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MODELLING COPPER SMELTING - THE FLASH SMELTING PLANT, PROCESS AND EQUIPMENT

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Abstract

The effectivity of present copper smelting technologies have their roots in industrial and laboratory-scale experience accumulated over the past decades. Since early '60s, the tools for improving the processing conditions and smelting vessel design included scale modelling and manual computing of homogeneous multicomponent equilibria. The scale models were isothermal, room temperature constructions where water or air was used as medium and dimensionless numbers ensured scale down and scale up similarities. Today, numerical modelling has opened new insight into the high temperature process modelling where chemical reactions and their heat sinks and sources can be included in the simulations. The utilisation of computational thermodynamics enables a rigorous management of the phase equilibria in industrial multi-components slag-matte-metal systems. This development will be visualised in the framework of various enabling techniques.

Key words: scale modelling, computational fluid dynamics, computational thermodynamics, flow-sheet simulation

1. Introduction:

Copper industry uses several technologies compared to most other metals [1]. As a result of its predominantly sulphidic raw materials, severe harmful emissions to atmosphere and soil have been local and global problems.

In the 70 year history of the Flash Smelting process for sulphide concentrates [2], significant equipment development was needed in the completely new smelting concept, and specifically the novel smelting furnace (FSF) and its key equipment. Already from beginning, the very low use of external fuel and a superb sulphur fixation were evident. The need for a continuous development in the last decades was induced by a continuous furnace capacity increase, initially 24 kt/a copper and today >400 kt/a [3]. That has been met, e.g., by detailed physical and numerical modelling of the furnace, its external concentrate and off-gas handling devices, and the process conditions. The Flash Smelting technique was developed initially for the copper sulphide concentrates but adopted shortly thereafter for nickel, iron and lead sulphide concentrates [4-5] which brought additional dimensions to the process chemistry. Altogether 58 flash smelting furnaces have been built so far, including the flash converters (FCF) for continuous solid matte converting.

Improved recoveries and resource effectivity, increasingly complex and lean raw materials are today putting pressure to the control of the material streams. A stable furnace operation and high on-line availability are drivers in the smelting, enabling a high primary recovery of copper and the effective sulphur and impurity fixation, as well as optimisation of the industrial operation. These have been in focus of the continuous research and development of FSF, which from the early years included versatile physical simulation competences and since '90s numerical modelling both in the equipment and process chemistry development. The need of replacing time-consuming experimental work and to intensify the pace of development were the main drivers for the adoption of digital tools, as soon as the experimental data allowing that had been accumulated.

The scope of this review is to introduce the time-line of modelling and the industrial challenges the developers of a novel copper smelting technology faced, including a new approach of process chemistry, a specialised heat balance and its management, and the smelting vessel geometry. That is why the break-through experimental observations and modelling issues are introduced without a deeper analysis of their significance. This gives, however, a well-defined insight to the phenomena which are critical for understanding the coupled phenomena in the suspension smelting of sulphide minerals.

Drivers for the modelling

In most pyrometallurgical processes, the closed furnaces with thick refractory linings allow only limited sampling and direct, continuous observations and measurements are often impossible. These factors combined with hostile environments at high temperatures and aggressive gas atmospheres are drivers for applying modelling on various levels for advanced simulations. Modelling is a tool for linking the discrete experimental data points by a solid scientific framework, such as continuum mechanics or thermodynamics, for gaining deeper understanding of the phenomena, reaction paths and their mechanisms. Elevated temperatures together with high reaction rates and effective mixing have emphasised already in early stages the use of equilibrium thermodynamics in pyrometallurgy. In sulphide smelting, the heat balance of the furnace, in particular that of the reaction shaft, and the process chemistry must be regulated simultaneously, because the matte grade has a direct link to heat generation in the exothermic sulphide combustion as well as to fluxing of iron oxides.

The challenges for modelling copper making are variable raw materials often changing in custom smelters within a time span of days which should provide high metal value recoveries and allow high on-line availability. This requires constant adjustments of the feed, slag chemistry and fluxing, as well as active process control. Needs for on-line quality prediction of the products are also evident.

The scope may be an equipment or a furnace vessel in the smelting, converting and fire refining steps or the whole production chain on the industrial scale, starting from smelter concentrate storage and extending to operations of the tank house.

In the metallurgical industry, increase of the production volume is only occasionally accomplished by adding a next smelting line. A typical case is a throughput increase by new and more efficient auxiliary equipment related to concentrate handling and combustion, including advanced furnace cooling and process control systems [6]. In a long term, the need of increasing effectivity of the process equipment and achieving a higher on-line availability are targets typically tackled today with modelling and simulation.

An increased throughput with minimum CAPEX requires de-bottlenecking of the smelter and its combined refinery flow-sheet. It is benefitted from feed-forward and feed-backward information of the product streams as well as from the state and performances of the unit processes [7-8], such as off-gas train and its gas handling capacity, or solution purification of the tank house and its limits in, e.g., arsenic elimination from the electrolyte. Sometimes, this approach can be applied from ore mining to refining, and even linked with metal prices [9].

Most copper smelters of today run with essentially continuous smelting step and use batch-wise operating matte converting and blister copper fire-refining steps. The smooth linking a continuous (Flash Smelting) and batch (Peirce-Smith matte converting) steps at the smelter requires accurate scheduling of matte tapping and the materials transport operations because of a limited storage capacity for melts at the copper smelters [8].

2. Copper smelting basics

In early stages of the technology development, several pilot-scale flash smelting furnaces of different feed rates, typically between 0.1 and 1 mt/h, were built for detailed process chemistry and fluid flow purposes [2, 10-13], see Fig. 1.

Figure1. A mini-pilot FSF for examining the basic metallurgy of concentrates: sampling from settler [with permission by Outotec© 2019].

The need for completely understanding the performance and dynamics of reactors, process chains and production plants was a strong driving force for the modelling activities. Equilibrium properties of phase and reaction enthalpies form one ultimate boundary condition to reactor intensification and performance at elevated temperatures. For that purpose we also need continuous updating of the physical and chemical property data, as well as filling the gaps in the available values.

The overall smelting reaction for producing matte from chalcopyrite in a gas-solid suspension, in free fall oxidation in the reaction shaft, and settler reactions can be written in an approximate way as:

where ν ($\nu < 1$) is related to the copper grade of produced sulphide matte. Along with Eq. (1), side reactions occur for fluxing solid iron oxides at smelting temperatures for generating molten slag and matte, forming, e.g., ferric oxide and chemically dissolved copper in the slag.

The overall reaction (1) is not detailed enough for estimating the heat generation in the furnace, because we do not know the exact locations where the reactions take place. A fraction of the sulphides is over-oxidised and partially desulphurised in the reaction shaft consuming all free oxygen, but only to balance the under-oxidised coarse fraction for producing the desired matte grade in the subsequent settler reactions. It is, therefore, unknown which fraction of the reaction shaft reactions deliver to the overall process energy balance, according to Eq. (1), and the contribution of bath reactions. In a general case, the settler reactions may even be endothermic and to complete the smelting heat is needed. The combustion phenomena in the reaction shaft are effected by mineral composition of the feed, but also its particle size distribution. It determines the fraction of Eq. (1) taking place in the suspension, prior to completion of the matte and slag formation reactions in the settler [14]. An additional practical challenge is variable mineral composition of the sulphide ores and concentrates which causes variations in the heat generation and oxygen demand in smelting from one concentrate to another at the same matte grade.

Figure 2. A scale model experiment of a FSF concentrate burner at Outotec Research in early '80s leading to a new concentrate burner design [15] [with permission by Outotec © 2019; feed rate scale about 1:25].

Physical modelling in reactor performance design and optimisation has been used in the non-ferrous metallurgical field already for more than 50 years [16-17]. Various tracer assisted 2D and 3D scale models were used when developing the flash smelting concentrate burners, e.g., for introducing the single burner concept in the '70s and '80s, see Fig. 2. They were also commonly used for industrial troubleshooting when browsing, e.g., accretion formation and refractory wear problems. The development often required detailed data on industrial conditions, which could be gained with post mortem excavations after campaigns and by using laboratory scale experiments. In particular, sampling numerous post mortem studies are outside the public domain due to commercially

sensitive process data, e.g. metal recoveries and the behaviour of trace elements. There are, however, some sampling campaigns in industrial [18] as well as in pilot scale furnaces [11, 19-20] available in the literature.

As transport phenomena, especially gas-solid suspension fluid dynamics, form the backbone for the reactions of sulphidic concentrates in the FSF reaction shaft and in the continuous development of the furnace and accessory equipment, the role of two and three dimensional scale models using water or gas flows has been elemental [17].

3. Phase equilibria and smelting chemistry

The original invention of flash smelting was to use the furnace off-gas energy for preheating the process air [21]. The key problem of practical importance was the energy or heat balance of FSF, which is today controlled with the oxygen enrichment of the process gas and managed by accurate enthalpy balance calculations [22]. Once that has been satisfied in a continuous smelting operation, tapping slag and matte requires suitable fluidity for the slag and thus a careful and continuous control of fluxing of the feed mixture. The quality of sand is also of major importance, as typical minority minerals of sand containing alumina and alkalis enhance the surface energy of the slag, its viscosity and the melting point.

The necessary data for process calculations and simulations are typically the pure substance properties of the minerals, product oxides and sulphides, available in compilations. For proper heat effects and the phase boundary simulations, validated solution data(bases) for modelling smelting equilibria are essential. Pure substance data and the ideal solution approach are not accurate enough for forecasting locations of the primary phase fields and the single-phase boundaries of silicate slags and sulphide mattes. They also omit the heat effects involved in the mixing of molten substances when slag and matte are formed. To model the non-ideal interactions of complicated multicomponent solution phases, such as slags, mattes, solid oxide solutions, and alloys, the most powerful approach is the so-called Calphad methodology [23]. It is a thermodynamic assessment technique, where the solution properties for a specific phase are optimized using the appropriate solution model (e.g., associate model, quasichemical model, ionic sublattice model for slags), using all available experimental thermodynamic and phase equilibrium data, as well as *ab initio* calculation results and crystallographic input. The chemical systems are modelled hierarchically going from unary to binary and ternary chemical systems (e.g., Cu, Cu-S, Cu-Fe-S), and finally to high-order multicomponent systems (e.g., Co-Cu-Fe-Ni-S-O).

Selected databases for copper and other non-ferrous smelting processes available commercially have been reviewed recently by Jak et al. [25], including their current advancements in the FactSage simulation environment. Another long-term and industrially coordinated activity is in progress in various metal making and related fields, as summarised by Gisby et al. [24]. It provides a vast database for phase equilibrium and process simulations in the MTDATA environment [26].

Table I. Coverage of the systems assessed in the Mtox database, vers. 8.2 [24]

System definition (components)	Core applications	Features/comments
Na ₂ O-K ₂ O-CaO-MgO-Cu-Fe-O-Al ₂ O ₃ -SiO ₂ -S*	Base slag system	A complete assessment
Co-Cu-Cr-Fe-Ni-Pb-Zn-O-S	Sulphide mattes	A complete assessment
CaO-CoO-Cu-Cr-Fe-O-MgO-NiO-PbO-ZnO-Al ₂ O ₃ -SiO ₂	Zn, Pb, Ni slags	Missing some high-order (>3)
Li ₂ O-Na ₂ O-CaO-MgO-Fe-Mn-O-Al ₂ O ₃ -B ₂ O ₃ -SiO ₂	B ₂ O ₃ slags	Missing some high-order (>3)
CaO-Fe-Ti-O-MgO-Al ₂ O ₃ -SiO ₂ -ZrO ₂	TiO _x slags	Missing some ternaries
CaO-Fe-O-MgO-Al ₂ O ₃ -SiO ₂ -P ₂ O ₅	Phosphate slags	
Fe-Nb-O-Al ₂ O ₃ -SiO ₂ , Fe-Nb-O-MgO-Al ₂ O ₃ -TiO ₂ -Nb ₂ O ₅	Niobia slags	CaO to be added
Na ₂ O-K ₂ O-MgO-NiO-Al ₂ O ₃ -ZrO ₂ -V ₂ O ₅	Vanadia slags	To be completed with V ₂ O ₃
CaF ₂ , NaF and Na ₃ AlF ₆ with CaO-Fe-Mn-O-MgO-Al ₂ O ₃ -SiO ₂	Fluorine in slags	
As-O binaries with Cu-Fe-O-CaO-CoO-MgO-NiO-Al ₂ O ₃ -SiO ₂	Arsenic in slags	To be completed

Bi-Cu-Fe-O-CaO-MgO-Al ₂ O ₃ -SiO ₂	Bismuth in slags	Missing some ternaries
Trace elements: hydroxyl, carbonate, sulphate capacity	Various slag matrixes	

*trace element concentrations only for sulphur in slags

A short list of slag and matte systems available in the Mtox database was collected in Table I. The base dataset of Mtox is the system Na₂O-K₂O-CaO-MgO-Cu-Fe-O-Al₂O₃-SiO₂-S, which has been assessed completely up to the highest order system and covers its components up to the highest oxidation degrees of the cations [24]. Its sulphur data includes the concentrations typically dissolving in industrial silicate slags in smelting and refining operations. The sulphide matte sub-set Co-Cu-Cr-Fe-Ni-Pb-Zn-O-S contains dissolved oxygen at concentrations typical to matte smelting in the SO₂ bearing atmospheres of industrial interest. These two datasets can be used alone or combined together for the copper smelting and converting equilibria. In large systems with more than 8-10 components, they can be combined with the third dataset from top for more detailed equilibrium analyses. The data sets include mono- and divalent copper and also several other elements typical as traces in the industrial non-ferrous raw materials, such as e.g. alkalis potassia and soda, chromium and cobalt. As the assessments have been carried out from one pure oxide to another, the Mtox database can also be used for the nickel, lead and zinc smelting equilibria. The models and modelling issues of Mtox have been discussed earlier elsewhere [27].

Figure 3. A quasiternary liquidus contour plot of the Cu-Fe-O-K₂O-MgO-Al₂O₃-SiO₂ system at fixed oxygen partial pressure of $p(\text{O}_2)=10^{-7}$ atm, copper saturation and constant concentrations of $w(\text{K}_2\text{O})=0.015$ and $w(\text{MgO})=0.05$, thermodynamic data from the Mtox database [24]; w is mass fraction.

An example of capabilities of an advanced thermochemical solver in visualising the liquid phase domains is shown in Fig. 3, where the liquidus surface of an alumina-iron silicate slag with 5 wt% MgO and 1 wt% K₂O, saturated with metallic copper, under constant oxygen partial pressure of 0.01 Pa (10^{-7} atm) is depicted. The calculations were carried out using the MTDATA software, vers. 6.0, and the Mtox database, vers. 8.2.

The assumption of a complete thermodynamic equilibrium in a flash smelting or flash converting furnace is, however, a huge oversimplification. Due to the reaction sequences in suspension smelting, the gas-solid reactions are possible in the reaction shaft only, and the final matte and slag forming processes in the settler are far from the equilibrium with the process gas [14]. Their heat generation is also linked with the balance between reaction shaft and settler reactions. In this

respect, the two furnace sections in a FSF or FCF must be treated separately in detailed mass and heat balance calculations [28].

The use of computational phase diagrams with constraints, which degrade the multi-component industrial systems to quasiternaries, is the only rational way to understand and visualise the multidimensional molten slag spaces forming in smelting operations. In such cases, the artificial intelligence by a fluxing advisor [29] used for selecting the essential variables must secure robustness to the approach, in order to tolerate, e.g., the practical control and feed inaccuracies. The soft sensor tool may be useful in the copper smelters using raw materials with difficult gangue chemistry, such as high silica or alumina contents.

The use of secondary copper materials as feedstock in the smelting brings new challenges to the equilibrium modelling of slags, mattes and copper alloys. The assays of feedstock contain increasing but still trace concentrations of precious and platinum group metals, as well as metals not present in sulphidic copper concentrates, like indium and gallium. In order to cope with their distributions and deportments, new fundamental data are needed. Some early attempts to generate, e.g., distribution properties are, however, already available in the literature [30-33].

(4) Fluid flow simulations in copper smelting

The phenomena occurring in the reaction shaft of a suspension smelting furnace are very fast and difficult to observe on an industrial scale. Therefore, the numerical modelling is the only way to investigate, e.g., how different operating parameters and raw material mixtures affect transport phenomena, process dynamics and change the products of smelting. A realistic enough modelling for decision-making is only possible by taking all relevant phenomena into account, including the chemical reactions and their kinetics.

After introduction of the first FSF into industrial use in 1949, systematic development work started using scale models in Outokumpu Research Centre in late '50s and early '60s [17]. The driver was a constant demand on knowledge, more specifically answers to questions like 'What's wrong with our FSF?' or 'Can we add feed beyond the name plate capacity?' The physical modelling efforts were mainly focused on the flow pattern development for the fluids and equipment dimensioning in the FSF area, including waste heat boiler (WHB), electrostatic precipitator, and the concentrate burner. The results were then validated in pilot-scale trials [13], see Figs 1-2.

Modelling the smelting and converting have been developed from 1D fully mixed cross sections in '80s and '90s [e.g., 34-36] to more detailed 2D and 3D descriptions where local variations in the flow pattern and temperature history are taken into consideration [37]. This development was enabled by cheaper computing time, using computational grids and, e.g., momentum transfer for the fluids reacting in time-dependent dynamic systems. There local rate constants of chemical reactions were used [37] or equilibrium was assumed in each computational cell.

Reaction sequences in the suspension oxidation

The gas-solid reactions of concentrates in the reaction shaft have been extensively studied in the last decades by several university groups [e.g., 38-43], and in the industry [10, 43-50]. Most studies have been conducted in a laminar flow or drop-tube furnace. The materials studied included chalcopyrites and pyrites, several impure copper sulphide minerals as well as different nickel sulphides [51]. Furthermore, the development of the continuous matte converting technology triggered studies on copper and nickel mattes [52-54]. In addition, a few reports from industrial research campaigns have been published [e.g., 20, 55-56].

Single particle studies for combustion kinetics of sulphide minerals typically included oxidation degrees up to blister copper with a total desulphurisation. The key difference in the flash converting

reaction sequences compared to matte smelting is the chemically inert nature of ground matte [52-54] which sets requirements to its pre-treatment, ignition in the reaction shaft and the reaction shaft conditions.

The way to computational fluid dynamics (CFD)

An early attempt to study numerically the FSF process was presented by Ruottu [57], followed by other authors [58-60]. During the recent decades, mathematical models have been adopted, e.g., by [34, 61-69] for simulating the coupled phenomena in gas-solid suspensions, but also for supporting engineering issues, on the scale shown in Fig. 4 [70-72]. Also, detailed studies on thermal radiation have been conducted, [e.g., 73-74].

Figure 4. The 3D CFD model geometry for a combined FSF and WHB [78].

The conditions in the smelting are harsh and the refractory linings have to be protected by suitable engineering solutions to prevent wear and enable long campaigns. Currently, water-cooled copper elements are assembled between the refractory bricks and their operation as well as heat losses are monitored with advanced instrumentation and visualisation systems [75]. Mathematical modelling has also been used for predicting the freeze lining thickness, which is formed by cooling elements in the reaction shaft wall and elsewhere in the smelting vessel [76-77].

A completely new approach was recently taken with the coupled CFD–DEM modelling (discrete element method) by using the EDEM[®] software tool [79]. It has been developed for simulating discrete (solid) particles' interactions and behaviour in a fluid. It has now been tested and found suitable for modelling liquid matte droplet settling through a liquid slag layer [80]. The ultimate objective of these efforts was to digitalize fundamental observations from the experimental research and the industrial experience into an intelligent process model and optimization system [e.g., 81-82]. The close collaboration of the experimental and CFD-groups provided a solid basis for development of the necessary sub-models for describing various unit processes in the flash smelting domain. It is a long-term target, but the current research activities are paving the way towards it by developing realistic unit process models that can predict changes in the process outcomes when the feed mixture and operating parameters are altered.

The reaction shaft and concentrate reactions

The modelling of FSF using a commercial CFD software started in early '90s and a comprehensive review has been published recently [37]. The scope initially included detailed models of the FSF reaction shaft [83] and the WHB [84-85]. A more advanced approach with chemical reactions of

sulphide particles for both FSF and FCF were made public later [62-63, 86]. One of the first developers of FSF simulations by a commercial CFD code published a reaction shaft flow simulation in 1992 [64], and they included chemical reactions in the model in 2003 [87-88]. Adams et al. [89] modelled drop-tube experiments followed by combustion in 1999. In the industrial context, a CFD model of an independent burner development was published in 2006 [90] materialising in a new side blown burner in 2013 [91]. A CFD model for improved FSF burner performance has also been published [92], and Li & Xiao [93] used numerical simulation for browsing production enhancement. An early attempt of FSF simulation with a commercial code was provided already in 1994 by Seo & Rhee [94].

The nickel matte smelting FSF has been modelled and a full simulation was presented by Varnas et al. [95]. Doblin & Nguyen [96] investigated detailed concentrate burner gas flows. Other authors included an industrial flash smelting furnace gas space modelling using a commercial CFD software [e.g., 67]. In addition to commercial software, there have been many modelling efforts done by in-house fluid dynamics codes and tools [42-43, 97].

The modelling approaches include also minor element behaviour in the FSF and FCF [e.g., 98]. They developed a fundamental model based on thermodynamics and mass transfer in the suspension smelting environments. Another phenomenon was the particle-particle interaction or particle agglomeration in the suspension [99-100]. In a recent paper, Zaim & Mansouri [101] included a particle fragmentation scheme in the combustion simulation of a copper concentrate. They found out that particle fragmentation was remarkable in suspension and it should, therefore, be considered in the sulphide combustion modelling. In experimental concentrate and matte particle studies, fragmentation in the molten state due to vigorous oxidation reactions [42, 83, 102] has been identified to generate a wide droplet size distribution. There is, therefore, a need for a thorough review of this particle size issue (i.e., fragmentation vs. agglomeration) in the literature, both in laboratory and industrial scale results.

Perez-Tello et al. [102] developed mathematical correlations and a model for representing size distributions and dust formation from oxidized copper matte particles in FCF conditions, in a laboratory-scale pilot furnace. The dust formation in FCF was discussed [103-104] in connection with reaction kinetics of a chalcocite concentrate. A kinetic model for oxidation of chalcocite has been developed by [86]. Its utilisation in the direct-to-blister smelting for low-iron concentrates has been crucial.

The FSF settler

In the settler, slag-matte reactions, phase separation and the settling processes are important. Direct experimental data about the microscale processes on settler surface, leading to matte-slag formation and phase separation, are scarce [105-106]. As mentioned earlier, the suspension arriving in the settler is not homogeneous or isothermal, and we do not understand the details influencing these processes.

Some studies have been conducted related to effects of matte droplet diameter in the settling process and metal losses during slag tapping [105-106]. Yang et al. [107] modelled liquid droplet collisions and, consequently, improving settling in a slag cleaning process [108]. However, these studies focused on steady state conditions. Thus, the effects of the droplet size on settling rates remain to be studied in transient cases.

Xia et al. [71] studied a continuous matte and slag tapping system and the effect of matte droplet size on copper losses by entrainment. They suggest that copper losses are inversely proportional to the droplet size, i.e. decrease with increasing droplet size, and that position of the tapping hole and rate of tapping also affect mechanical copper losses. Slow tapping decreased losses and the slag tapping hole should be at least 100–150 mm above the slag-matte interface [71, 109]. Various other

phenomena, such as a higher turbulence at the inlet, matte-slag velocity distribution, and impingement of faster droplets on hitting the matte slag mixture in the settler, have been studied in recent years [106, 110]. The feed mixture falling from the reaction shaft is concentrated in the centre of its cross section area, instead of covering it uniformly. After fluid flow modelling, more challenging processes, as coalescence of matte droplets and chemical reactions between matte and slag, have to be included in the model.

Recently, a refined CFD modelling work [72] aimed at a settler model by user-defined-functions for the matte-slag reaction kinetics [e.g., 111]. The advanced modelling started from a transient two-phase flow of slag and matte layers where the feed material was landing on the slag surface. It was a realistic starting point for the liquid flow in the settler, which was modelled as a continuous tapping with a fixed flow rate in order to include the slag flow and matte entrapment. The second stage involves coupling of fluid dynamics and chemical reactions. One challenge in this approach was that single droplets cannot be used in the simulation due to very large computing resources needed, and because in CFD, discrete particle method (DPM), cannot model collisions between particles and particle agglomerates. They, therefore, must be estimated stochastically [112].

Coupling CFD and DEM methods attains growing interest in processing areas involving particulate solid materials in different applications, such as cyclones and blast furnaces [113-114]. A novel alternative approach for including chemical kinetics into fluid flow by a coupled CFD-DEM software has been made for testing feasibility of the CFD-DEM technique for describing droplets' settling through a slag layer [79-80, 115]. A physical modelling study for water droplet settling through an oil was used for validating the CFD-DEM model [80, 115]. In the simulation, fluid and particle flows were computed separately in CFD and DEM, respectively. First, fluid flow with drag forces were calculated in CFD and data was transferred to DEM, which calculated the particle movements. DEM time step is usually much smaller than that used in CFD.

The FLASH code for industrial sulphide smelting and converting

The experience gained in the industrial, pilot plant and laboratory studies on sulphide concentrate particles' combusting were compiled in '90s into a CFD tool extensively applied today for simulating various mineral and metallurgical processes, including, e.g., the flash smelting, flash converting [63, 70], and TSL smelting and refining furnaces [116]. The transport phenomena data of the FCF was based on experimental studies of copper concentrates and on the models developed for the FSF, whereas the models for chemical reactions, kinetics and particle fragmentation were obtained from experimental investigations of copper matte particles [53, 117] in copper matte-gas reactions using laboratory scale single-particle furnaces.

This in-house CFD tool for fluid flow, heat and mass transfer simulations in chemically reacting systems was incorporated into an advanced FLASH code [62, 64]. It is currently implemented in the commercial ANSYS Fluent software [112] for simulating complex heat and mass transfer processes, solids distributions, and chemical reactions of sulphide particles occurring in the reaction shaft of a FSF and FCF. By using the proprietary software package, complex material flows and heat transfer phenomena in various smelting furnaces, concentrate burners and in different operating conditions can be simulated and visualised. As an example, Fig. 5 compares particle mass densities (kg/m^3) in the reaction shaft with low and high distribution air flow rates [78].

Figure 5. Particle mass concentration (kg/m^3) in the reaction shaft with low (left) and high (right) distribution air flow rate [78].

Another example how to use a CFD tool to understand flow and heat transfer in FSF is shown in Fig. 6. The entire furnace including concentrate burner, reaction shaft, settler, uptake shaft, and waste heat boiler with radiation section and a simplified convection section were taken into consideration. Fig. 6a shows the simulated (massless) particle trajectories originating from the concentrate burner and Fig. 6b describes the SO_2 mass fraction contours across a sulphation air jet in the waste heat boiler, respectively.

Figure 6(a). Trajectories of particles in the FSF and its waste heat boiler, seeded at the burner inlet on top of the reaction shaft and travelling through the furnace and the radiation section of the boiler.

Figure 6(b). Contours of SO_2 mass fraction across a jet and a nozzle for sulphation air inlet in the front end of a waste heat boiler in the furnace off-gas train.

From the CFD simulation data of the properties of fluid flow, both mass and heat transfer in the flash smelting furnace, optimized configurations of sulphation air jet and its spray nozzle location were

obtained to prevent buildups and possible corrosion occurring in the WHB. In addition, in a new 'hat-like' design of the uptake shaft roof was proposed based on the CFD approach.

5. Future trends and needs

According to recent forecasts, the demand of copper will be growing by 2050 more than 2× and by 2100 more than 4-5× the current level [118]. This will place further requirements to the smelting and refining technologies of copper, including effective use of the secondary copper resources from end-of-life goods [32]. Complexity of the secondary raw materials and low grade of the primary resources will evidently challenge the future generations, as well as the growing global awareness of emissions to air and environment. This will be assisted by the systemic approach using advanced process and plant simulations [119-121].

Computational thermodynamics is the only tool enabling the rational description of phase equilibria in complex, multicomponent industrial slags where the use of suitable boundary conditions allow their presentation as quasibinary and –ternary sections. This is not possible in a reliable way by curve fitting of experimental data from various sources but needs accurate thermodynamic modelling of the phase properties as a function of composition and temperature.

In early '90s, the CFD was taken as a tool to the flow laboratory work and the physical models helped to develop modelling skills, as there were experimental results in abundance. Gradually, CFD tools partly superseded the physical modelling, especially when it became feasible to include heat transfer and chemical reactions in the simulations. Today, CFD has reached a rather mature and reliable state with enough experienced and knowledgeable users and user communities. Therefore, the parameters and initial conditions used have to be carefully and accurately described in the publications, and the results prudently discussed and critically evaluated.

In the future, CFD will grow in importance along with computing power as new detailed user-defined functions can be included for chemical and physical phenomena occurring in metallurgical reactors. However, CFD alone cannot solve the future knowledge needs, as the feed mixtures are increasingly complex, including secondary materials, like WEEE, or complex primary minerals. Therefore, their behaviour have to be experimentally studied for developing accurate UDFs. CFD is also a critical tool in developing real time process advisors [82] that will be based on digitalized metallurgical knowledge both from industrial practises [122-123] and experimental basic research.

Debottlenecking the operation and throughput maximisation, depending on feed mixture quality will lead to higher recoveries, maximised production, and high resource efficiency and lower dust (solids), fugitive, SO₂ and CO₂ emissions. The control of impurities, or the gangue is typically limiting the smelting capacity when amounts of volatile elements in the feed mixture are high or the fluidity of the slag becomes a problem, including the deportment of various elements in the product streams. The downstream processing would benefit from accurate knowledge of the matte quality, which makes the scheduling of converter aisle, cranes and the actual converter operation easier, the sulphur fixing at acid plant more effective and operations in tank house more effective. Scheduling of the conventional smelting-converting operation from raw materials handling to cathode allows maximisation of the production capacity – i.e. maximising production rate by volume and quality – as the time constant of the *process chain* is several hours (instead of fractions of seconds in RS), even a week considering the tank house operation. This enables use of computation intensive models and algorithms [119, 124-125] for process control and on-line optimisation. This approach can, thus, be extended over the whole copper production chain from raw materials to cathode copper [126]. An active control of fluxing based on feed mixture analysis, automatically as an off-line system as a process advisor and the use of thermodynamic (enthalpy, slag & fluxing chemistry) data is possible even today [127].

Conclusions

The extensive thermodynamic databases of today are the key to comprehensive understanding and analysis of high-temperature chemical processes and environments. They are mostly based on experimental data measured over a long period of time, in many cases more than 50 years ago. The present advanced experimental and analytical techniques give equilibrium data with less systematic errors, higher reliability and accuracy. Several technically important systems are still without experimental observations and those white gaps must be filled in order to improve the reliability of the digital data and the thermodynamic substance databases.

The FSF has been extensively investigated on laboratory scale and later on by using CFD software. Many research groups have developed kinetic models and coupled them with CFD, at least in Australia, China, USA and Finland. Today, the maturity of the FSF reaction shaft models has reached a level, where they can forecast effects of changes in operational parameters, and be used in process and equipment development. Thus, CFD and other simulation methods are reliable tools, but their use requires metallurgical expertise, especially from those who interpret the results and draw conclusions from them. Even though the reactions of sulfidic particles have been included in numerical reaction shaft models, a complete and detailed FSF model, using computational fluid dynamics (CFD) with both gas-solid and gas-liquid sub-models included is still too demanding for today's computer hardware.

As next phase of the FSF modelling, the first steps have now been taken to model immiscible liquid phase phenomena and interactions in the settler of a FSF. This goes with the same procedure as the reaction shaft modelling: starting with a reliable fluid flow simulation and acquiring experimentally the kinetics and mechanisms of the slag-matte forming reactions on top of the settler bath. Other phenomena, such as the droplet interactions by collisions and coalescence, have to be included in a physically reliable way as the suspension from the reaction shaft falls as a rather dense layer of particles and droplets, which will form larger phase volumes separating and forming the matte and slag layers. However, there are always a number of slowly settling individual droplets causing copper losses in slag tapping. A comprehensive and realistic CFD-based model will give the possibility to find ways to enhance settling and reduce mechanical copper losses. In the FCF area, the fundamental research has been so far much less intensive.

In the future, with the strongly increasing importance of the circular economy, the challenges in the development of modelling of both FSF and FCF can be seen in the need for kinetic models for new feed materials, especially for effective recoveries of technical and critical metals included in secondary materials. The experimental work is already going on in Aalto University for obtaining the kinetic data of e.g. WEEE and EoL energy storage materials behaviour when used as a feedstock in the FSF process. Furthermore, time dependent modelling sets challenges to the software and sub-models. Additionally, the concerns of the carbon and sulphur footprints need new tools for seeking ways to minimise the use of fossil fuels and replacing them with renewables, as well as solutions for washing tail gases and fugitive emissions for S-fixing and reduction of dust emissions. As an ultimate goal, the development of artificial intelligence needs deeper understanding of the process details that can only be achieved by the new and advanced, comprehensive CFD models.

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