnB5 with k = 1.73 µm/$s^{0.5}$. In the study of Springer et al., the reaction kinetics of the formation of AlFe and AlFeSi intermetallic compounds has been investigated for stir welding of steel DC01 and AlSi5 [12]. The measured coefficients k in this study were k = 0.201 µm/$s^{0.5}$ for the heat treatment temperature of T = 500 °C and k = 2.303 µm/s$^{0.5}$ for the heat treatment temperature of T = 600 °C.

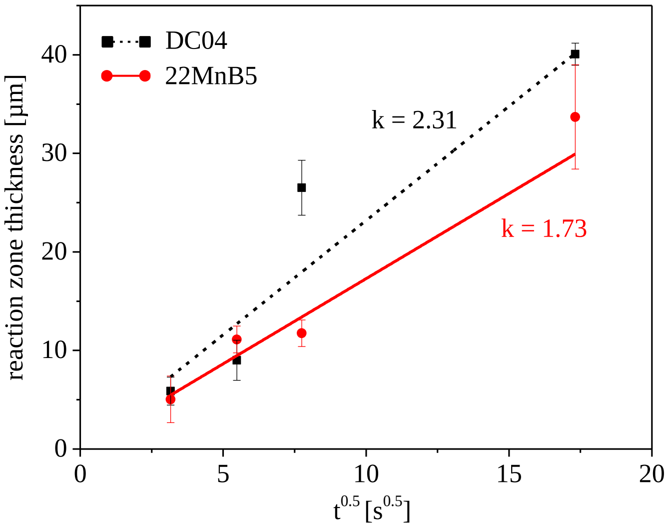

Figure 6. Mean thickness of the reaction zone between steel and AA7075 coating after the heat treatment at the temperature T = 550 °C

3.3. Hardness measurement

Nanoindentation measurements were performed to further characterize the reaction zone at the interface between steel and coating. The deep drawing steel DC04 exhibits hardness values of HU = 2.09 ± 0.12 GPa. For both substrate materials the maximal hardness in the reaction zone was measured in the intermetallic layers adjacent to the steel: HU = 11.30 ± 1.13 GPa for steel 22MnB5 and HU = 12.73 ± 0.30 GPa for steel DC04. As expected, in the region with a low iron content between the two intermetallic layers the hardness was considerably lower. In the region of the reaction zone bordering on the AA7075 coating, the hardness of the intermetallic layers was HU = 7.71 ± 0.89 GPa in case of steel DC04 and HU = 5.22 ± 1.30 GPa in case of steel 22MnB5. The results of the hardness measurement reflect the structure and morphology of the reaction zone observed by means of SEM/EDS analysis. The measured hardness of the intermetallic layer assumed as Fe_2Al_5 with a hardness of around HU = 12 GPa is a little bit higher as commonly reported in the literature, for example by Ogura et al. with HU = 7.8 GPa [14] or by Dybkov et al. with HU = 8.9 ± 0.9 GPa [11]. This can be possibly explained by the presence of alloying elements and in particular Zn as a substitute for Al in the $Fe_2Al(M)_5$ intermetallic phase with M = Zn, Cu or Mg.

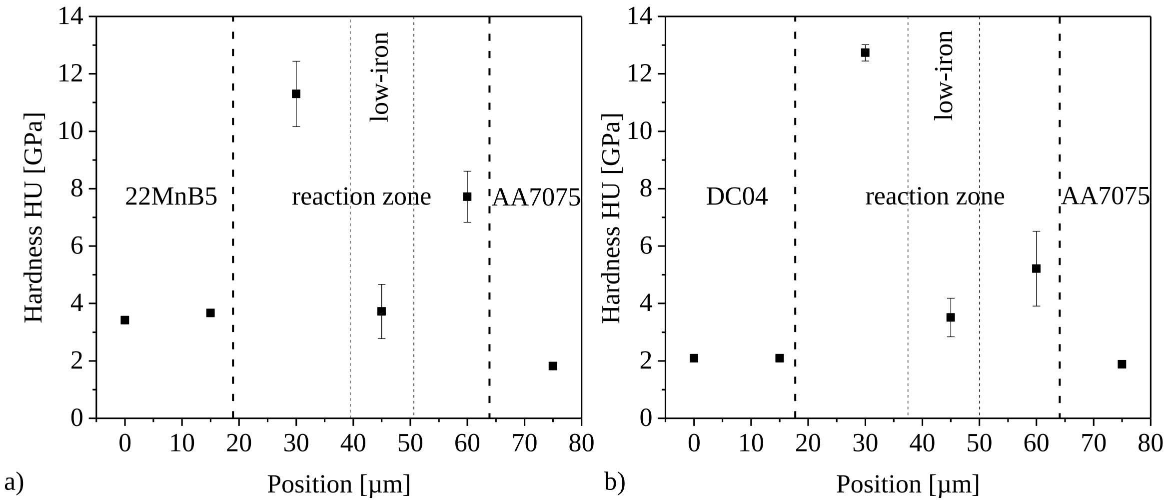

Figure 7. Hardness of the reaction zone after heat treatment time of t = 5 min at the interface between a) steel 22MnB5 and AA7075 coating, b) steel DC04 and AA7075 coating

4. Conclusions

In the current study, the formation of the reaction zone between cold gas sprayed AA7075 coating and steel DC04 or steel 22MnB5 during the heat treatment at T = 550 °C was experimentally examined. It could be shown that despite different chemical composition of both steel grades, the structure and morphology of the intermetallic layers at the interface between steel and coating were nearly identical. The following findings can be reported based on the results of the current study:

- For both substrate materials, the intermetallic phases of $FeAl_2$, Fe_2Al_5, Fe_4Al_{13} or $FeAl_3$ detected by means of SEM/EDS analysis were found to be the main components of the reaction zone between steel and AA7075 coating.
- The reaction zone consists of two AlFe intermetallic layers separated by a thin zone with a low-iron content. Furthermore, pores occurred at the interface between the reaction zone and coating, possibly due to the Kirkendall effect.
- Growth kinetics of the intermetallic compounds for steel DC04 with parabolic coefficient of growth k = 2.31 µm/s$^{0.5}$ was slightly faster than for the steel 22MnB5 with k = 1.73 µm/s$^{0.5}$.
- For steel 22MnB5, an occurrence of the crack formation in the reaction zone after cooling from heat treatment temperature was distinct compared to the steel DC04. It can be a crucial factor for the reliability of high pressure die cast Al/steel hybrids.

5. References

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