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Numerical study and experimental investigation of solidification phenomena in semi-continuous casting of Bronze

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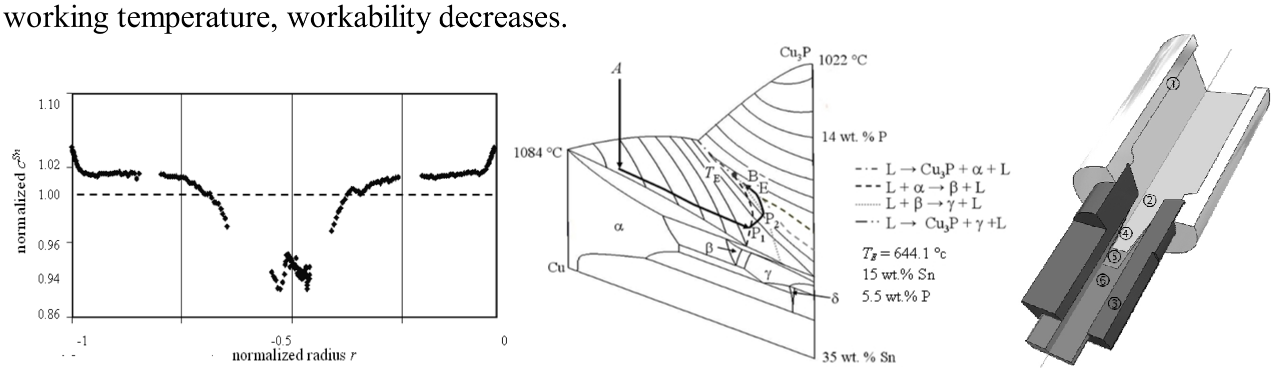
Abstract. In semi condinuous casting of technincal bronze alloys homogenuous microstructure is very important for the assurance of material properties. The improvement of the knowledge about both, thermodynamics of the ternary system Cu-Sn-P and the solidification process is of main interest for the involved industry. To describe solidification of these alloys, the ternary system Cu-Sn-P in the Cu-rich corner is experimentally investigated. DSC measurements, diffusion and annealing experiments have been performed and compared with computational thermodynamics based on the Calphad approach. The so defined thermodynamic information is coupled with solidification simulation. For this, CFD calculations are done with a two phase solidification model including mass, momentum, energy and concentration transfer and applied to semi-continuous casting of technical Bronze alloys. The predicted macrosegregation pattern is in good qualitative agreement with experimentally observed result.

Introduction

Bronzes, especially phosphor bronzes, are among the oldest metals produced. Nowadays these alloys have to provide even higher qualities for improved applications in for example electronics. Based on the fact, that these bronzes have a rather large mushy zone (we talk of about 200 °C of solidification interval dependent on the alloy composition [1,2]) macrosegregation often appears as displayed in Fig.1a [3]. This can be observed after casting even in wrought alloys with a tin content from 4 to 8 wt.% Sn. Since the tin and phosphor rich phases are brittle at room temperature as well as at hot working temperature, workability decreases.



(a) (b) (c)

Fig. 1 (a) Normalized macrosegregation profile of Sn for a round bloom (CuSn7.6P0.022 alloy). Positive macrosegregation is obtained at the surface and negative one at the centre [1]. (b) 3D liquidus surface of the ternary phase diagram Cu-Sn-P in the Cu rich corner [4]. (c) Geometry configuration: ① graphite tundish, ② position of the inlet for the calculation, ③ graphite mold, ④ liquid region, ⑤ mushy zone and ⑥ solidified strand.

Macrosegregation is known as the inhomogenuous solute distribution observed at the scale of a casting. It is caused by a combination of microsegregation and relative motion between liquid and solid. The relative motion in the mushy zone is thought to be induced by different flow phenomena, for example, forced convection, thermosolutal convection or feeding flow. To describe solidification of these alloys, the ternary system Cu-Sn-P in the Cu rich corner (Fig.1b shows the liquidus surface) is experimentally investigated by DSC (differential scanning calorimetry) measurements and annealing experiments [4,5]. The experimental results are compared with computational thermodynamics and literature. The validated thermodynamic information is included in CFD calculations for solidification simulation based on the multiphase model published by the authors [6-9] and applied to Bronze semi-continuous casting.

Performance

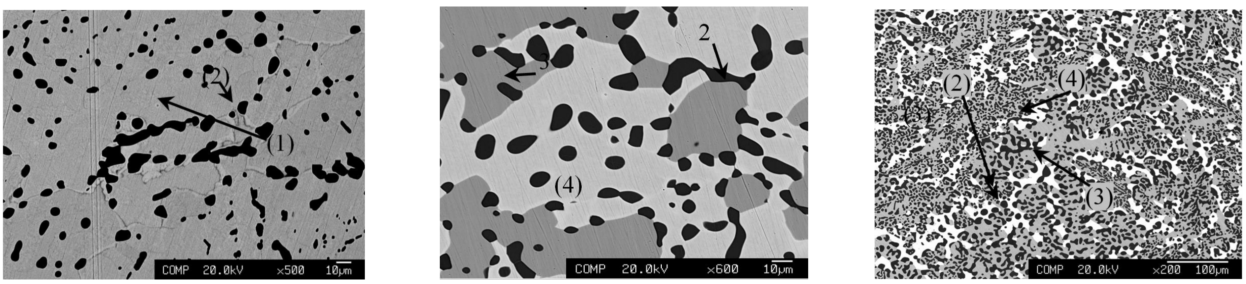
Brief Description of the Benchmark and Numerical Model. The benchmark problem is shown in Fig.1c. Details about the boundary and the initial conditions as well as the applied material properties for the simulation are given in [10]. Here, the main assumptions are mentioned: (i) A two-phase (liquid and solid phase) solidification model [6-9] is applied. (ii) Ternary thermodynamics of the system Cu-Sn-P is included by coupling computational thermodynamics performed with Thermo-Calc with solidification simulation [11]. (iii) Solidification shrinkage is included by applying different densities for liquid and solid. Thermosolutal convection as well as nucleation of grains is ignored. The grid size for calculation is 1.2 mm in the strand and 5 mm in the mold.

Experimental Performance. In the experimental investigations DSC, annealing and diffusion experiments have been performed to define specific phase regions, e.g., γ phase. The region around the ternary eutectic point (T_E, Fig. 1b) was investigated by diffusion experiments with the sample partners Cu and CuSn20P6 (in wt.%). The experimental description is given in [12,13].

Computational Thermodynamics. The ternary system was assessed by [14] considering literature data on the binary system and the experimental study on the ternary system [15]. Based on the phase diagram Cu-Sn the γ phase is included in the numerical assessment although it was not observed in the experimental study of [15]. The ternary eutectic point (T_E, Fig.1b) is still under discussion as mentioned by [16]. The shown thermodynamic data is calculated with the commercial software Thermo-Calc which is based on the Calphad approach using the database CuSnII.

Results and Discussion

Experimental Results. The experimental investigation includes the study of phase distribution, concentrations and phase regions of binary and ternary samples. For that the microstructure and concentrations of both, unannealed and annealed samples has been investigated with light microscope and SEM (scanning electron microscope). The phase distribution is on the one hand compared with thermodynamic phase diagram data (calculated with the database CuSnII) and on the other hand with literature [15, 16]. In Fig. 2 micrographs of ternary annealing samples are shown.



(a) CuSn30P1, 640 °C, 24 hours (b) CuSn17P2, 640 °C, 42 hours (c) CuSn11P5, 640 °C, 42 hours

Fig. 2 SEM pictures of ternary samples after annealing at 640 °C. (a) CuSn30P1 (nominal concentration) for 24 hours, (b) CuSn17P2 and (c) CuSn11P5 for 42 hours and following quenching. Phases labelled with (1) γ (gray), (2) Cu_3P (black), (3) α (dark gray) and (4) β (bright gray to white).

Based on the performed investigations, a two phase region of γ+Cu_3P is detected in sample CuSn30P1. Sample CuSn17P2 has a microstructure containing three phases, namely α, β and Cu_3P as well as the third sample CuSn11P5 but different volume fraction of the three appearing phases. Fig. 3a shows the ternary diffusion sample Cu and CuSn20P6 after annealing and quenching. Fig. 3b shows the isothermal section of the ternary system at 640 °C for comparison with the proposed thermodynamics. The dark gray dots mark the concentrations detected in the annealing samples and the bright gray ones in the diffusion sample CuSn20P6 (Fig. 3a). Fig. 3c shows the same isothermal section indicating the observed concentrations along a sample cross section from the outside towards the center (see Fig. 3a, (i) to (iv)). Comparing the detected phase distributions with the proposed ones by computational thermodynamics and literature, it can be stated that good agreement is observed. However, although the γ phase was not detected by the experimental work performed by [15], its presence in the ternary system is confirmed by this study. In addition the existence of the ternary eutectic point is also confirmed by the detected three phase region in the range of interest. Therefore the database CuSnII is applicable for solidification simulation.

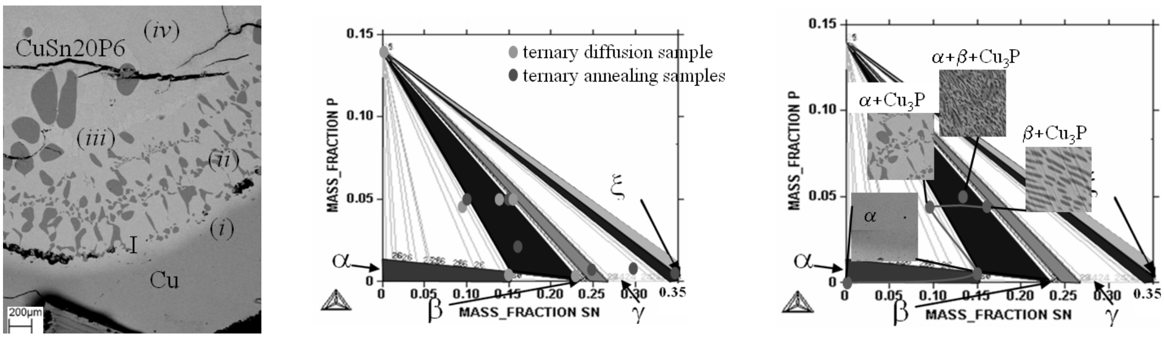


Fig. 3 (a) Ternary diffusion sample CuSn20P6 and Cu after annealing at 648 °C and quenching. (*i*) diffusion depth of Sn into the Cu sample (α see also in c); (*ii*) two phase region α + Cu_3P; (*iii*) two phase region β + Cu_3P; (*iv*) three phase region α + β + Cu_3P. I marks the interphase between the diffusion partners. (b) Detected concentrations of annealing samples (dark gray dots) and diffusion samples (bright gray dots) on isothermal section of 640 °C. (c) Detected phase distribution in the ternary diffusion sample.

Simulation Results. The ternary thermodynamics is coupled to solidification simulation [11]. Fig. 4a shows the predicted liquid concentration of Sn (c_ℓ^{Sn}) as a function of volume fraction liquid (f_ℓ) compared with ternary Scheil calculation done with Thermo-Calc. The displayed lines are taken in casting direction along the symmetry plane. It can be seen that the simulation results are in good agreement with the Scheil curve. However, it is visible that the f_ℓ versus c_ℓ^{Sn} curves are slightly shifted compared to the ternary Scheil curve. One reason for that is that Scheil does not consider any influence of flow on the concentration distribution but the simulation takes into account feeding flow.

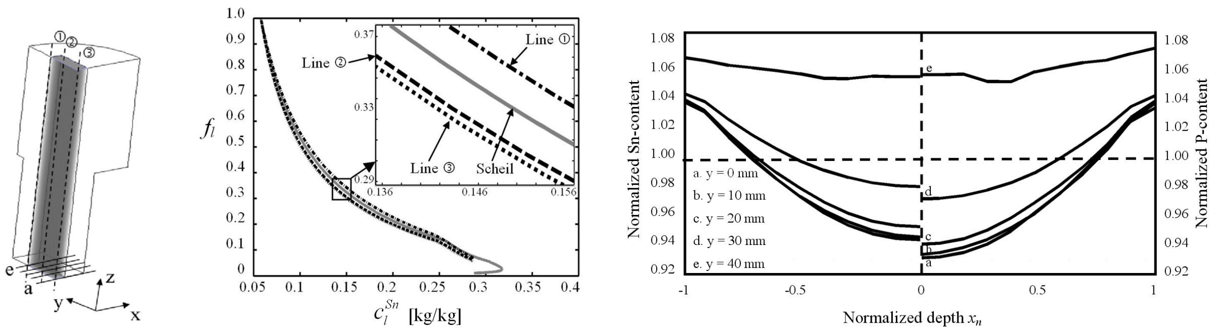


Fig. 4 (a) Scheil curve calculated with Thermo-Calc (gray line) for Sn compared to simulation results along three lines on the symmetry plane labelled in 3D geometry. (b) Calculated macrosegregation profiles of Sn (left) and P (right) along five lines at the outlet plane marked in 3D geometry.

Fig. 4a shows that there are differences in the concentration profiles depending on its position in the strand. This is caused by the fact that in the centre, where strong feeding flow is observed, c_ℓ^{Sn} is shifted to lower f_ℓ, whereas at the surface, where the solute is accumulated, c_ℓ^{Sn} is shifted to higher f_ℓ

Fig. 4b shows macrosegregation of Sn and P profiles taken along lines at the outlet plane. The predicted macrosegregation distribution shows the same tendency as observed in the experimental work (Fig. 1a) namely, positive macrosegregation occurring at the surface and negative one in the centre of the casting. At the moment the predicted values are qualitatively comparable with the experimental results. However, there are still deviations in the quantitative comparison. That is because just feeding flow was considered in the simulation but thermo-solutal convection was ignored up to now to allow the comparability with Scheil calculation. So the next steps are to include more complex flow phenomena and to adapt the simulation to specific process parameters.

Conclusions

In this paper, results of thermodynamic investigation, including DSC measurements, diffusion and annealing experiments and computational thermodynamics, coupled with solidification simulation for technical bronze alloys are presented. It can be stated that the recent experimental observations are in good agreement with the assessment work of Miettinen [14]. The thermodynamic information is used in multiphase solidification simulation taking microsegregation and relative velocity into account. Macrosegregation induced by feeding flow is predicted as accumulation of solute at the surface and depletion in the centre of the casting. The predicted results show the same tendency as observed in the experimental work.

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