

Spray-formed Copper Alloys – Process and Industrial Applications

Hilmar R. Müller, Robert Zauter

Spray forming is a fairly new method of manufacturing metal semi-finished products. In this process, a melt is converted into solid state by the intermediate step of atomization. At the same time a compact preform is produced. The preform is a billet which is further processed in conventional forming processes. Spray forming is a process in between powder metallurgy and continuous casting. Its main advantage is the ability to produce materials which cannot be cast or can only be cast with strong segregation. Materi-

als produced successfully by spray forming are high-tin bronzes, aluminium bronzes and high-strength CuMnNi alloys. These materials and their industrial applications are presented.

Keywords:

Spray forming – Copper alloys – Aluminium bronze – High-strength – High-alloyed

Sprühkompaktierte Kupferlegierungen – Verfahren und industrielle Anwendungen

Sprühkompaktieren ist eine noch junge Methode zur Herstellung von metallischen Halbzeugen. Dabei wird eine Schmelze über den Zwischenschritt Zerstäubung in den festen Zustand überführt und gleichzeitig eine kompakte Vorform erzeugt. Diese Vorform ist ein Bolzen, der mit konventionellen Umformverfahren weiterverarbeitet wird. Sprühkompaktieren wird zwischen Pulvermetallurgie und Strangguß eingeordnet. Die Stärken liegen besonders bei der Herstellung von Werkstoffen, die nicht oder nur sehr schwierig zu gießen sind, da sie zu starken Seige-

rungen neigen. Werkstoffe aus den Gruppen der hochlegierten Zinnbronzes, Aluminiumbronzes und hochfesten CuMnNi-Legierungen haben inzwischen Anwendungen in der Industrie gefunden. Werkstoffe und Anwendungsbeispiele werden vorgestellt.

Schlüsselwörter:

Sprühkompaktieren – Kupferlegierungen – Aluminiumbronze – Hochfest – Hochlegiert

Alliages de cuivre obtenus par compactage au spray – procédé et applications industrielles

Aleaciones de cobre por proyección compactada – Elaboración y aplicaciones industriales

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Compared with other forming processes, such as continuous casting, spray forming is a fairly new method of manufacturing semi-finished products. In this process, a metal melt is converted into solid state by the intermediate step of atomization. Already in the early fifties, some proposals were published for atomizing metal, collecting it on a substrate and solidifying it [1]. The current method of spray forming is based on developments by Professor SINGER, Wales/UK [2]. At the beginning of the seventies, initial publications and patents came out. Efforts were focussed on the production of strip directly from the melt [3]. As a result of these activities at the University of Swansea, the company Osprey Metals was founded dealing with the development and marketing of the spray forming process. They give licenses to manufacturers of plants and users in the steel, aluminium- and copper-alloy industry. The atomization in droplets and the subsequent fast cooling with rapid solidification opens up new opportunities for the production of materials which, up

to now, could not be produced or produced only with strong segregation.

1 The spray-forming process

1.1 Classification of the process

The designation spray casting, which is also used, indicates that the process can be associated neither with powder metallurgy nor with classical casting methods. While the casting process deals with the production of semi-finished products made of castable alloys and powder metallurgy with the production of semi-finished or finished products made of powders of special alloys, spray forming is a combination of both. Main applications are the production of semis made of

- materials not castable or castable only with difficulty
- high-alloy materials
- strongly segregating materials
- composite materials (metal matrix composites).

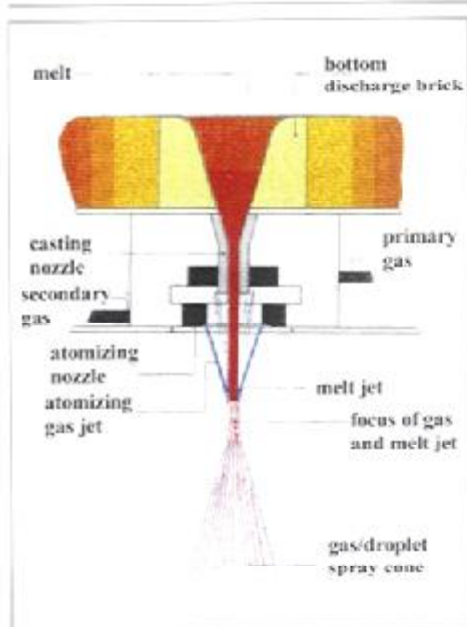


Fig. 1: Atomizer

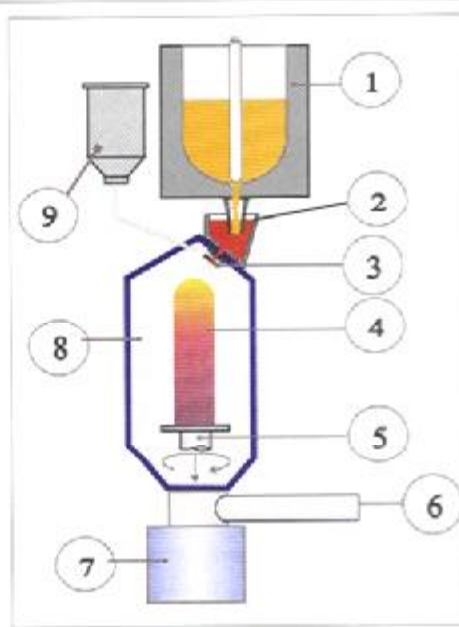


Fig. 2: Principle of the Wieland-Spray Forming unit. 1 = Holding furnace, 2 = Tundish, 3 = Atomizer, 4 = Billet, 5 = Withdrawal/rotation unit, 6 = Exhaust gas, 7 = Overspray container, 8 = Spray chamber, 9 = Particle injector



Fig. 3: Finished billet in the spray chamber

1.2 Process description

The melt is first guided from a tundish via a ceramic casting nozzle to a concentric atomizing gas nozzle. The melt jet is then dispersed to small droplets by an inert gas according to Figure 1. The average droplet diameter is about 60 μm . Compared to the metal powder production, the mass flow rate is significantly higher, namely up to 35 kg/min with a single atomizer and up to 70 kg/min with a twin atomizer. The primary gas nozzle runs with slightly elevated pressure and is used for guiding the melt jet. Figure 2 shows a typical spray forming arrangement. The melt is prepared in a melting furnace not being shown and then transported to the spray forming plant with a holding furnace. Then it flows through an inert gas shrouded tube into the tundish and from there through the casting nozzle to the atomizer.

A controlled stopper rod keeps the melt level in the tundish and therefore the mass flow rate constant. Bottom pouring and shrouding of the melt surface keep off oxygen coming from the surrounding air.

The spray chamber is also flooded with an inert gas, usually nitrogen. The atomization is similar to the production of metal powder but the flight distance of the droplets during spray forming is significantly shorter. Just before complete solidification the droplets hit a suitable substrate, which rotates and can be moved in a vertical direction, and solidify. A metal layer is formed and grows according to the flow rate of the droplets. Proportional to the growing layer, the substrate is withdrawn in order to keep the distance between the atomizer and the tip of the billet constant. The angle between the axis of the atomizer and the billet allows the diameter to be controlled merely by adjusting the withdrawal speed. Figure 3 shows the finished billet after the end of the spraying process with opened spray chamber.

and Figure 4 the billet being formed shortly after the beginning of the spraying process. The maximum diameter reached up to now is about 500 mm. Figure 5 shows an example.

Also other particles can be added to the primary gas flow. So it is possible to produce dispersion materials with big density differences between the particles and the matrix. The particles are homogeneously embedded in the solidifying metal.

Part of the solidifying droplets do not stick to the billet surface but follow the gas flow. This so-called overspray means a loss of metal in the order of 10 % to 40 %. Most of

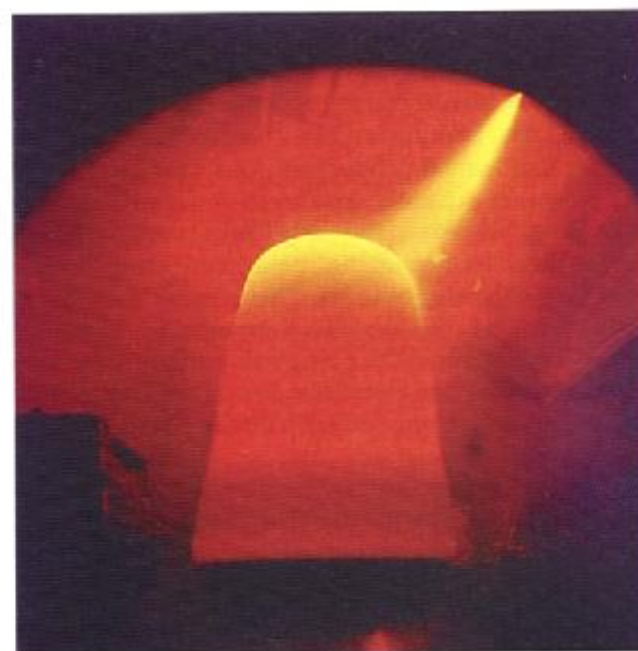


Fig. 4: View inside the spray chamber at the beginning of the process

Fig. 5: Billet CuCrZr, \varnothing 500 mm

the droplets are separated from the gas already at the bottom of the chamber, the remainder in a cyclone and bag filter. In Table 1 some typical technical data of a spray forming plant are collected.

1.3 Differences from conventional casting processes

The mere comparison of the macrostructure of continuously cast and spray-formed billets shows big differences. In Figure 6, the cross section of a continuously cast billet (on the left) is compared with that of a spray-formed counterpart (on the right). Both are made of the alloy Cu-Cr0.8%-Zr0.1%. The diameter is just the same. While on the left the typical cast structure is visible, it totally disappears on the right. The average grain size is about 60 μ m.

Microstructures and segregation are also clearly influenced by the process. Figure 7 compares the microstructures of permanent mould cast and spray-formed Cu-Sn16%. The permanent mould cast is of dendritic structure. Between the dendrites the tin-rich δ -phase is en-

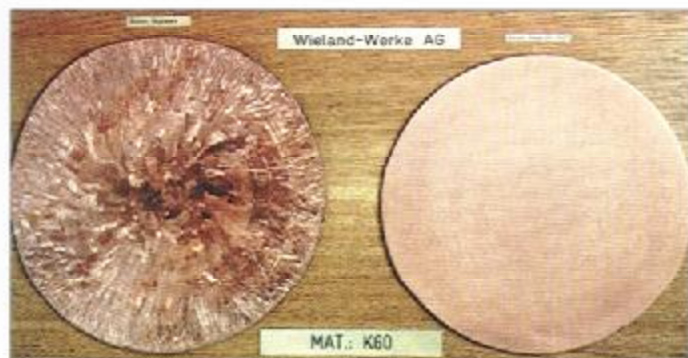
Fig. 6: Macrostructure of continuously cast (left) and spray-formed (right) billet, \varnothing 250 mm, CuCrZr

Fig. 7: Microstructure of permanent mould cast (left) and spray-formed (right) CuSn16

Table 1: Technical data of the Wieland spray forming unit

Semi-finished product	Billet
d_{min}	160 mm
d_{max}	500 mm
l_{max}	2200 mm
m_{max}	2400 kg
$input_{max}$	3200 kg
production rate, single-atomizer	35 kg/min
production rate, twin-atomizer	70 kg/min
overspray	10 to 40 %
additional equipment	Particle injector

riched. The high fraction of this brittle, low melting phase prevents cold and hot forming. In the spray-formed structure (on the right) the fraction of δ -phase is much lower and not connected. This structure allows cold and hot forming. While prematerial made of conventional – permanent mould or continuous – cast is formable only up to 8 % Sn, spray-formed material can be hot and cold formed by extrusion and drawing up to 16 % Sn without any prior homogenization. The classical border between wrought and cast alloys is shifted to considerably higher tin contents.

With Sn-contents of up to approx. 14 %, no δ -phase is observed in the spray-formed structure at all. In the solidified droplet a relatively high fraction of a second phase is found, as shown by the section of an overspray particle in Figure 8, on the left, which can be identified by chemical analysis to be β -phase. During slow cooling of the billet this β -phase converts to δ -phase and disappears totally due to the thermal equilibrium (Figure 8, on the right). Interesting is the twin accommodation in the sprayed structure indicating a deformation of the already solidified fraction in the droplet when hitting the substrate and subsequent recrystallization.

But also macroscopic segregation, known as inverse segregation, can be reduced to a minimum. On continuously cast bronze billets, differences in concentration between core and surface of up to 2 % will be measured. If the outermost solidified shell is included, these differences are even 9 %, as shown in Figure 9 [4]. Also during spray forming, this type of segregation can be observed. However, the differences in concentration are much smaller. If the gas flow rate G chosen is unfavourable, the difference is < 1 % (see Figure 10, lower curve). With a suitable gas flow rate, the difference can be reduced to < 0.5 % (see

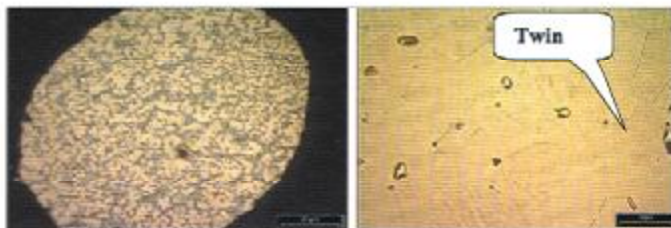
Fig. 8: Section of a solidified CuSn13.5-droplet with β -phase (left) and fine grained homogeneous structure in the billet (right)

Figure 10, upper curve). It is to be considered in this context that the billet diameter in Figure 9 is only 7" and that the alloy contains 8 % Sn, whereas Figure 10 is based on a 12" billet with nominal 12 % Sn. The influence of the process parameters has been described already [5, 6].

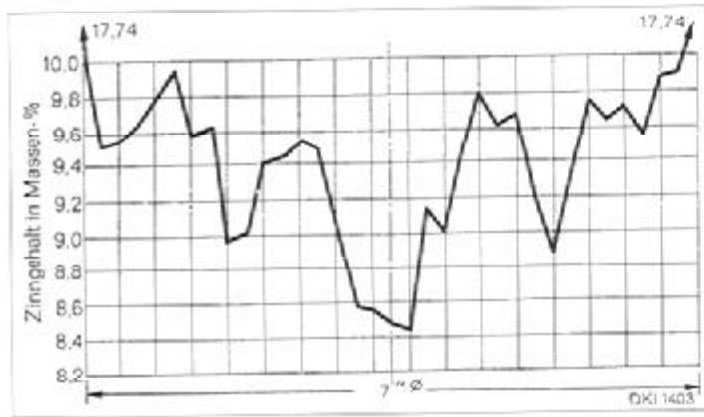


Fig. 9: Sn concentration across the section of a continuously cast CuSn8 billet [4]

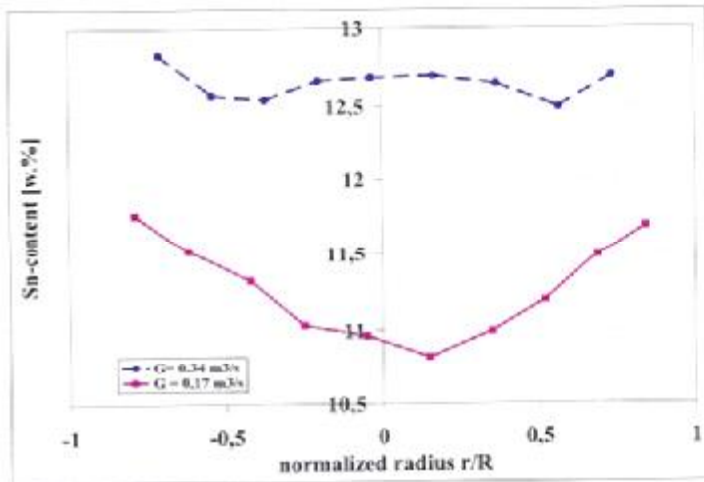


Fig. 10: Sn concentration across the section of a continuously cast CuSn12 billet spray-formed with different gas flow rates G at constant melt flow

2 Materials

2.1 Tin bronze

As mentioned already, the special advantage of the spray forming process is the production of high-alloy systems. In the phase diagram Figure 11, the areas for conventional casting and spray forming are marked. In the latter case, the limit of the homogeneous α -mixed crystal is exceeded. Nevertheless, the material can be formed by hot extruding and drawing due to the low segregation and the fine grained, homogeneous structure.

By adding Sn, the strength of copper is increased with simultaneous constant elongation. The curves for Sn contents of 0 % to 16 % run almost parallel (Figure 12). With increasing Sn content, cold formability decreases.

2.1.1 Application for sliding elements

High-alloy tin bronzes are also suitable as material for slide bearings and sliding elements. For slippers in hydrostatic drive hydraulics (Figure 13 and Figure 14) the de-

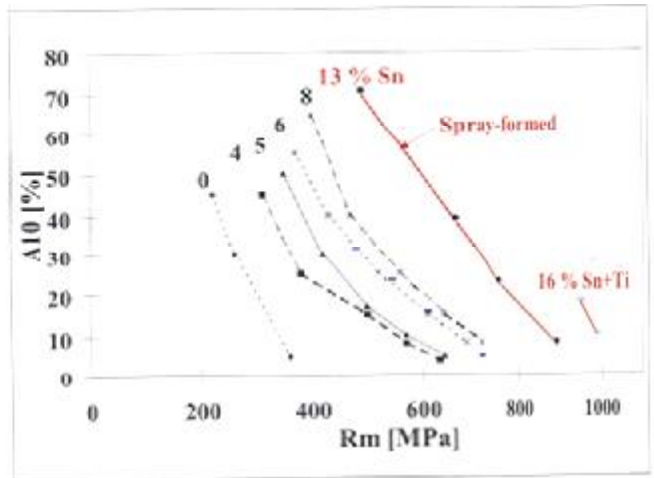


Fig. 12: Influence of Sn concentration on elongation A10 und tensile strength Rm

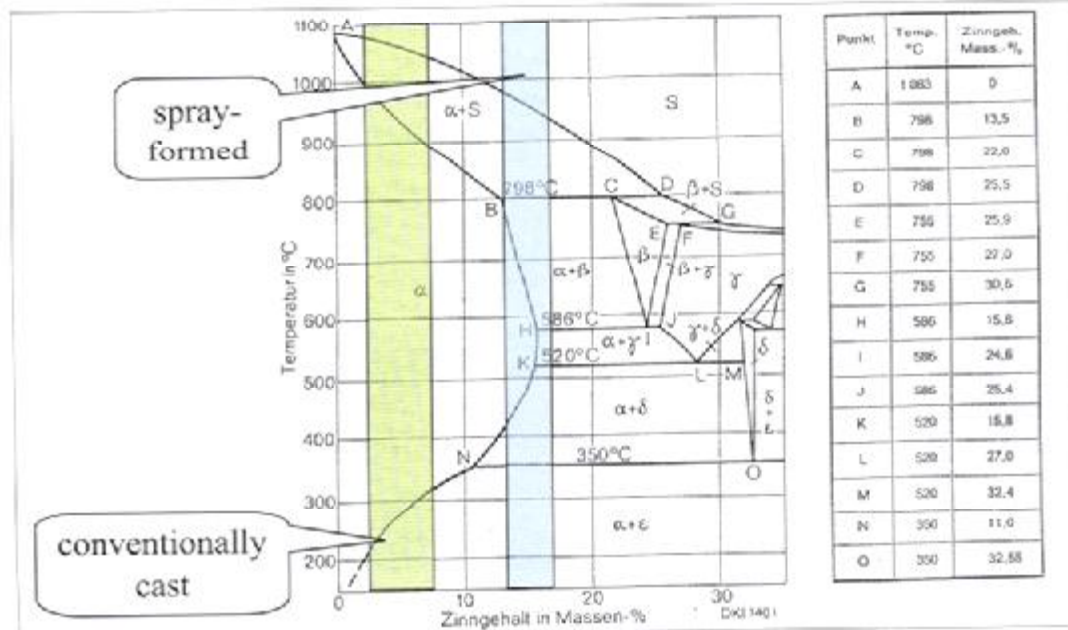


Fig. 11: Phase diagram Cu-Sn [4]



Fig. 13: Slipper and plunger for hydrostatic pump

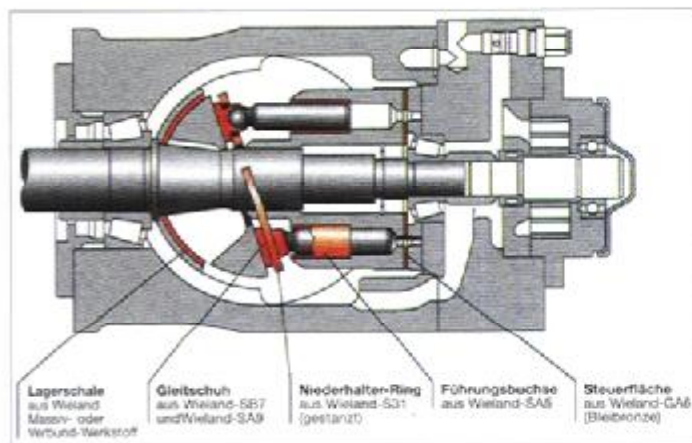


Fig. 14: Hydrostatic drive hydraulic

mand for higher strength with at least the same tribological properties as those of the standard material had to be met. A large number of tests has shown that e.g. thermal conductivity influences the emergency running properties of sliding elements [7]. In Figure 15, the results of bench tests on real drive hydraulics are compared for different materials. The tests were carried out in four steps. The conditions of these steps are plotted on the abscissa. The standard material for this application is conventionally produced CuZn37Mn2Ni2Si1Pb. It passed successfully the steps 1 to 3 of the test. The spray-formed bronze CuSn14.8Ti is the only material which passed step 4 of the test. So it is qualified for the application in slippers for hydrostatic drive hydraulics when the requirements to strength become higher.

2.1.2 Application for superconductive wire

The application of spray-formed high tin bronzes for superconducting wires is described in detail elsewhere [11]. The tin bronze is used as a carrier of tin for the final reaction with Nb to the superconducting Nb₃Sn phase. The production process can be split into six steps (Figure 16, next page):

1. Spray forming of bronze billet
2. Extrusion and cold drawing of bronze sheath tubes and hexagon tubes
3. Assembling bronze tubes and Nb-rods
4. Co-working of Niobium and bronze (sequences of extrusion, cold drawing, heat treatment)
5. Coiling of wire to final shape on the magnet
6. Formation of Nb₃Sn by final annealing process

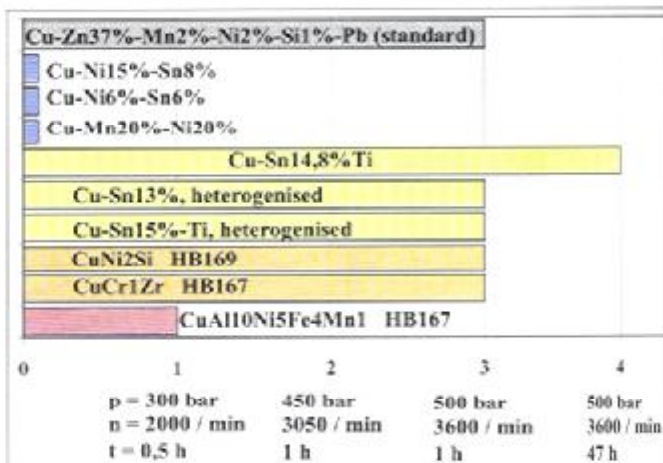


Fig. 15: Results of tests in hydrostatic drive hydraulics

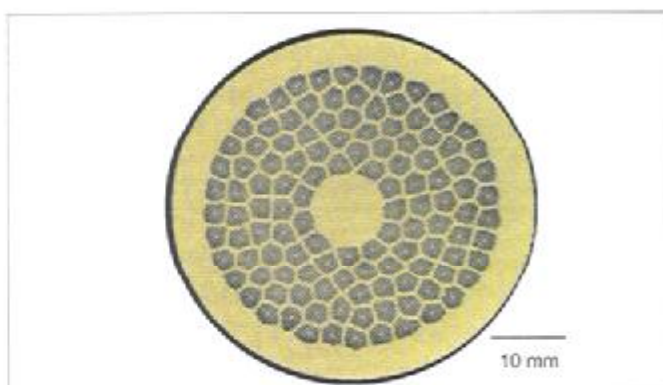


Fig. 17: Cross section of superconductor wire in production stage after hot extrusion. Bronze matrix contains $120 \times 84 = 10\,080$ Nb filaments, with courtesy to Vacuumschmelze GmbH Co KG, Hanau, Germany

During this process a multi-filamentary composite material is generated. Figure 17 shows a cross section through a non-stabilized type of superconductor in a production stage after extrusion and before cold-drawing of the niobium-bronze-composite (production step 4). At a first glance 120 hexagonal grey dots surrounded by bronze are visible. Each dot consists of 84 niobium filaments which also are surrounded by a bronze matrix. This matrix has been generated during the extrusion of 84 hexagon tubes of bronze (e.g. hexagonal dimension of 19 mm) filled with niobium rods (e.g. diameter of 9 mm).

The following production steps are numerous cold-drawing steps and heat treatments. The dimensions of niobium filaments and bronze channels finally reach the size of 5 to 20 μm .

2.1.3 Requirements to high-tin bronze semi-finished material

The manufacturers of super-conducting wire need a good workability of the tin-carrying bronze. By means of forming processes the tin is brought as near as possible to the niobium. The final heat treatment lets tin diffuse to the niobium and transfers both into the Nb₃Sn phase. Accordingly the manufacturers state two main requirements to the semi-finished bronze material:

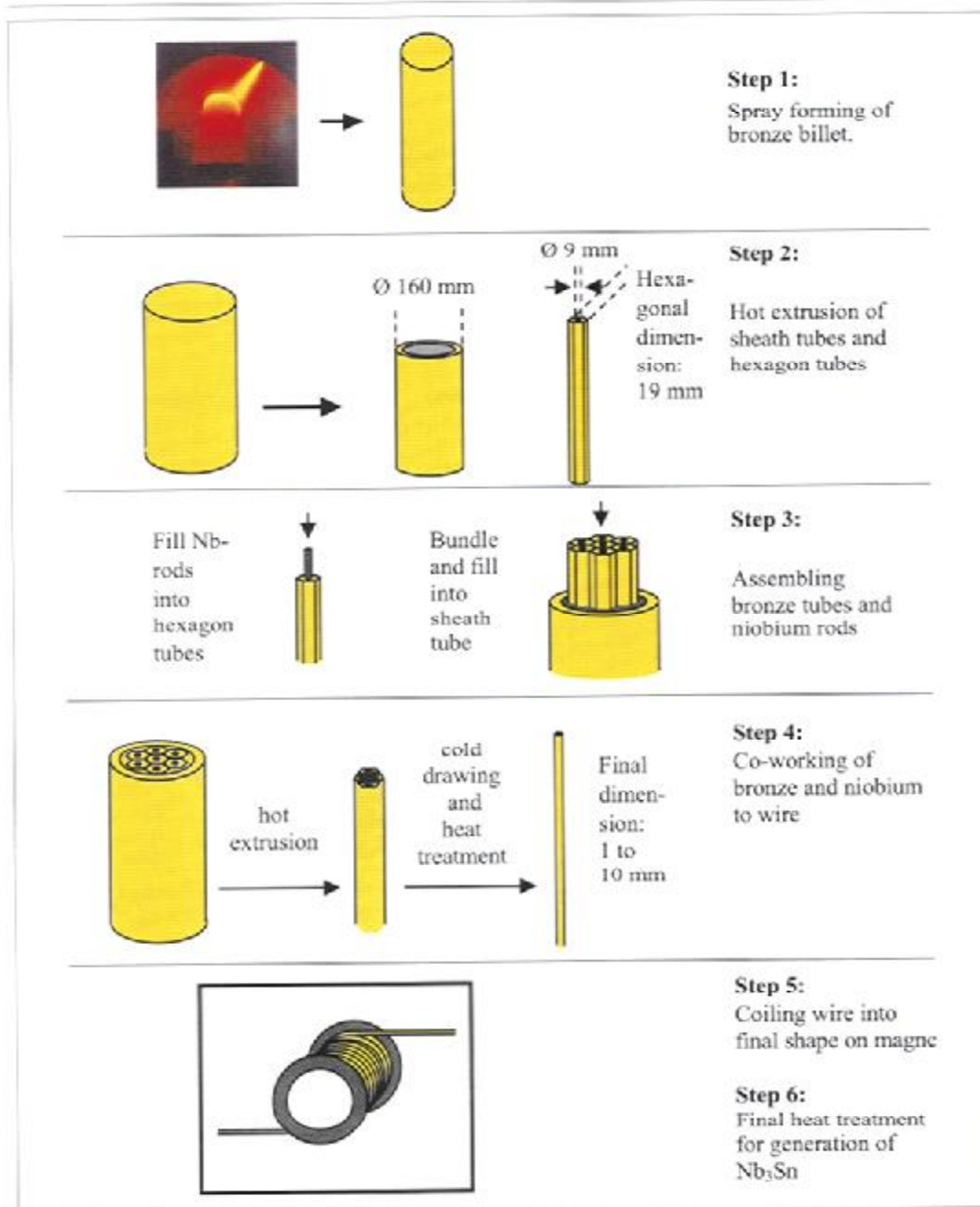


Fig. 16: Production process of bronze for super-conducting Nb₃Sn wire (scheme)

- Good hot and cold workability.
- Suitable high tin content (> 13 %) and homogeneous tin distribution.

2.1.4 Advantages of spray-formed high-tin bronze

The requirements to the material are a challenge to the material manufacturer. Bronzes produced by casting processes have a high tendency to grain growth, tin segregation and precipitation of the inter-metallic delta-phase, a copper-tin phase with 32 to 33 % tin. These two effects are detrimental to the further production processes of the super-conducting material. Cast bronzes therefore have to be time and cost intensively heat treated before being suitable to the production process.

Spray-formed high-tin bronzes have various advantages compared to cast high-tin bronzes. The grain structure of

spray-formed material is fine and homogeneous. Bronzes with tin content up to 13.5 % are single-phased. Those with tin content of more than 13.5 % have a homogeneous distribution of small delta-phase particles. Inverse segregation is minimized to 1 % tin concentration difference across the billets. All these advantages are beneficial to a good workability and to process stability as well as they help to simplify the production process, e.g. help to make initial heat treatment unnecessary. So they contribute to reduce production cost.

2.2 High-strength material CuMnNi alloy

For a number of applications, such as drilling components for oil exploration, antimagnetic copper base alloys with very high strength and low galling tendency are required. The system CuMnNi basically has the potential to meet these requirements. However, also in this case, production

with conventional casting methods is very difficult due to the high alloy contents. The phase diagram Figure 18 shows the range of an age-hardened alloy. The position of the alloy CuMn20Ni20 is marked with a blue dot.

Figure 19 compares CuMn20Ni20 in the A5-R_m diagram to beryllium copper and different steel grades. The entire

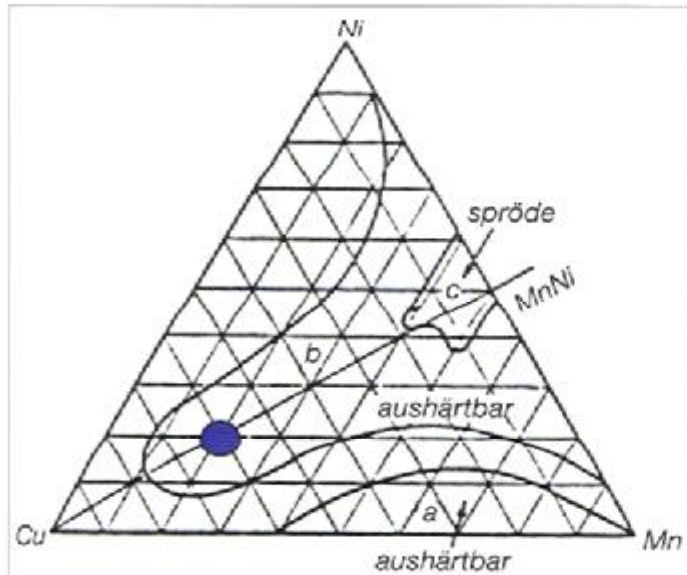


Fig. 18: Ternary phase diagram CuMnNi [8]

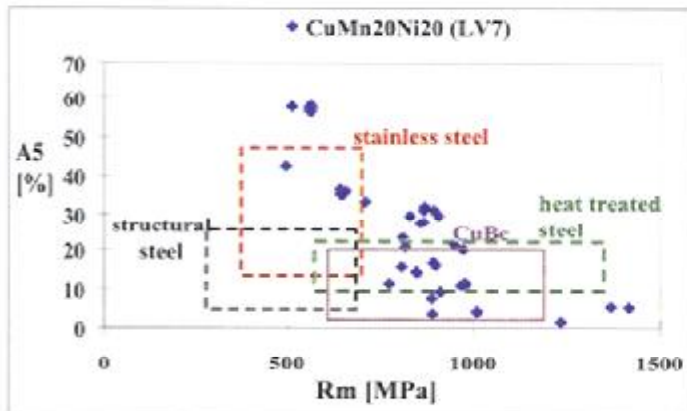


Fig. 19: Mechanical properties of CuMn20Ni20 compared with other high-strength materials

range can be covered by CuMn20Ni20. In Table 2 the standard values of the mechanical properties for three different tempers are listed. The material is able to fulfil the API specification for high-strength non-magnetic material for oil drilling equipment.

Table 2: Mechanical properties of the high-strength material CuMn20Ni20

Material	Rm [Mpa]	Rp0.2 [Mpa]	A5 [%]	HV
CuMn20Ni20, soft	540	200	35	120
CuMn20Ni20, age hardened	875 to 1075	760 to 980	13 to 30	240 to 310
CuMn20Ni20, 20 % cold worked + age hardened	< 1300	> 1050	> 5	350

2.3 Aluminium bronzes

Complex aluminium bronzes are characterized by high hardness and very good corrosion resistance [10]. The phase diagrams for Cu-Al and Cu-Al-Fe3 (Figure 20) [9] show that a large number of complex phases is to be expected particularly with the grades having a high aluminium content. The range preferred for spray forming is marked in blue.

The uniform hardness distribution shown in Figure 21 is a result of the process-related homogeneous distribution of

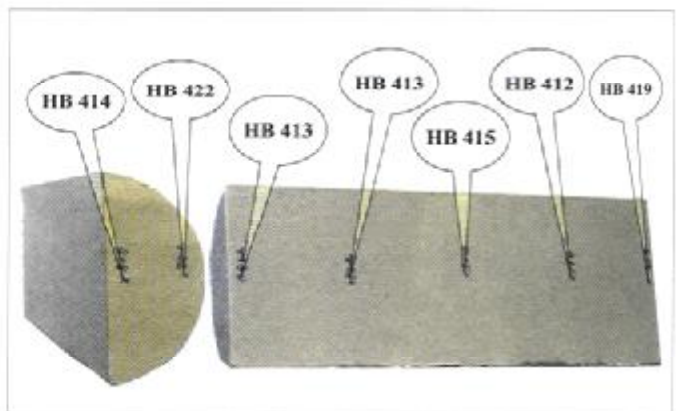


Fig. 21: Hardness distribution in a CuAl15Fe4.5CoMn rod, Ø 80 mm, spray-formed and extruded

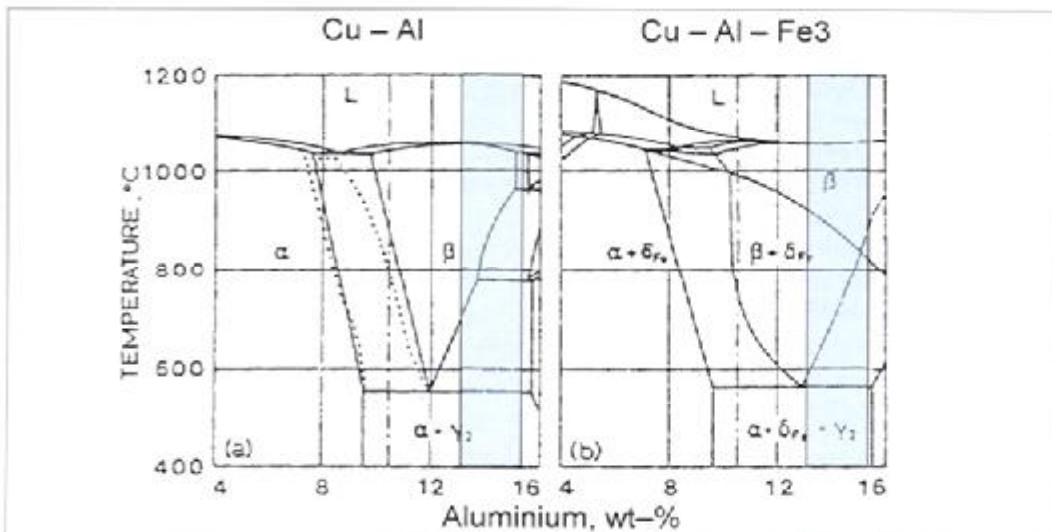


Fig. 20: Phase diagrams Cu-Al and Cu-Al-Fe3 [9]

Conventional
material:
rough chips



Material with
improved
machinability:
smooth chips



Fig. 22:
Al-bronze chips of conventional
and improved material at same
machining conditions

all alloying elements. The \varnothing 80 mm rod has been extruded from a spray-formed \varnothing 300 mm billet.

Main applications of this type of Al bronzes are forming and deep drawing tools for steel, especially stainless steel. The requirements are high pressure strength and high hardness. Controlled by the Al content the hardness can reach more than 400 HB and compressive strength is up to 1350 MPa. Usually these alloys are not easy to machine. New developed types have considerably improved machinability, which is realized in better surface quality, less tool costs due to reduced wear and in less roughness of the chips, because they break more easily during the machining process (Figure 22).

3 Conclusion

Spray forming is a fairly new process. Introduction into industrial production has reached a considerable level. The process offers interesting production possibilities on the one hand for materials already known, which cannot be cast or are difficult to cast, and on the other hand for innovative materials with special properties. So tin bronze up to 16 % Sn can be produced by spray forming exhibiting excellent ductility and high mechanical strength. Workability is excellent without the need for preceding homogenization.

Above about 14 % Sn, a brittle phase appears, which is much lower and finer distributed compared to permanent mould or continuously cast material and, therefore, allows good hot and cold working. The spray-formed tin bronze covers a field of applications in sliding elements and superconductive wire.

CuMn20Ni20 is a high-strength construction material with mechanical properties which can compete with those of beryllium copper and high-strength steels. This and the non-magnetic property introduced it in on- and off-shore industry for oil-drilling equipment.

Spray-formed complex aluminium bronzes are characterized by high hardness, high compressive strength and uniform distribution of the properties in the workpiece. Additionally the machining properties are improved considerably. So this type of material now is widely applied in forming and deep drawing tools for steel and stainless steel.

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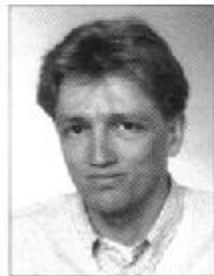
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Dr. Robert Zauter studied materials sciences at the technical faculty of the University Erlangen from 1982 to 1988, following which he worked as scientific assistant at the same institute. He graduated in 1992 to work in industry from 1993, namely for seven years as senior materials engineer in power plant and flue gas desulfurizing design. He then worked for one and a half years for a medium-sized material producer in the technical marketing of special materials. Since 2001, the author has been working for Wieland-Werke AG and is now engaged in developing and marketing spray-formed copper alloys.

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