OPTIMIZATIONS IN CONTINUOUS CASTING OF CU BASED ALLOYS BY NUMERICAL SIMULATION

Robert PEZER [1], Stjepan KOŽUH [1], Ivan ANŽEL [2], Mirko GOJIĆ [1]

[1]*University of Zagreb Faculty of Metallurgy, Sisak, Croatia,*
rpezer@simet.hr, kozuh@simet.hr, gojic@simet.hr
[2]*University of Maribor Faculty of Mechanical Engineering, Maribor, Slovenia,*
ivan.anzel@um.si

Abstract

Experimental study and numerical model for the thermo-mechanical properties in continuous casting (CC) of Cu-Al alloys is presented. This is a prerequisite basis for further development of the Cu based shape memory alloys (SMA) with various alloying elements (like Ni or Mn). A coupled thermo-mechanical numerical simulation of the CC process is implemented and applied to the full non-equilibrium process conditions. In the experimental part, we have used simple yet effective vertical continuous casting system. For the quantitative comparison, we have implemented special temperature measurement system within graphite crystallizer that enables us to monitor temperature profile at several spots around solidification front in real time. The present analysis of the various process parameters effect on the solidification process includes: casting speed, thermal contact conductivity, liquid metal temperature and cooling system set up. For microstructural examination we have prepared samples from rods that were subsequently investigated by optical microscopy, scanning electron microscopy (SEM) and energy-dispersive X-ray spectroscopy (EDXS) analysis. Comparisons of the experimental results and numerical simulation have been carried out. We have developed a comprehensive numerical simulation model that quantitatively describes non-equilibrium time-dependent temperature profile, solid fraction and microstructure features in as cast state. Results from the coupled thermo-mechanical and microstructural simulations compare favorably with experimental data suggesting the casting speed as a key parameter in process optimization.

Keywords: Material processing, continuous casting, solidification, thermo-mechanical, multiphysics

1. INTRODUCTION

One of the continuous efforts by the scientific community is to effectively transfer the basic knowledge from research results into practical industrial applications. Metallurgy and more specifically physical metallurgy is such a particularly successful story that made huge impact on our society over the last hundred years. Before the fundamental understanding of the physical phenomena involved in metallurgical processes and real nature of matter it was tedious and rather slow improving science. One of the basic technologies is CC that is in the focus of present investigation. It is appropriate whenever we need to produce long lengths of bars with uniform cross-section and is usually shrinkage-free. Copper based alloys are one of the main materials in many practical applications and among them aluminum bronzes (AB) [1] are best known for their high corrosion/oxidation resistance with high thermal and electrical conductivity. One further direction in the development of the Cu-Al systems applications is design and production of the SMA-s where in addition to copper and aluminum, third alloying element (like nickel or manganese) is usually introduced [2]. Despite the long history of usage, there are many open questions in CC technologies on the right values of the key process parameters for target as cast microstructure. There has been substantial progress and many refinements of the technology but mostly in steel production [3]. In the context of copper based alloys one of the recent developments is Ohno continuous casting technique [4] that can be effectively used for AB production. In fact,

by the very nature of the CC process, it is difficult to get quantitative information about what is going on in the mold so physical models became the strong alternative to direct characterization [3].

Today we are equipped with many advanced and highly precise physical models that utilize readily available computing power giving us completely new opportunities to study and improve our technologies. The basic fact is that macroscopic properties of the material is determined by underlying microstructure resulting from thermodynamics rarely close to equilibrium but more often than not the ones selected by relevant kinetics. One of the very successful approaches in predicting the microstructure evolution during solidification is phase field (PF) method and more specifically multicomponent alloy solidification version of it. Relatively comprehensive survey of the topic can be found in [5] and [6]. Cellular Automaton (CA) - Finite Element (FE) or CAFE model is another very successful simulation approach for the prediction of dendritic grain structures in solidification process [7] which is readily interfaced with standard FE codes like ProCAST and used for the same purpose as PF in this work.

Figure 1 Continuous casting system. (Left) Induction furnace filled with pure Cu and Al rods prepared for melting. (Right) The exit of the as cast rod from the crystallizer.

In present work, we have performed CC of the Cu-9Al system on our LAB equipment (similar to Delta Encon process). Straightforward characterization of the material grain structure and chemical composition is revealed by standard optical microscopy, SEM microscopy, and EDXS examination. We have developed two complementary simulation systems. First one is macroscopic scale simulation of the heat transport by conduction and convection that dictates the cooling of the alloy implemented in the industry FE code ProCAST. Second is our own PF implementation that features a simulation of the dendritic grain growth of the primary α phase into the undercooled melt. The model includes orientation dynamics following nucleation described by an additional order parameter. This code acquires local thermal gradient information from macroscale FE calculation. In the last section, PF simulation results are shown and compared to the experimental results. In concluding section we summarize the results and outline possible improvements of the present model.

2. EXPERIMENTAL PROCEDURE

The CC experimental setup in our LAB is shown on **Figure 1**. Cu-9Al melt was prepared using inert atmosphere at 1250ºC, which is well above liquidus temperature, from a commercial pure aluminum (99.9%) and copper (99.99%). The casting speed profile is a critical process parameter for ensuring CC product quality. In our case it is essentially regular sequence of 5 mm pulls with 0.5 - 0.7 s waiting intervals resulting in average casting speed of 260 mm/min. Water cooling system was set at 10 l/min (scheme of the system is shown on **Figure 3**). In the experiment and in the simulation we let system achieve stationary state so that temperature

field is constant on the time average. Reason is to avoid transient effects at the beginning since we cannot control this part of the process accurately. By CC we have obtained several meters long 8 mm in diameter rod with characteristic surface texture reviling pulls of the transport system.

Material thermal properties: liquidus is estimated from equilibrium thermodynamic database value at T_L = 1064 °C and solidus T_S = 1031 °C, latent heat 233 kJ/kg and thermal conductivity, density, specific heat are all given as functions of temperature as given by standard databases including one within ProCAST.

As cast material rod is cut, polished and etched in order to reveal grain structure by optical and SEM microscopy. **Figure 2** shows a typical optical (left) and SEM micrographs (right) with well-established microstructure in the present system that is a mixture of α and β phases [8]. Since the driving force in this solidification process is thermal activation in high-temperature gradients α phase dominates even α phase precipitation might be suppressed since the β phase transforms to the α phase as cooling goes on.

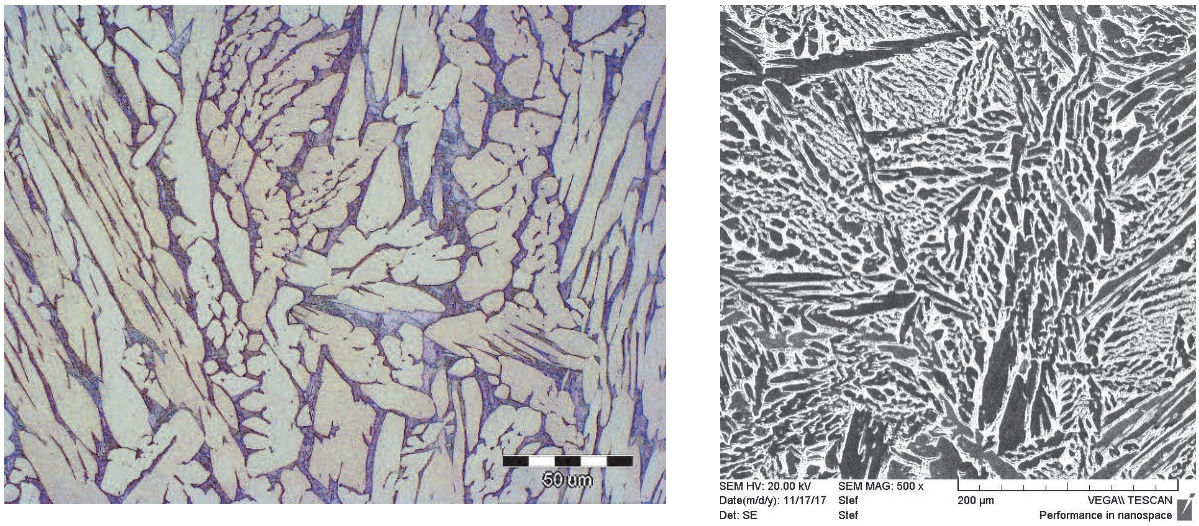

Figure 2 Micrographs with 500X magnification in the middle of the as cast rod: (Left) Optical microscopy. (Right) SEM image approximately at the same position in the sample as in the optical case.

EDXS reveals homogenous chemical composition along the cross section giving results in the range 6.66-7.58% of aluminum on the spots examined (near the edge and in the middle of the rod). This is in accordance with the observation that with high cooling rates β-phase transforms to the α-phase with fixed chemical composition, changing only the crystal structure.

3. PHYSICAL MODELING

The first part of the physical modeling consists of macroscopic thermo-mechanical simulation performed in ProCAST FE casting software package with CC module activation. Solidification simulation strategy is based on the fundamental observation that macro heat and mass transport processes govern mesoscopic microstructure evolution process, by determining a number of key conditions like local thermal gradients. Data obtained in this simulation were exported and imposed as thermal conditions for the micro system. We have calibrated uncertain parameters of the process (mainly heat transfer coefficients among different materials) in order to faithfully represent thermal gradients in the CC system compared to the measured data. Positions of the thermometers in the experimental set up are depicted on **Figure 3** (left). **Figure 3** shows main simulation results on the macro scale: on the left image we give temperature field after thermal stabilization (40 s from the start of the simulation) representing stationary temperature distribution. The right image on **Figure 3** gives solid fraction distribution (at the same time) with characteristic meniscus shape of the solid-liquid interface that

is very heart of the CC solidification process. In the subsequent micro simulation, we will numerically follow the tiny piece of the alloy in the middle of the mold where very high heat fluxes are present giving rise to steep gradients of the relevant quantities. One of the main simulation findings is that, in contrast to the typical steel CC process, we clearly see that the main thermal transport route is down the rod itself due to the very high alloy thermal conductivity. It calls for special care in the experiment as cutting the rod as it goes out of crystallizer reduce total heat capacity of the rod thus effecting casting conditions significantly. The main control parameter at hand to process adjustment is the casting speed that is the same as in experimental set up (on average 260 mm/min, with characteristic pulls and waiting intervals). Waiting time increase is a consequence of the graphite crystallizer heating up during the process since in CC process heat and material constantly enter the system from the crucible.

This simulation gives us full information about the thermal environment for the microstructure evolution. During the simulation residual stress is also calculated (not shown in this report) that can be used to further optimize process parameters. Effective residual stress after initial thermal stabilization is below 100 MPa throughout the rod. To continue further with simulation we define 100x100 μm 2D model box that we are going to follow from the state of the complete melt to solid grain structure that arises from the dendritic growth of the primary phase. We are going to perform simulation in the reference frame of the moving cast rod looking at the small quadratic piece that is on the symmetry axis passing through the top of the solidification meniscus. Already calculated temperature and heat flux fields in FE calculation provide changing the thermal environment for our micro subsystem.

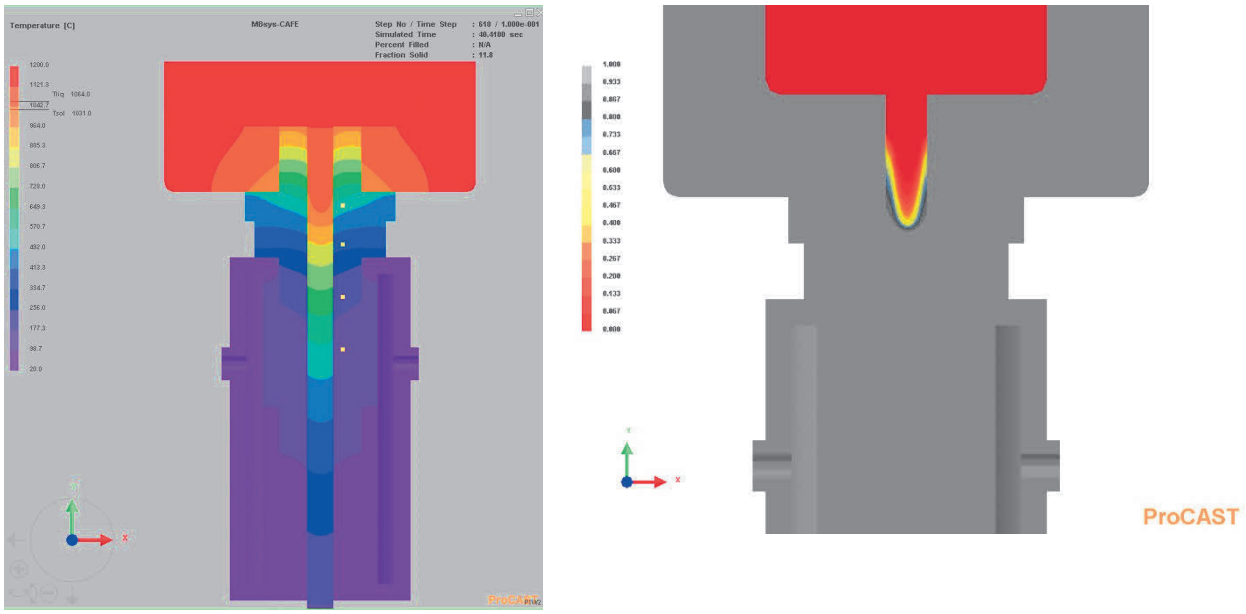

Figure 3 Stationary state in the CC process. (Left) Temperature field with thermometers positions within graphite crystallizer (shown as yellow squares). (Right) Solid fraction at the same time.

As for grain growth inside the model box we choose 2D PF model that is based on liquid and solid (α) phase but also include the possibility of different grain growth orientation [10]. There is a great deal of physical information from the full 3D simulation which can be studied in a 2D case but with considerably less computational cost. In this short paper we cannot give a proper account of the physical formalism involved in the phase field modeling of the nucleation and grain growth but refer the reader to the appropriate literature and references therein [5], [6] and [10]. It is based on the concept of order parameter that in this case consist of two 2D space-time function: one for the solid fraction and the other for grain orientation governing the impingement and misorientations of grains. We closely followed the formalism developed in [10].

PF formalism for the α phase order parameter governing equation gives:

$$\frac{1}{M_\phi}\frac{\partial \phi}{\partial t} = \nabla \cdot D\nabla\phi + \phi(1-\phi)m(\phi,\Delta T), \quad m(\phi,\Delta T) = \phi - \frac{1}{2} - \frac{\kappa_1}{\pi}\arctan(\kappa_2\,\Delta T) \tag{1}$$

The coefficient M_ϕ is solid liquid interfacial mobility and D is an anisotropic diffusion tensor in 2D defined with following auxiliary quantities:

$$D = \alpha^2(1+c\beta)\begin{pmatrix} 1+c\beta & -c\frac{\partial\beta}{\partial\psi} \\ c\frac{\partial\beta}{\partial\psi} & 1+c\beta \end{pmatrix}, \qquad \beta = \frac{1-\tan^2(\frac{N}{2}\psi)}{1+\tan^2(\frac{N}{2}\psi)}, \qquad \psi = \vartheta + \arctan(\frac{\partial\phi/\partial y}{\partial\phi/\partial x}) \tag{2}$$

where function $m(\phi, \Delta T)$ describe surface energy anisotropy with parameters $\kappa_{1,2}$. N is crystal symmetry (4 in our case). The governing equation for local crystallographic orientation field order parameter is given by:

$$\frac{P(\varepsilon|\nabla\vartheta|)}{M_\vartheta}\frac{\partial\vartheta}{\partial t} = \nabla\cdot\left[\phi^2\left(\frac{s}{\nabla\vartheta}+\varepsilon^2\right)\nabla\vartheta\right], \qquad P(x) = 1-\exp(-\beta x) - \frac{\mu}{\varepsilon}\exp(-\beta x) \tag{3}$$

where s and ε specify strength of the ϕ-θ coupling P being inverse mobility function. Third coupled differential equation is heat transport equation for the undercooling temperature (defined dimensionless as the difference from fusion temperature T_M) field:

$$\frac{\partial\Delta T}{\partial t} = \nabla\cdot D_T\nabla\Delta T + \frac{\partial\phi}{\partial t} + q(T_0 - T), \qquad \Delta T = (T - T_M)/T_M \tag{4}$$

where the last term is an additional source term that controls warming of the model box with q and T_0 adjusted to keep undercooling distribution realistic at the boundary. In this simulation, it is an effective heat sink that corresponds to net flux out of the model box. Namely, as solidification proceed undercooling is almost linearly increasing (temperature gradient is approximately 115 K/cm) on the macro scale as shown on **Figure 4** (left) but locally is severely influenced by massive latent heat production.

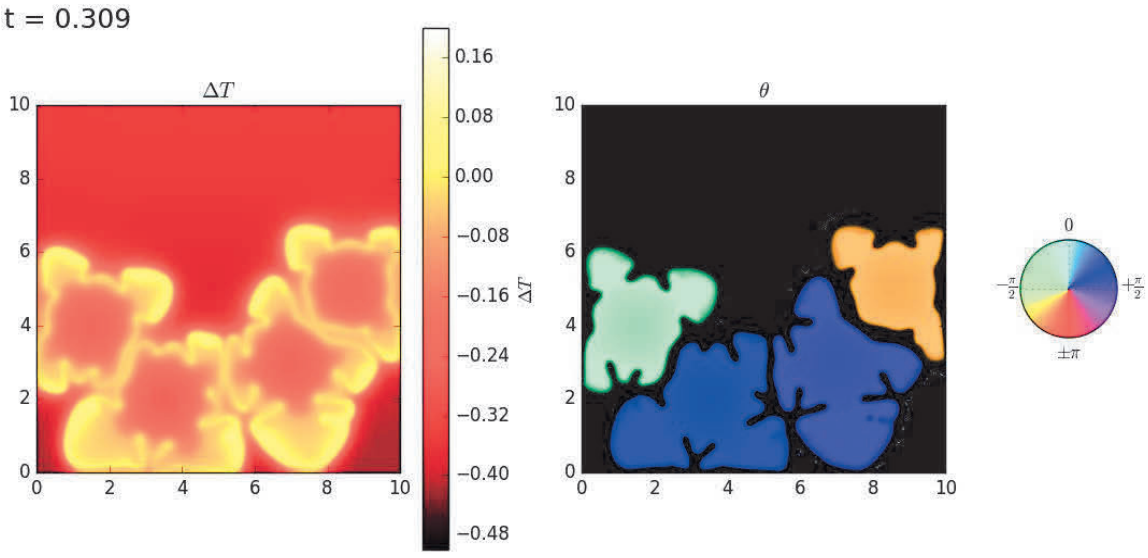

Figure 4 (Left) Undercooling distribution with the clear signature of the solidification latent heat local effects. (Right) typical dendrite structure of the solidification into an undercooled melt. Colors show the local crystallographic orientation of the dendrite with respect to the fixed axis of the simulation box.

Initial condition for the simulation is undercooled melt in accordance with the gradient obtained from FE macro calculation and seeding 4 grains with random orientation. The coupled system of equations is evolved by finite volume method with the help of implementation in FiPy numerical suite [11].

4. CONCLUSION

To summarize, in this paper we have described experimental CC of Cu-9Al binary alloy in rods. The as cast material was subsequently prepared for optical, SEM and EDXS characterization by standard procedure. Physical macro model of the CC system is prepared with the help of ProCAST software suite. We show main findings of the simulation that is calibrated by matching of the experimental and simulated temperature field in the graphite crystallizer. Besides macro temperature field, we also presented solid fraction field under stationary state during casting. Temperature field obtained in this step is used as an input for the undercooling field that constrains PF simulation of the single primary phase solidification. PF simulation (eq. 1-4) shows kinetics of the dendritic grain growth into the undercooled melt and is performed in the reference frame of the moving rod. Heating of the simulation model box is treated by including artificial heat sink into the thermal equation (4). As a further development of the microstructure model we plan to include composition field in the spirit of the Kim-Kim-Suzuki model [13] for interfacial energy and second phase order parameter for quantitative predictive physical simulation o the ternary SMA Cu-based alloys.

ACKNOWLEDGEMENTS

This work has been supported by Croatian Science Foundation under the project (IP-2014-09-3405).

REFERENCES

[1] MEIGH, Harry J. *Cast and Wrought Aluminium Bronzes: Properties, Processes and Structure*. IOM Communications, 2000. p. 47.

[2] GOJIĆ M., VRSALOVIĆ L., KOŽUH S., KNEISSL A., ANŽEL I., GUDIĆ S., KOSEC B. and KLIŠKIĆ M. Electrochemical and microstructural study of Cu-Al-Ni shape memory alloy. Journal of Alloys and Compounds. 2011. vol. 509, pp. 9782-9790.

[3] MILLS K. C., RAMIREZ-LOPEZ P., LEE P. D., SANTILLANA B., THOMAS B. G. and MORALES R. Looking into continuous casting mould. *Ironmaking & Steelmaking*. 2014. vol. 41, pp. 242-249.

[4] OKAYASUA Mitsuhiro, TAKASUA Satoshi and YOSHIEB Shigeki. Microstructure and material properties of an Al-Cu alloy provided by the Ohno continuous casting technique, *Journal of Materials Processing Technology*. 2010. vol. 210, pp. 1529-1535.

[5] STEINBACH, Ingo. Phase-field models in materials science. *Modelling and Simulation in Materials Science and Engineering*. 2009. vol. 17, no. 7, pp. 073001.

[6] NESTLER Britta, GARCKE, Harald and STINNER, Björn. Multicomponent alloy solidification: Phase-field modeling and simulations. *Physical Review* E. 2005. vol. 71, pp. 041609

[7] RAPPAZ M., GANDIN Ch.-A., DESBIOLLES J.-L. and THÉVOZ Ph. Prediction of grain structures in various solidification processes. *Met. Trans.* 1996. vol. 27A, pp. 695-705. GANDIN Ch.-A., DESBIOLLES J.-L., RAPPAZ M. and THÉVOZ Ph. A three-dimensional cellular automaton-finite element model for the prediction of solidification grain structures. Me*tal. Trans* A. 1999. vol. 30A, pp. 3153-3165.

[8] (PDF) Interactions between columnar.... Available from: https://www.researchgate.net/publication/50370483_Interactions_between_columnar_solidification_and_segregation_a_comparison_of_the_predictions_of_the_CAFE_model_with_in-situ_observations [accessed Jul 30 2018].

[9] CENOZ I. and GUTIERREZ M. Phase Transformations in Cu - Al Alloy. *Metal Science and Heat Treatment.* 2011. vol. 53, no. 5/6, pp. 265 - 269.

[10] ProCAST 2015.0 User's Guide, ESI Group.

[11] WARREN James A., KOBAYASHI Ryo, LOBKOVSKY Alexander E. and CARTER W. Craig. Extending phase field models of solidification to polycrystalline materials. *Acta Materialia.* 2003. vol. 51, no. 20, pp. 6035-6058; WARREN J. A. , PUSZTAI T., KÖRNYEI L. and GRÁNÁSY L. Phase field approach to heterogeneous crystal nucleation in alloys, *Physical Review* B. 2009. vol. 79, pp. 014204.

[12] GUYER J. E., WHEELER D. and WARREN J. A. FiPy: Partial differential equations with python. *Computing in Science & Engineering.* 2009. vol.11, no 3, pp. 6-15.

[13] OTSUKA K. and WAYMAN C. M. *Shape memory alloys*. Cambridge University Press, Cambridge, 1998, pp. 97-116.

[14] KIM Seong Gyoon, KIM Won Tae and SUZUKI Toshio. Phase-field model for binary alloys. *Physical Review* E. 1999. vol. 60, no. 6, pp. 7186-7197.