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# Using ultrasonic atomization to recycle aluminium bronze chips for additive laser directed energy deposition

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**Abstract.** In the post-processing of large maritime components, a considerable amount of waste in the form of milling and grinding chips is produced. At the same time, additive manufacturing technologies have shown great potential in producing high-volume parts for maritime applications, allowing novel design approaches and short lead times. In this context, this study presents a sustainable approach to recycle and use aluminium bronze waste material, generated during post-processing of large cast ship propellers, as feedstock for laser-powder directed energy deposition. The recycling technology used to produce powder batches is inductive re-melting in combination with ultrasonic atomization. The derived metal powders are characterized using digital image analysis, powder flowability tests, scanning electron microscopy as well as energy dispersive X-ray spectroscopy. Compared to conventional metal powders produced by gas atomization, the recycled material shows excellent sphericity and a powder size distribution with a higher content of finer and coarser particles. Metallographic sections of deposited additively produced specimens show an increased hardness and reduced ductility, but also competitive densities and higher yield and ultimate tensile strength compared to cast material. The process chain shows high potential for the maritime sector to enable circular and sustainable manufacturing.

## 1. Introduction

The majority of modern ship propellers are produced from aluminium bronze via sand casting. The alloy offers good material properties and high resistance to cavitation and corrosion in sea water which makes it especially suitable for maritime applications [1]. After casting and several machining steps, ship propellers are ground in a dry grinding process to generate the final geometry of the propeller blades. Due to the large component sizes, up to 200 kg of



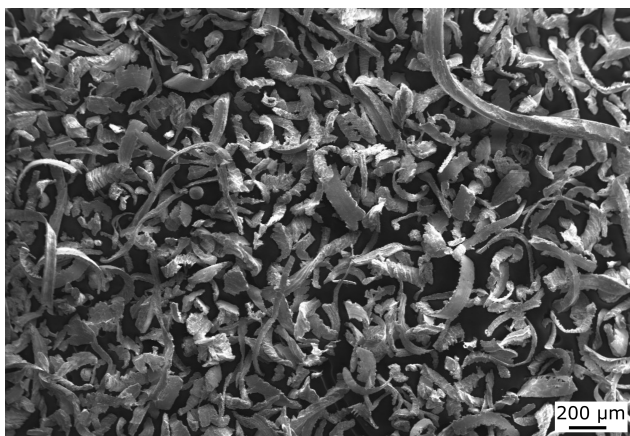
material is removed from the part during this process. This paper investigates an approach to recycle this aluminium bronze swarf to feedstock for additive manufacturing (AM) to be used in a powder-based directed deposition process with laser beam (DED-LB).

There are some authors who investigate the use of metal swarf as feedstock for AM processes. For example, Jackson et al. [2] produce specimens from ball milled 304L steel machining chips with good material properties of additive specimens. Razumov et al. [3] generate metal powder by melting steel machining chips with a plasma torch. They are able to produce spherical particles with acceptable particle size distribution (PSD) for a laser powder-bed fusion process. After heat treatment, the mechanical properties of produced specimens exceed the conventional material properties.

For AM feedstock, the characteristics and properties of the metal powder are very important for good processability and part quality. In order to analyze the powders, certain characterization processes can be applied. They often include measurements of PSD, morphology of particles, flowability or chemical composition. In the literature, applicable methods and procedures for characterization are investigated extensively. For example, Wimler et al. [4] and Mitterlehner et al. [5] investigate various methods to measure PSD and morphology of AM powders. Kiani et al. [6] use statistical approaches for powder characterization. The studies of Baesso et al. [7] and Spierings et al. [8] apply and compare methods of standardized flowability tests.

The raw material that was used in this study is shown in the SEM image in Figure 1. It can be stated by sight alone that these particles are not well suited for a DED-LB process where flowability is important for a stable process. In a previous study, the authors of this paper managed to deposit these grinding chips without further processing. While this was generally possible, the flowability was not sufficient and the contamination with foreign particles caused significant welding defects [9].

Therefore, this study utilizes a recycling process called ultrasonic atomization (UA) to produce metal powders from the chips. The UA approach is described by Lierke and Griebhammer [10]. More recently, Żrodowski et al. [11] investigated UA to produce AM powders. For UA, the material is melted down and dropped on a sonotrode that vibrates with frequencies in the kHz-range. As a result, fine metal droplets are ejected. Due to their surface tension they form into a spherical shape in which they solidify. The process takes place in an inert atmosphere of Argon to prevent oxidation. The particle size distribution (PSD) can be controlled by the frequency of the sonotrode.



**Figure 1.** SEM image of grinding chips before the ultrasonic atomization process. The grinding chips consist of long and sharp-edged particles with a very inconsistent PSD and morphology.

## 2. Materials and Methods

### 2.1. Feedstock

The material used in this study was from grinding chips of aluminium bronze (CuAl10Ni5Fe5). The material was produced during post processing of cast propellers by means of grinding belts utilizing corundum as abrasive particles. The abrasive was primarily composed of aluminium oxide ( $Al_2O_3$ ). No cooling lubricants were used during the process. The material was not subjected to any cleaning or purification measures before or after the ultrasonic atomization process.

To compare the properties of recycled grinding chips, commercially available gas atomized aluminium bronze powder (CuAl9.5Fe) from Oerlikon Metco was characterized as well. The nominal PSD according to the producer was 45  $\mu m$  to 125  $\mu m$ .

### 2.2. Ultrasonic atomization

The UA process was carried out using the induction crucible-based ultrasonic atomization system rePowder from Amazemet. The heating zone consisted of a graphite-based crucible and pouring system. The nozzle to feed the melt had a diameter of 1 mm to avoid clogging with possible contamination of leftover ceramic particles from the grinding process. The feedstock material was heated to  $T = 1300^\circ C$  and poured down through the nozzle system onto a carbon fiber sonotrode, vibrating with a frequency of  $f = 40$  kHz. Both the melting and atomization processes were carried out in an inert gas atmosphere of Argon (grade 5.0). Powder yielded from the process was 77%. The remaining material was mainly slag consisting of organic and ceramic contaminations.

### 2.3. Material characterization route

To measure PSD and particle morphology, dynamic image analysis was performed on a Microtrac Retsch CamSizer X2. Scanning electron microscopy (SEM) as well as energy dispersive X-ray spectroscopy (EDX) were conducted on a Zeiss LEO 1455VP. The flowability of the material was evaluated using two characteristics: The Carney flowmeter test according to ASTM B964 and the Hausner ratio. To calculate the Hausner ratio, the bulk density was measured using a Pharma Test SV100 based on DIN ISO 3923, and tapped density was measured with a JEL STAV 2003 based on DIN ISO 3953. Furthermore, the level of oxygen content was measured on a Leco ONH836 elemental analyzer.

### 2.4. DED equipment

The DED-LB system consisted of a Trumpf TruLaser Cell 7020 equipped with a Yb:YAG disc laser ( $P_L = 2$  kW, focal length of 200 mm, wavelength of 1030 nm) a Trumpf three-jet powder nozzle and a GTV PF2/2 powder disc feeder. The welding process parameters are listed in Table 1. As substrate, 10 mm thick 1.4404 steel plates were used. To determine the mass flow rate under realistic conditions, the powder feed rates for disc speeds of 1, 3, 5, 7 and 9 rpm were measured three times and averaged.

**Table 1.** DED-LB welding process parameters as used in this study to manufacture specimens.

Laser power (W)	Feed rate (mm/min)	Material mass flow (g/min)	Laser spot size (mm)	Argon shielding gas (l/min)	Helium carrier gas (l/min)
1300	750	11	1	10	4



### 2.5. Analyses of properties

Metallographic cross sections were prepared using multiple grinding steps, 6, 3 and 1  $\mu\text{m}$  diamond polishing suspensions and then etched with Adler reagent. To obtain tensile values, flat tensile specimens according to ISO 6892-1 were manufactured from the deposited cuboid in the X-Y-plane and cut out by means of wire electrical discharge machining (EDM). The tensile tests were carried out on a Zwick/Roell Z150 at room temperature.

## 3. Material characterization

In order to compare the recycled powder from ultrasonic atomization with the reference powder, selected material characteristics were analyzed and evaluated. The steps included dynamic image analysis to measure the PSD and morphology of particles, SEM to gather qualitative shape information and EDX to detect contaminations. Furthermore, standardized flowability tests and oxygen measurements were carried out.

### 3.1. Dynamic image analysis

The PSD measured in the dynamic image analysis of the recycled powder shows a distribution of  $d_{10} = 45 \mu\text{m}$  to  $d_{90} = 179 \mu\text{m}$  with  $d_{50} = 96 \mu\text{m}$ . The variable  $d$  represents the equivalent particle diameter that corresponds to the diameter  $x_{\text{area}}$  of a circle with an equivalent area. The span of the PSD is wider both on the finer and coarser end compared to the gas atomized reference powder which has a PSD of  $d_{10} = 66 \mu\text{m}$  to  $d_{90} = 133 \mu\text{m}$  ( $d_{50} = 99 \mu\text{m}$ ). Both measurements are shown in Figure 2 and, together with other measurement data, are summarized in Table 2. Besides the PSD, the sphericity of particles also plays an important role in flowability. To evaluate this factor, the aspect ratio  $b/l$  as a factor of each particle's shortest chord  $x_{c,\text{min}}$  and longest chord  $x_{Fe,\text{max}}$  can be evaluated:

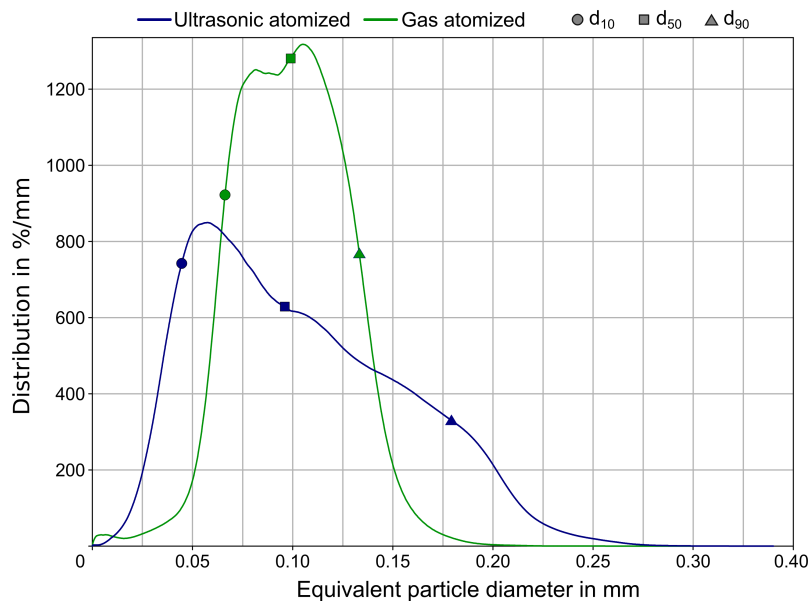
$$\frac{b}{l} = \frac{x_{c,\text{min}}}{x_{Fe,\text{max}}} \quad (1)$$

A value of  $b/l = 1$  indicates a perfectly spherical particle, a deviation results in values of  $b/l < 1$ . The results for the powders of this study reveal that the ultrasonic powder shows a significantly higher number of spherical particles than the commercial gas atomized powder. Approx. 82% of the analyzed ultrasonic particles have an aspect ratio of  $b/l \geq 0.9$ . In other words, 82% of all particles are almost perfectly spherical. This is superior to the reference powder, where only 43% fall into the same sphericity range.

**Table 2.** Summary of the results of the different material characterization tests for sieved chips, ultrasonic atomized powder and gas atomized reference powder.

	$d_{10}$ ( $\mu\text{m}$ )	$d_{50}$ ( $\mu\text{m}$ )	$d_{90}$ ( $\mu\text{m}$ )	$b/l_{\text{mean}}$	$b/l \geq 0.9$ (%)	Carney flow rate (s/150g)	Bulk density ( $\text{g}/\text{cm}^3$ )	Tap density ( $\text{g}/\text{cm}^3$ )	Hausner ratio	Oxygen content (%)
Chips <sup>1</sup>	81	131	188	0.51	2	44.8	1.80	2.50	1.39	0.0727
Ultrasonic	45	96	179	0.92	82	8.6	4.67	4.90	1.05	0.0125
Reference	63	91	133	0.84	43	10.5	3.97	4.44	1.12	0.0196

<sup>1</sup>Chips were sieved with a 125  $\mu\text{m}$  mesh analysis sieve



**Figure 2.** Particle size distribution for recycled, ultrasonic atomized powder and commercial, gas atomized powder.

#### 4. SEM and EDX analysis

In order to obtain image-based information on the metal powders, SEM analyses were carried out. Furthermore, EDX was performed to analyze the chemical composition of the droplets and to detect possible contamination.

The EDX analyses of the ultrasonic atomized powders did not reveal any deviations in the chemical composition compared to the cast material CuAl10Ni5Fe5. While Źrodowski et al. detected a depletion of elements like Manganese in certain alloys in previous studies [11], aluminium bronze seems to be stable in ultrasonic atomization. EDX analysis of the reference material CuAl9.5Fe also shows a chemical composition of the particles that was well within the expected range.

In the SEM images depicted in Figure 3 a), the reference powder has a low number of spherical particles and contains a significant number of spattered and agglomerated particles as well as many adhesions in the form of satellites. Some elongated particles are also clearly visible. Compared to other studies, such as from Mitterlehner et al. [5], the quality of the gas atomized powder seems to be in line with industry standards where elongated particles, satellites and agglomerates can be observed quite frequently.

The images of the recycled chips (see Figure 3 b) are uniformly round in majority. Only a few agglomerated, fused particles or satellites can be seen. The findings confirm the data gathered by the dynamic image analysis in which the recycled powder showed a higher percentage of particles with a favorable aspect ratio of  $b/l \geq 0.9$  and significantly fewer spherical particles were measured in the commercial material.

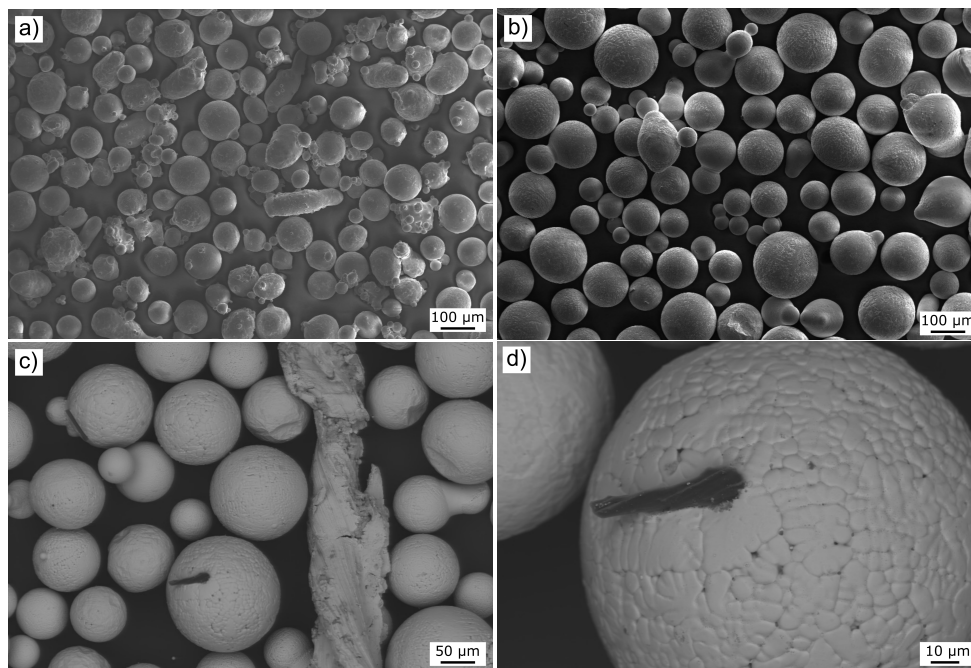
While no foreign particles could be detected in the SEM images of the reference powder, impurities were found in the recycled material. For instance, few elongated particles could be detected that seem to be larger droplets of metal, potential contamination from a chamber wall of the equipment (see Figure 3 c). The number of such particles was very low and a relevant influence on the AM process can likely be ruled out.

In the image in Figure 3 c) and d), a fiber adhering to powder particles can be observed. Such fibers of approx. 5  $\mu\text{m}$  to 10  $\mu\text{m}$  width were also detected on other particles. It is highly likely that these fibers originate from the vibrating sonotrode used in the ultrasonic atomization process of this study. The sonotrode was made of binder-free, woven carbon

fibers. This type of material is beneficial due to its high fatigue strength and resistance to high temperatures. Apparently, however, fibers detach from the plate when exposed to the hot melt, and subsequently contaminate the powder. While these impurities are not expected to affect the flowability due to their small size, traces of carbon can have a negative influence on the welding process and the component quality, as they could cause inclusions or porosity. To avoid this, a sonotrode from a different material, ideally with a similar alloy to the chips should be used in the future.

As mentioned above, a previous study by the authors of this study [9] analyzing the as-collected grinding chips showed high contamination with organic and ceramic particles that were residues from the grinding belt used during manufacturing. In the recycled material of this study, no such impurities were found in the final powder. This is due to the complete re-melting of the swarf prior to the atomization process. During the melting, all organic and ceramic elements float to the top of the melt. This brings the advantage that nearly all foreign particles and contaminations can be removed during or after the process as a slag. This significantly improves the quality of the powder compared to other recycling approaches like milling or grinding, where additional cleaning steps must be considered.

Measurements via an elemental analyzer show that the UA powder has an average oxygen content of 0.0125%. The reference powder was measured with 0.0196% of oxygen on average. Both results are in an acceptable range for AM powders. The lower values for the recycled powder indicate a sufficient inert gas atmosphere in the UA equipment.



**Figure 3.** a) SEM images of gas atomized reference powder, b) ultrasonic atomized grinding chips, c) intact chip and carbon fiber in recycled powder and d) detail of particle with adhering fiber.

## 5. Standardized flowability tests

In order to quantify the powder flow characteristics, the Carney flow test was performed and the Hausner ratio was calculated as a factor of bulk and tapped density. The flowability values

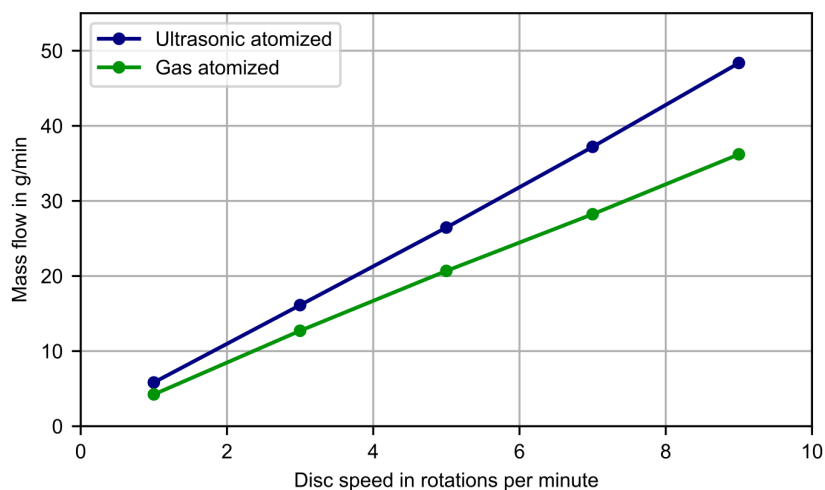
are summarized in Table 2. In the Carney test, 150 g of recycled materials needed 8.6 s on average to flow out of the funnel, while the gas atomized material took on average 10.5 s. The measured Hausner ratio of 1.05 for the recycled powder indicate an excellent flowability. With a Hausner ratio of 1.12, the commercial powders show a good flowability (compare Baesso et al. [7]). It can be noted that the results of the two flowability tests are consistent with the measurements from dynamic image analysis and SEM images. As pointed out by Kiani et al. [6], a wider range of particle sizes as present for the UA powder can lead to a reduced flow rate due to more contact points between the particles and tighter packing. For the UA powders, the influence of the wider PSD compared to the reference powder seems to be lower than the high sphericity and the homogenous shape of the particles, leading to better flow behavior.

In summary, the results of the material characterization show that ultrasonic atomization is well suited to produce high-quality, spherical AM powders with better flow and morphology properties compared gas atomized powders in this study. While few process-induced foreign particles (carbon fibers) can be found in the recycled powder, nearly all contaminations from the grinding process were removed during ultrasonic atomization.

## 6. DED-LB processing and results

### 6.1. Flowability and processability with disc feeder

While standardized flowability tests as performed during material characterization can give a theoretical implication to compare different powders, processability in specific DED-LB systems must be ensured as well. To verify sufficient powder flow and to determine the powder mass flow in g/min in relation to the disc rotation speed of the powder feeding systems, the powder output after 2 min was measured for increasing disc speeds. The results are shown in Figure 4. Each data point was measured three times. The average error was  $< 2\%$  for all measuring points and is not displayed in the graph. Both powders show a steady and linear trend with increasing disc speeds. The powder flow from UA, however, is characterized by a mass flow that is constantly about 30% higher compared to the gas atomized material. While the relatively high difference of 30% was not expected, these results are again in line with the measurements in dynamic image analysis, SEM and standardized flow tests.



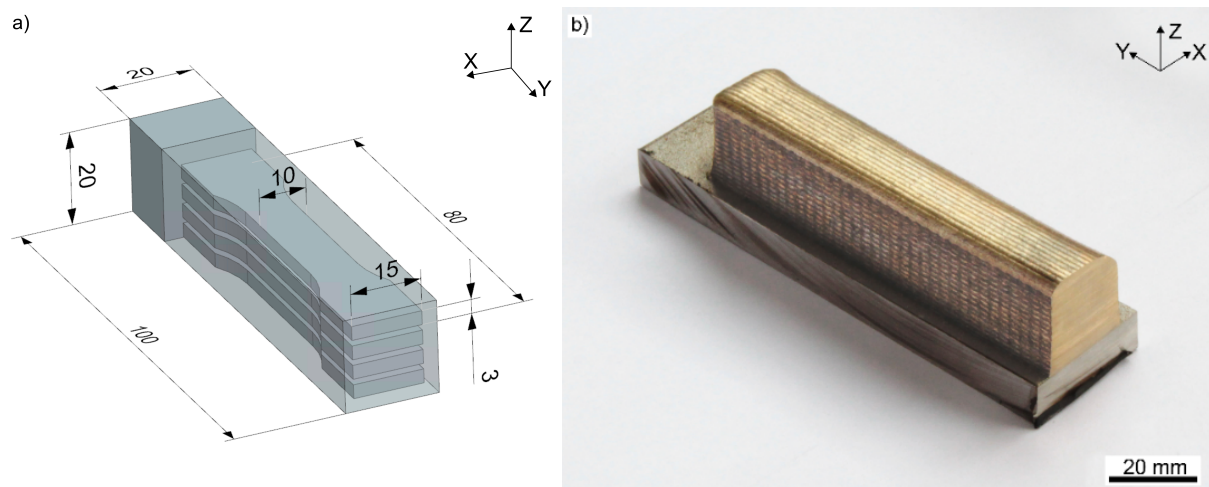
**Figure 4.** Results of analysis of flowability of recycled and commercial powders in a disc feed powder delivery system as used in the DED-LB system in this study.

### 6.2. Manufacturing of specimens

To gather mechanical properties of the recycled bronze powder, a specimen block was deposited, consisting of 35 layers with a total height of 20 mm. A meandric deposition strategy was

employed with a 90° rotation of the track direction after each layer and 50% track overlap. The specimen dimensions are depicted in Figure 5 a) and were chosen to extract a series of test specimens for metallographic analysis, hardness measurements and flat tensile specimens. The manufactured cuboid after separating the specimen for microstructure analysis is shown in Figure 5 b).

It should be noted that the welding parameters used (see Table 1) were optimized for the gas atomized material and not adapted for the recycled material due to the limited amount of powder available. As can be seen in the cross-section in Figure 6, the manufactured block shows a sag at the top left edge which is due to the DED parameters used. However, the geometrical deviation did not impair the mechanical properties of the material, and the targeted four tensile specimens could still be manufactured out of the block.



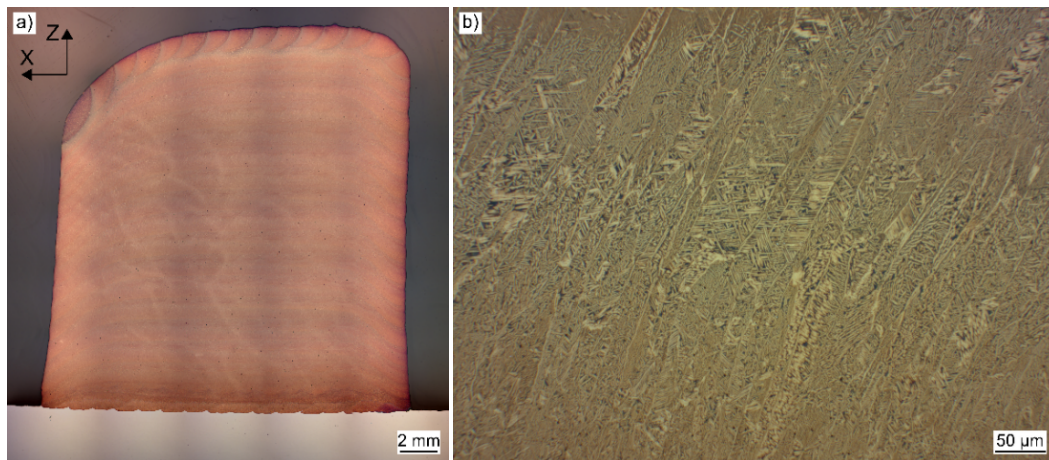
**Figure 5.** a) CAD drawing with nominal dimensions, the position of the micro section and flat tensile specimens, b) manufactured specimen block after sawing of substrate and micro section specimen.

### 6.3. Microstructure and mechanical properties

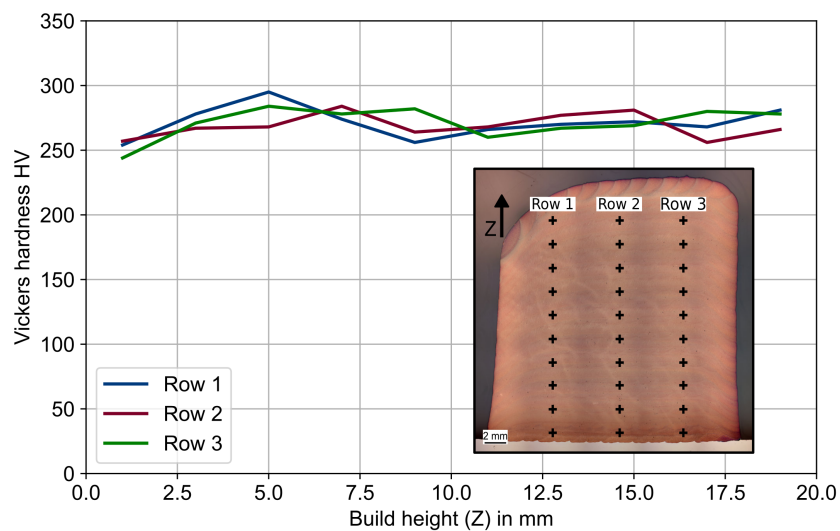
The etched metallographic cross-section in the X-Z-plane is shown in Figure 6 a) and b). No significant defects can be observed. Only very few small spherical gas pores can be detected in the cross-sections. The relative density is  $\rho > 99,8\%$ . The etched cross-section reveals a fine dendritic microstructure with a preferential grain orientation parallel to the build direction, which is common for AM microstructures.

The same specimen was also used to determine the hardness profile of the structure. Three parallel series of measurements were carried out with eleven measuring points, each in increasing sample height (direction of build-up). The results are plotted in Figure 7 and show a fairly constant hardness profile with an average hardness of 266 HV. According to the data sheet of the German Copper Institute for the investigated aluminium bronze [12], the hardness of cast microstructures usually is in the range of 150 HV to 180 HV. Compared to these values, the additively manufactured sample shows a hardening that exceeds the requirement for end use components.

In a casting process, usually low cooling rates prevail. For the alloy CuAl10Ni5Fe5 this leads to  $\alpha$ - and  $\kappa$ -phases that precipitate and grow at the boundaries of initially formed  $\beta$  grains (compare Culpan and Rose [13]). However, during a DED-LB process, this precipitation process



**Figure 6.** a) Overview image of the etched micro section, b) light microscopy close-up image of the microstructure. A grain structure with fine needles is visible.



**Figure 7.** Lines of three hardness measurements at different positions with increasing height of the additively manufactured specimen from ultrasonic atomized powder. A uniform hardness profile can be observed.

is suppressed due to the rapid cooling. As a result, a martensitic-like structure is generated (compare Hájek et al. [14]) that mainly consists of fine needles in the  $\beta'$ -phase and leads to the elevated hardness of the additive specimen.

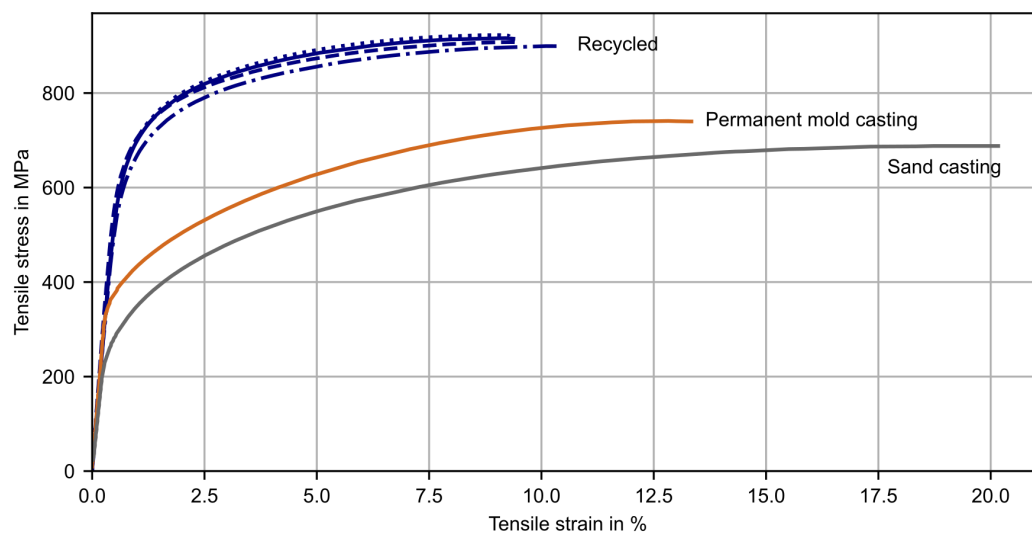
#### 6.4. Tensile properties

In order to obtain tensile properties for the specimen from recycled powder, flat tensile specimens were machined via wire EDM and tested. The results show a relatively brittle structure with fracture elongation of around  $\epsilon = 9\%$ , but a high yield strength of  $R_{p0.2} = 646$  MPa and tensile strength of  $R_m = 911$  MPa. The deviations in the measured data between the tensile specimens are small and confirm the observation of a homogeneous deposition that was mainly free from defects or large pores. Figure 8 shows the tensile stress curves for the recycled powder. The graph



also includes exemplary tensile curves for casting processes commonly used for aluminium bronze. Sand casting processes are characterized by their very slow cooling rate, and the permanent mold casting process by more rapid solidification. The plot in Figure 8 clearly indicates the influence of the cooling rate for this alloy: rapid solidification leads to high tensile strength and lower ductility, while slow solidification leads to higher values in elongation at break and lower values in tensile strength.

While the tensile properties combined with the elevated hardness values do not meet the requirements for end-use parts like ship propellers, other applications are possible. The high toughness and hardness could be advantageous, e.g. for hard coatings. For additive structures, heat treatment could be applied to allow the microstructure to precipitate  $\alpha$ - and  $\kappa$ -phases to reduce the hardness and increase tensile strength.



**Figure 8.** Results of tensile tests of DED-LB specimens from recycled powder in blue. The graph also contains tensile curves for the same alloy that were produced by sand casting in gray (very low cooling rate) and permanent mold casting in orange (faster cooling rate). The tensile test data for the casting processes are from material tests conducted by Mecklenburger Metallguss GmbH.

## 7. Conclusion

In this study, ultrasonic atomization was used to produce AM powder from aluminium bronze grinding chips from ship propeller manufacturing. The following summary can be drawn:

- The recycled powder was highly spherical, had excellent flowability properties, exceeding the gas atomized reference powder and was low in oxygen content. Ceramic and organic contaminants from the grinding process were successfully removed during UA.
- Metallographic cross-sections of a cuboid produced by DED-LB from the recycled powder reveal an almost defect-free microstructure with fine needle-shaped grains. Hardness measurements indicate that a martensitic-like  $\beta$ -phase was predominantly present which likely formed due to the high cooling rates.
- Tensile tests show a high yield strength and ultimate tensile strength but reduced fracture elongation in comparison to cast properties, caused by the relatively hard and therefore brittle microstructure of the additively manufactured specimens.

It should be noted that UA is an energy-intensive process, as all the metal must be melted in a crucible prior to atomization. This is at least partially at odds with the concept of sustainability. On the other hand, powder yields from UA are over 60% compared to typical 30-40% from gas atomization, with sustainably lower gas consumption. Therefore, mechanical recycling approaches should be investigated to reduce the energy input during recycling. However, this study shows that UA is a suitable production technology to produce high quality AM powders with excellent properties and low contamination.

### Acknowledgments

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