9th International Symposium on High-Temperature Metallurgical Processing

EDITED BY

Jiann-Yang Hwang Tao Jiang Mark William Kennedy Dean Gregurek Shijie Wang Baojun Zhao Onuralp Yücel Ender Keskinkilic Jerome P. Downey Zhiwei Peng Rafael Padilla





The Minerals, Metals & Materials Series

Jiann-Yang Hwang · Tao Jiang Mark William Kennedy · Dean Gregurek Shijie Wang · Baojun Zhao Onuralp Yücel · Ender Keskinkilic Jerome P. Downey · Zhiwei Peng Rafael Padilla Editors

9th International Symposium on High-Temperature Metallurgical Processing





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Preface

This book features selected papers presented at the 9th International Symposium on High-Temperature Metallurgical Processing organized in conjunction with the TMS 2018 Annual Meeting & Exhibition in Phoenix, Arizona, USA. More than 125 abstracts were submitted. Among them, 74 were selected for oral presentation and 45 were provided with a poster presentation opportunity. After reviewing the 86 submitted manuscripts, 83 of them were accepted for publication on this book.

As the title of symposium suggests, the interests of the symposium is on thermal processing of minerals, metals, and materials that intends to promote physical and chemical transformations of materials to enable the extraction and production of valuable materials such as metals, alloys, ceramics, and compounds.

The symposium was open to participants from both industry and academia and focused on innovative high-temperature technologies including those based on nontraditional heating methods as well as their environmental aspects such as handling and treatment of emission gases and by-products. Because high-temperature processes require high energy input to sustain the temperature at which the processes take place, the symposium addressed the needs for sustainable technologies with reduced energy consumption and reduced emission of pollutants. The symposium also welcomed contributions on thermodynamics and kinetics of chemical reactions and phase transformations that take place at elevated temperatures.

We hope the book will serve as a reference for both new and current metallurgists, particularly those who are actively engaged in exploring innovative technologies and routes that lead to more energy efficient and environmental sustainable solutions. There could not be this book without contributions from the authors of included papers, time and effort that reviewers dedicated to the manuscripts, and help from the publisher. We thank them all! We also want to thank Mr. Mingjun Rao and Mrs. Feng Chen for their assistance in collating the submitted abstracts and manuscripts.

Jiann-Yang Hwang Tao Jiang Mark William Kennedy Dean Gregurek Shijie Wang Baojun Zhao Onuralp Yücel Ender Keskinkilic Jerome P. Downey Zhiwei Peng Rafael Padilla

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About the Editors



Jiann-Yang (Jim) Hwang is a Professor in the Department of Materials Science and Engineering at Michigan Technological University. He is also the Chief Energy and Environment Advisor at the Wuhan Iron and Steel Group Company, a Fortune Global 500 company. He has been the Editor-in-Chief of the Journal of Minerals and Materials Characterization and Engineering since 2002. He has founded several enterprises in areas including water desalination and treatment equipment, microwave steel production, chemicals, fly ash processing, antimicrobial materials, and plating wastes treatment. Several universities have honored him as a Guest Professor, including the Central South University, University of Science and Technology Beijing, Chongqing University, Kunming University of Science and Technology, and Hebei United University.

He received his B.S. degree from National Cheng Kung University in 1974, M.S. in 1980, and Ph.D. in 1982, both from Purdue University. He joined Michigan Technological University in 1984 and has served as its Director of the Institute of Materials Processing from 1992 to 2011 and the Chair of Mining Engineering Department in 1995. He has been a TMS member since 1985. His research interests include the characterization and processing of materials and their applications. He has been actively involved in the areas of separation technologies, pyrometallurgy, microwaves, hydrogen storages, ceramics, recycling, water treatment, environmental protection, biomaterials, and energy and fuels. He has more than 28 patents and has published more than 200 papers. He has chaired the Materials Characterization Committee and the Pyrometallurgy Committee in TMS and has organized several symposiums. He is the recipient of TMS Technology Award and the Michigan Tech Bhata Rath Research Award.

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His research interests include agglomeration and direct reduction of iron ores and extraction of refractory gold ores. He has undertaken more than 50 projects from the government and industry, including National Science Fund for Distinguished Young Scholars Program. He and coworkers invented the direct reduction process of composite binder pellets, and three plants were set up in China based on the invention. He proposed the innovative composite agglomeration process of iron ore fines, which was put into production in Baotou Steel Company, China. He is actively involved in the areas of utilization of nontraditional ferrous resources such as complex ores and various solid wastes. He has published 340 technical papers, 6 books including Direct Reduction of Composite Binder Pellets and Use of DRI, Principle & Technology of Agglomeration of Iron Ores, Chemistry of Extractive Metallurgy of Gold, Electrochemistry and Technology of Catalytical Leaching of Gold. He holds 39 patents and has made more than 35 conference presentations.



Mark William Kennedy is the Chief Technology Officer of Proval Partners of Lausanne Switzerland and part-time Associate Professor at NTNU in Trondheim, Norway. He has a Ph.D. from NTNU and Licentiate degree from KTH in Stockholm, Sweden, both in Materials Science and Engineering, and related to electromagnetic phenomena in metallurgical systems. He also holds a master's degree in Metallurgical Engineering from McGill in Montreal, Quebec, and a bachelor's degree in Chemical Engineering from the University of Waterloo, Ontario. He has previously worked for Noranda and Falconbridge (both now Glencore), and Elkem, working in research and development, engineering projects, and plant operations. He has experience with Cu, Ni, Zn, ferro-alloys, Si, Al, and Mg. He has extensive background in the development and commercialization of new metallurgical technologies including associated financial risks. He is highly knowledgeable about the thermal and electrical behavior of various smelting furnaces and their simulation using finite element modeling, as well as the design and optimization of metallurgical processes using flowsheet simulation software.



Dean Gregurek is a Senior Mineralogist in the RHI AG Technology Center, Leoben, Austria, since 2001. He received his master of science degree at the University of Graz in 1995 and his doctorate degree in Applied Mineralogy from the University of Leoben in 1999. Prior to RHI, he worked two years for Luzenac Europe in the talc business. His current research interests and technical expertise are focused on chemical and mineralogical studies related to interactions between refractories, molten metals, and slags from pyrometallurgical furnaces. He has been a TMS member since 2012, JOM advisor (2014-2017), vice-chair for the Pyrometallurgy Committee, and a co-organizer for the 7th-9th International Symposium on High-Temperature Metallurgical Processing (TMS Annual Meetings 2016–2018).



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Baojun Zhao is Codelco-Fangyuan Professor in the School of Chemical Engineering at The University of Queensland, Brisbane, Australia. His primary fields of research are fundamental investigations of high-temperature processes. He has developed a number of novel research techniques to enable high-temperature properties of the complex oxide systems to be accurately determined. Systematic studies have been undertaken to directly support industrial operations of copper, zinc, lead, ferroalloys, iron, and steel. Development of new refractory materials related to high-temperature processes is also important in his research.

He has received a number of international awards to demonstrate his high-quality research including Spriggs Phase Equilibria Award from The American Ceramic Society, Billiton Gold Medal from Institute of Materials, Minerals and Mining, UK, Non-Ferrous Pyrometallurgy Best Paper Award from Metallurgical Society of CIM, Canada and Science and Technology Progress Award from Chinese Society of Non-ferrous Metals, China. He has long-term collaborations with international



metallurgical companies including Baosteel, Codelco, Dongying Fangyuan Nonferrous Metals, Hesteel, Rio Tinto Iron Ore, Shougang and Luzhong Refractories.

Onuralp Yücel completed his technical education with a Ph.D. in Metallurgical Engineering from Istanbul Technical University (ITU) where he is currently holding the post of Professor since 2002. He was a Visiting Scientist in Berlin Technical University between 1987 and 1988. He carried out postdoctoral studies at New Mexico Institute of Mining and Technology, Socorro, USA, between 1993 and 1994. He has as many as 250 publications/presentations to his credit, which include topics such as technological developments in the production of wide range of metals, ferroalloys, advanced ceramic powders, and application of carbothermic and metallothermic processes, among others. He was the Vice Chairman of ITU, Metallurgical and Materials Engineering Department (MMED), between 2004 and 2007. He has been a Director of ITU, Applied Research Center of Material Science & Production Technologies, between 2006 and 2012. He is currently the Chairman of ITU, MMED.

He is a member of the international advisory board of International Symposium on Boron, Borides and Related Materials (ISBB), International Symposium on Self Propagating High Temperature Synthesis (SHS), and International Metallurgy and Materials Congress (IMMC).

His areas of interest include:

Pyrometallurgy: Pretreatment of concentrates (production of WO₃, Sb₂O₃, As₂O₃, MoO₃), smelting and reduction of slags, production ferroalloys, alloys and metals carbothermic and metallothermic processes in EAF or in ladle (copper, cobalt, vanadium, chromium, ferroboron, cobalt boron, nickel boron, ferromolybdenum, ferromanganese, silicomanganese, ferrovanadium, ferrotungsten, ferrochromium, nickel-chromiummolybdenum-iron, and aluminum-titanium-boron alloys).

Ceramic Powder Production and Processing: Production of carbide, nitride, boride powders and their processing by explosive consolidation or sintering techniques. (B₄C, TiB₂, ZrB₂, SiC, CrB₂). *Beneficiation of Industrial Wastes:* Production of metals and compounds from galvanizing ash, brass production wastes and vanadium sludges produced aluminum production. Grit production from aluminum, copper and steel slags.

Ender Keskinkilic earned his undergraduate degree from the Department of Metallurgical and Materials Engineering of Middle East Technical University (METU), Ankara (the capital city of Turkey) in 1999. He continued M.S. and Ph.D. studies in the same department. He worked as a research assistant in METU between 1999 and 2003. After receiving the master's degree in 2001, he progressed further in the field of extractive metallurgy. During the Ph.D. period, he moved to Eregli-Zonguldak in 2003 and he worked in Quality Metallurgy & RD Department of Eregli Iron and Steel Works Co. (ERDEMIR), which is the leading steel company in Turkey regarding the qualities produced and the production capacity. After earning his Ph.D. degree in 2007, he returned to university and to work in the Department of Metallurgical and Materials Engineering of Atilim University, Ankara, in 2008. He has been working as a faculty in Atilim since then. He was assistant professor between 2009 and 2014. He has been working as an associate professor since 2014. His primary field of interest is extractive metallurgy and more specifically pyrometallurgical processes such as iron- and steelmaking, ladle metallurgy, ferroalloy production, and nonferrous extractive metallurgy.



Jerome P. (Jerry) Downey earned his Ph.D. in Metallurgical and Materials Engineering at Colorado School of Mines and his B.S. and M.S. degrees in Metallurgical Engineering at Montana Tech. He is a registered professional engineer with active licenses in Colorado and Montana. He has over 40 years of professional experience that includes industrial operations, applied process research and development, and corporate management. His technical expertise includes chemical and metallurgical thermodynamics, thermal processing, materials synthesis and processing, and hazardous materials treatment.



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Part I Energy-Efficient and Clean Metallurgical Technology

Simplified Process for Making Anode Copper

Zhi Wang, Haibin Wang, Xueyi Guo, Zhixiang Cui and Baojun Zhao

Abstract In conventional copper production, anode copper is produced from concentrate in three furnaces in a process that entails four oxidation steps and one reduction step. Three types of slags are produced that require further treatment to recover copper. Dongying Fangyuan Nonferrous Metals recently developed a simplified process which requires only two custom-designed furnaces instead of the conventional smelting, converting and refining furnaces. The first furnace continuously produces high-grade matte (>75 wt% Cu) that contains little iron (<2 wt% Fe). The liquid matte is continuously fed to the second furnace, which produces anode copper. The new process significantly reduces the capital and operating costs and increases productivity and environmental sustainability. This paper presents fundamental concepts that enable the simplified process to be developed. The detailed operations in Dongying Fangyuan Nonferrous Metals are also described.

Keywords Anode copper \cdot Two-step production \cdot Fundamental concepts Refining

Introduction

Copper is most commonly present in the earth's crust as sulfide minerals. Anode copper (>99% Cu) is conventionally produced through smelting, converting and fire refining processes [1]. Copper producers have long sought a shorter and more efficient way to produce anode copper from sulfide minerals. Direct-to-blister processes have been accomplished in several smelters using flash furnaces [2–4]. In

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this process smelting and converting are combined into one continuous process as they both have the same oxidation reactions. However, these processes are restricted to the concentrates with a low iron content to control the slag volume, and efficient recovery of copper from the slag remains a challenge.

In 2008, Dongying Fangyuan Nonferrous Metals (Fangyuan) started the first commercial scale bottom-blown oxygen copper smelting furnace, which represents the first modern copper smelting technology developed in China. The technology has a number of advantages: low off-gas and slag volumes, high grade matte produced with low copper content (2-3 wt%) in the slag, low temperature operation to avoid carbon-based fuels and increased furnace campaign life [5-8]. In 2015, Dongying Fangyuan Nonferrous Metals took a further step by developing a new anode copper production process [9]. The new technology combines smelting, converting and fire refining into a two-step process that can treat any copper concentrate. The new process includes one "Smelting Furnace" and two "Converting Furnaces'. The "Smelting Furnace" continuously produces high-grade matte (>75% Cu) that is sent directly through launder to the "Converting Furnaces". In the converting furnaces, the liquid matte is continuously treated through converting and refining steps to produce anode copper for casting and electrorefining. This paper presents the fundamental concepts that enable the new process to be developed as well as a brief description of the Dongying Fangyuan Nonferrous Metals operations.

Fundamental Concepts to Support Two-Step Process

Conventional pyrometallurgical smelting processes require five steps (four oxidation and one reduction) in three furnaces to produce anode copper from concentrate [1]:

Smelting furnace:

(1) concentrate (20–30% Cu) + $O_2 \rightarrow$ matte (50–70% Cu) + smelting slag + SO_2

Converting furnace:

(2) matte (50–70% Cu) + $O_2 \rightarrow$ white metal (Cu₂S) + converting slag + SO₂

(3) white metal $(Cu_2S) + O_2 \rightarrow blister copper (99\% Cu) + SO_2$

Refining furnace:

(4) blister copper (99% Cu) + $O_2 \rightarrow low-S$ copper + refining slag

(5) low-S copper + reductant \rightarrow anode copper (99.5% Cu).

The direct-to-blister effects the first three steps in a continuous process in a single furnace. All of the iron contained in the concentrate is oxidised to form a single slag phase that contains 10–30 wt% Cu [1]. The iron content of the

concentrate is necessarily limited in order to obtain a high direct recovery rate. Consequently, the direct-to-blister process cannot accept many copper concentrates.

In the new process at Fangyuan, anode copper is produced in just three steps and two furnaces:

"Smelting Furnace":

(i) concentrate (20–30% Cu) + $O_2 \rightarrow$ sub-white metal (>75% Cu) + smelting slag + SO_2

"Converting Furnace":

- (ii) sub-white metal (>75% Cu) + $O_2 \rightarrow$ low-S copper + converting slag + SO_2
- (iii) low-S copper + reductant \rightarrow anode copper (99.5% Cu).

The initial step in the Fangyuan process is to produce "sub-white metal" which contains only 1-2% Fe. A unique feature of the bottom-blown furnace is that it enables low-SiO₂ slag to be used with low-Cu in the slag. Therefore, slag volume is controlled even though most of iron is oxidised and reported to the slag. Because the "sub-white metal" brings little iron into the "Converting Furnace" the resulting slag volume is limited. After "low-S copper" is produced in the "Converting Furnace", the slag is removed and the reduction process is initiated to reduce the oxygen content in the melt.

The "Smelting Furnace" and "Converting Furnace" in Fangyuan process differ from those in the conventional process. Some oxidation reactions are combined and fire refining furnace is combined with the converting furnace. The major difference between the new process and conventional process is the converting and refining reactions. Thermodynamic calculations have been performed to examine the phase changes and copper compositions in the process.

The matte composition shown in Table 1 was used for the calculations by FactSage 7.1 and the databases "FactPS", "FToxide" and "FTmisc" were applied [10]. An operating temperature of 1200 °C and a basis of 100 g of matte were assumed. Figures 1, 2 and 3 show three cases where (1) no flux is added, (2) SiO₂ is used as flux, and CaO is used as flux.

As indicated in Fig. 1a, as oxidation progresses, matte mass decreases and white metal (Cu_2S) mass increases. Solid magnetite and SO_2 form at the beginning of the process. After all of the iron in the matte is oxidised, the solid magnetite reaches its maximum and remains constant. Further oxidation produces metallic copper and the amount of white metal starts to decrease. All of the white metal is oxidised to copper and SO_2 reaches maximum after 20 g oxygen is added. More oxygen reacts with Cu to form Cu₂O, which will react with magnetite to form liquid slag.

In this process, a layer of solid magnetite is always covered on the top of the bath before liquid slag is formed. This dense solid layer significantly increases the

Table 1 Matte composition	Cu	Fe	S	Pb	Zn	As
"Converting furnace" (wt%)	75.0	3.0	20.0	1.0	1.0	0.5



Fig. 1 Reactions and copper compositions in the "Converting furnace" at 1200 °C, 100 g matte with the composition given in Table 1, no flux



Fig. 2 Reactions and copper compositions in the "Converting furnace" at 1200 °C, 100 g matte with the composition given in Table 1, 3 g SiO_2 flux



Fig. 3 Reactions and copper compositions in the "Converting furnace" at 1200 $^{\circ}$ C, 100 g matte with the composition given in Table 1, 2 g CaO flux

pressure where the bottom lances faced inside the bath. The high pressure in the bottom of the bath may cause the lances to be blocked and damage by the melt. It is therefore important to use flux to absorb the solid magnetite to form liquid slag from the beginning. Figure 1b shows the impurities in metallic copper during the oxidation progress. Copper starts to form when 5.5 g O_2 is added into 100 g matte.

At the beginning, the arsenic, lead, and sulphur concentrations are relatively high in copper and oxygen is only 0.2 wt%. With increasing oxygen, the arsenic, lead, and sulfur concentrations in melt decrease gradually and oxygen in copper remains almost the same. When 21.5 g oxygen is added, the arsenic, lead, and sulfur concentrations are reduced to 0.6, 0.2 and 1.1 wt% respectively. However, oxygen in the Cu is increased to 0.3 wt%. Consideration of the information provided in Fig. 1a, b indicates the end-point of the oxidation and removal of the slag. However, the end-point of the oxidation varies in different smelters. To minimise the copper loss in the slag, oxidation can be terminated at 23 wt% oxygen, at the point where the solid magnetite has disappeared and a small amount of liquid slag is formed, and the lead, oxygen and sulphur concentrations in the copper are 0.17, 0.9 and 0.04 wt% respectively. More oxygen will be required if it is important to further lower the lead and sulphur concentrations in the copper loss in the slag and oxygen in the copper will increase significantly.

The oxidation process is slightly different when 3 g of SiO_2 is added as flux to 100 g matte. The graph in Fig. 2a shows that liquid slag forms immediately when the iron is oxidised. There is no solid oxide on the top of the bath. As the silicate slag absorbs lead oxide (PbO), the lead and oxygen concentrations in the copper is much lower. For example, 21.5 g oxygen addition results in the concentrations of arsenic, lead, and sulfur in Cu to be 0.6, 0.4 and 0.16 wt% respectively. The sulfur content of the copper is slightly higher due to lower oxygen in the Cu (0.2 wt%).

Calcium ferrite slag has been used in continuous converting processes due to its comparatively low viscosity and high magnetite capacity [1, 11]. Figure 3 shows the oxidation progress when 2 g CaO is added as flux to 100 g matte. The plot in Fig. 3a indicates that liquid slag forms immediately when the iron is oxidised. The slag contains approximately 10 wt% solid oxide but the apparent viscosity of the slag is still low. The slag volume increases when Cu₂O is formed. Calcium ferrite slag has different impacts on the copper quality. For example, Fig. 3b shows that 21.5 g oxygen addition results in the concentrations of arsenic, lead, and sulfur in the copper to be 0.6, 0.7 and 0.6 wt% respectively. Further increase of oxygen to 24 wt% decreases the concentrations of arsenic, lead, and sulphur to 0.3, 0.1 and 0.04 wt% respectively. However, oxygen in the copper will be increased to 2.5 wt%, which will require more reductant in the next step.

In addition to the oxygen and flux, temperature can also influence the impurity content of the copper metal. Figure 4 shows the concentrations of oxygen and sulphur as a function of temperature. After oxidation refining at 1300 °C, the copper contains 1.1 wt% oxygen and 0.1 wt% sulphur. After removal of the slag, if the temperature is decreased to 1140 °C, the concentrations of oxygen and sulphur in the copper will be reduced to 0.6 and 0.03 wt% respectively. This change indicates that, if strong stirring is provided, the oxygen and sulphur in molten copper can be significantly reduced by lowering the temperature.



Description of Two-Step Operation for Anode Copper Production at Fangyuan

Two-step process at Fangyuan includes one "Smelting Furnace" and two "Converting Furnaces" as shown in Fig. 5. The size of the "smelting furnace" is Φ 5.5 × 28.8 m equipped with 23 oxygen lances in two rows. Two slag tapholes are located on the end wall of the "Smelting Furnace" to match the slag tapping rate and transportation of the ladles. Two "Converting Furnaces" are required to treat the liquid matte produced from the "Smelting Furnace". The size of the "Converting Furnace" is Φ 4.8 × 23 m and 17 lances are arranged in two rows to provide different gases. The outer layer is usually used for nitrogen and natural gas while the inner one is used for oxygen and air. The flowrates and ratios of these gases can be accurately controlled through a computer system.



Fig. 5 New anode copper process at Fangyuan, (1) "smelting", (2) "converting", (3) casting [9]

Fig. 4 Concentrations of

oxygen and sulphur as a

function of temperature