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Solidification

The Separation Theory and its Practical Applications

Translated by Judy Wakabayashi

With 143 Figures

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Preface

It was when I saw the countless number of icebergs floating in the North Atlantic Ocean in August 1964 that I decided to commence research into the solidification mechanism of metals, and already I have been continuing this research for two decades.

In 1970 my former professor Susumu Miyata received a letter from President Jiro Komatsu of the Tokyo Keigokin Seisaskusho Co., Ltd. The letter enclosed a copy of an article written by Mr. Imao Sasaki, chief of the casting research section at Daihatsu Motor Co., Ltd., entitled "Aruminyumu gyoko riron no shimpo" (Progress in aluminum solidification theory) from a journal called "Kinzoku Zairyo" (Metals in Engineering).

This article introduced my research in considerable detail and well-written style. After stating that I "rebuffed conventional theories and enlightened casting engineers", Sasaki wrote that "These research results deserve great attention from casting engineers for the progress they have brought about in basic theories on improved soundness in casting quality."

The letter from President Komatsu was a request to arrange a meeting with me. Saying that "I've heard you mention your theory occasionally, but I'd like to hear a full discussion of it for once", Professor Miyata accompanied me to the Tokyo Keigokin Seisakusho Co., Ltd. at Gyoda in Saitama Prefecture.

Apparently my words there that "I have tried to observe the phenomenon of solidification with the clear eyes of a young boy" made a great impression on Professor Miyata. "What you discussed today is wasted in a mere lecture. You should definitely write it up into a book." Acting on his advice, I published the first edition of "Kinzoku Gyoko-gaku" (The Solidification of Metals) in 1973.

In 1975 I gave a lecture at Aahen Technical University in Germany. While I was there I was invited to the home of Teruaki Hiraoka, a Nippon Steel Corporation employee studying in Germany. Mr. Hiraoka had gone to the trouble of taking my book "Kinzoku Gyoko-gaku" all the way to Germany with him.

He handed me the book and asked me to write something on the fly leaf. I inscribed my motto "Resign yourself to your given circumstances".

This current book, "Solidification", is the outcome of Mr. Hiraoka's urging at that time. "You should definitely record the story of your research from its initial conception onwards. I am sure it would be of benefit to young people."

Even now I cannot forget Mr. Hiraoka's enthusiastic words on that occasion and the wonderful crystal-clear echo of our glasses clinking together in a toast.

Since then I have given numerous lectures on the solidification of metals at many universities and companies both in Japan and abroad, not only in the United States and Europe, but also in China, South America and Australia. Constantly keeping in mind Mr. Hiraoka's recommendation that I publish a new book, I have made alterations to the manuscript each time I have delivered a lecture.

In 1980 I collated part of the manuscript into a serialized lecture entitled "Chuzo soshiki contororu no genri" (The principles of cast structure control) and had it published in a journal called "Chutanzo to netsu-shori" (casting, Forging and Heat Treatment). In this series I discussed my motives for commencing research into the solidification of metals, my advocation of the separation theory, and the principles of cast structure control based on the separation theory.

I was amazed to learn that a far greater number of people than I had expected had read this series of lectures. Apparently copies were made at universities and companies all over the place and distributed to students and engineers.

Intending to add a discussion of the practical applications of the separation theory to these lectures and make them into a book, I continued to revise the manuscript whenever I delivered a lecture subsequently. I gave thought to the question of how to give readers a thorough understanding of the separation theory and how to explain it so that they could put it to effective use in actual casting. The questions raised at the conclusion of each lecture were valuable material, as I improved the contents of the book in order to answer these questions.

Putting the separation theory into practice, recently I have developed a continuous casting process in which ingots are solidified from the inside first so as to produce ingots that have no such defects as central segregation or cavities and which have a unidirectionally solidified structure with absolutely no equiaxed crystals. With the appearance of long ingots with absolutely no equiaxed crystals and a beautiful mirror-like surface, I feel that finally the time has come when readers will be able to understand the separation theory.

Already five years have passed since I first started work on this manuscript. There is no limit to the revisions that could be made every time I reread it. To complete the final check of the manuscript, I visited Toronto in Canada, which had been a major influence on my research, and during my week's stay there I finally managed to bring the manuscript to completion.

Having now completed it, I would like to express my profound gratitude not only to the many people from whom I have learnt and to the students and colleagues who have aided me in my research, but also to those who have raised arguments against my theory. To me solidification research is like a game of chess, and it was the existence of these formidable opponents that has made my research so enjoyable.

Toronto, June 5, 1983

Atsumi Ohno

Preface to the English Edition

The rapid development in electronic industrialization has led to a trend towards smaller equipment, and along with this strong calls have arisen for the development of new methods of casting metal materials which have no casting defects or crystals grain boundaries. No sooner was the Japanese edition of this book published in August 1984 than it was immediately adopted as a textbook for employees by many companies in the Japanese material industry, which regard it as providing a wealth of hints for developing metal solidification techniques capable of meeting these contemporary needs.

In the belief that this book can make a major contribution to the progress of casting techniques in English-speaking countries as well as in Japan, the author decided to publish an English edition of the book.

The Japanese language is known for its ambiguity of expression and extreme difficulty of translation, and accurately translating the contents and atmosphere of the original Japanese into readable and natural English is a very demanding task.

I was fortunate enough to have Mrs. Judy Wakabayashi, whose beautiful translation has previously been praised in "Nature", undertake the job of translating this book into English. I would like to take this opportunity to express my heartfelt gratitude to Mrs. Wakabayashi for her great and unsparing efforts in the preparation of this English edition.

Tokyo, January 1987 — Atsumi Ohno

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Introduction

Over the past two decades I have adopted the method of in-situ observations of the solidification phenomenon in metals in order to continue research into the questions of how molten metal solidifies within the mould and how to control the solidification structure of cast metals.

It gives me great pleasure to have this opportunity to report on the results of my research.

The question of how molten metal solidifies within the mould is of great interest, particularly because the inside is opaque. Not only is it of great interest, but a knowledge of the solidification mechanism is also of extreme importance in producing metal materials suited to contemporary needs.

With a few exceptions, nearly all of the metal products with which we come into daily contact undergo fusion and solidification before becoming cast metal or ingots, and then undergo various working, heat treatment and surface treatment to become finished products.

It is well known that structural or casting defects formed in the initial solidification process remain right through into the subsequent products, but since metal is opaque, this important first formulative stage of solidification which metal products must undergo had been disregarded without being fully understood, based on a belief that "solidification is difficult". Recently, however, increased demands for improved quality in metal materials have led people to think that "An understanding of the solidification mechanism is necessary after all."

In this book I will discuss my motives in taking up research into the solidification of metals, and trace the course of this research and propound the "crystal separation theory" and discuss its application. I will conclude with a discussion of the "O.C.C. Process" that I have developed recently, which is a continuous casting process that produces metal materials with a structure in which the crystals are unidirectionally elongated and which have no internal defects.

Based on my research, in 1973 I published a book called "Kinzoku Gyoko-Gaku", and the English edition (The Solidification of Metals) appeared in 1976, while a metallurgy publisher in Moscow put out a Russian edition in 1980. Some time ago I visited China and learnt that a Chinese translation of my book had also been printed and was in use as teaching material at universities throughout China.

Since the book "Kinzoku Gyoko-Gaku" was compiled for textbook purposes, it is a little stiff and terse in style, but in this book it is my intention not to be bound by format but to describe the solidification mechanism and cast structure control in considerable detail and complete freedom, including a full description of experimental results and ideas and arguments against my theory.

eutectic composition was quenched only hypo-eutectic primary crystals were distributed throughout the whole, but when it was cooled gradually both kinds of primary crystals appeared separately in the upper and lower parts. This is regarded as being due to the fact that the formation, separation and floating up (or precipitation) of the primary crystals caused the composition of the liquid in local contact with the mould wall to become hyper-eutectic. Apparently this phenomenon frequently occurs in either the hypo-eutectic or hyper-eutectic side of eutectic alloys. It also seems to be related to the specific gravity of the primary crystals, and is an interesting topic that should be taken up in future research.

3.5 Separation Theory and the O.C.C. Process

So far I have discussed how "Application of the separation theory makes it relatively easy to explain various issues with regard to the solidification of metals that had been regarded as difficult to understand in the past."

It seems, however, that people who adhere to the old concepts cannot readily accept the separation theory. When papers on the formation of equiaxed crystals or the refining of crystals are published, I firstly open up to the last page and check whether or not our papers have been used as references, but in nearly all cases they are not.

I feel that acceptance of the separation theory would lead to the birth of one after another new techniques for cast structure control. Conversely, however, this failure to accept the theory provides those of us with little equipment or funds with plenty of time to develop new casting techniques.

Let me conclude by introducing as one example of these techniques the O.C.C. Process* that I developed recently.

O.C.C. is an abbreviation for Ohno Continuous Casting, and is a process for continuously casting metal materials with any desired sectional form and consisting only of a unidirectionally solidified structure with absolutely no equiaxed crystals.

Since ingots solidify first from the inside and the surface solidifies last, this process enables the casting of beautiful materials with a smooth surface and having no segregation of impurities or gas bubbles and cavities inside. When the ingot is a fine wire or thin plate, it is easy to produce a structure consisting only of single crystals and having absolutely no crystal grain boundaries.

No doubt the reader will think "Something like a cold wire must have been inserted continuously inside, or else how would it be possible to actually cause solidification from the inside first?" But this is possible in actu-

* Ohno A (Japan patent) 1049146; (U.S.A. patent) 4515204; (German patent) 3246470

al practice. Figure 3.40 shows Al ingots cast continuously by the O.C.C. process, and their macro structures. The specular surfaces and the long crystals growing unidirectionally were actually produced by continuous casting.

There are various conventional methods of continuous casting of ingots, but their principles are fundamentally similar, involving pouring molten metal into a cooled hollow mould and causing many crystals to nucleate and grow on the mould wall, and then extracting the ingot after a stable solid shell has formed. When such processes are used, the ingots solidify from the sides inwards, as shown in Fig. 3.41(a), so impurities and gas are concentrated in the center part of the ingots. Cavities frequently form there. Many crystals exist, so naturally impurities and microscopic defects exist in their grain boundaries.

With the rapid development of the electronics industry recently, equipment has become increasingly compact and precise, and demands have arisen for the metal materials used to be of higher quality material, finer and thinner. For instance, extremely fine wires and thin plates with a diameter and thickness of 10 microns or 20 microns and no internal defects have become necessary, such as lead frames and bonding wires for use in ICs and LSIs. At this stage, the crystal structure of the ingot and such casting defects as segregation and gas in the center of the ingot,

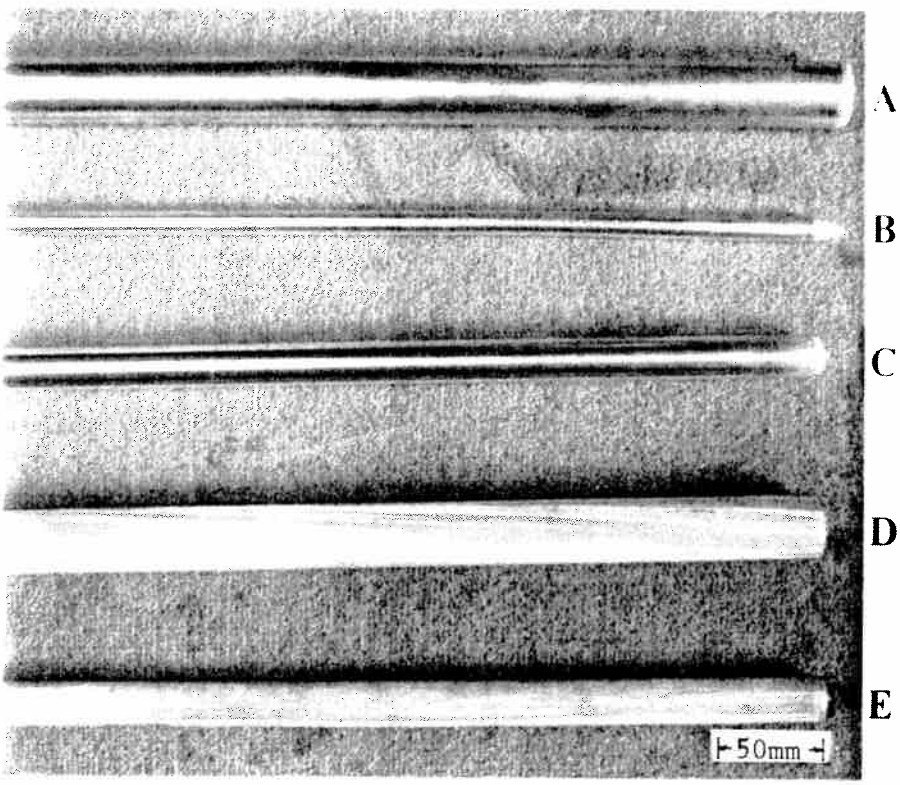


Fig. 3.40 99.8%Al and Al-1%Si alloy ingot cast continuously by the O.C.C. Process. A. B. C. Surface; D. E. Etched structure; A. 30 mm diameter Al; B. 12 mm diameter Al; C. 20 mm diameter Al-Si; D. 30 mm diameter Al; E. 20 mm diameter Al

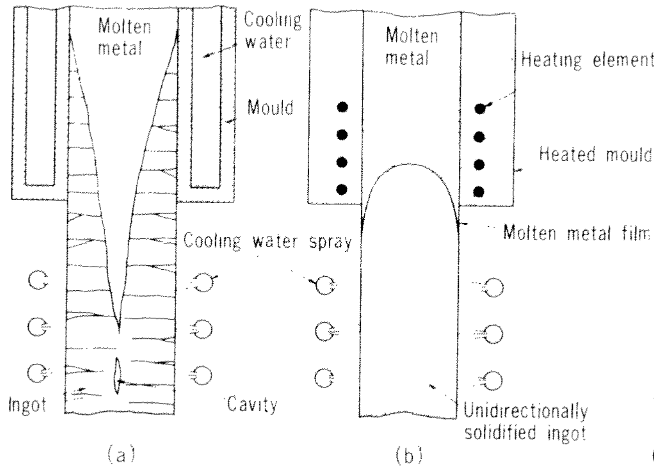


Fig. 3.41 Principle of the O.C.C. Process. (a) Conventional method; (b) O.C.C. Process

which is the original material prior to plastic working, come to have a great effect on the nature of the product. Extremely fine wire breaks during working, and such defects as very fine holes occur in foil.

Recently there have also been calls for materials with no crystal grain boundaries, which in the past was inconceivable, such as copper wires for use in acoustic equipment and the memory disks of computers.

Those of us engaged in cast solidification must supply ingots capable of meeting these sophisticated demands for materials that keep pace with the progress made in such advanced technology. Based on my past research, I developed the O.C.C. process in order to respond to such demands.

Figure 3.41(b) illustrates the principle of the O.C.C. process. When a heated mould is used instead of the conventional cold mould and the temperature of the inside walls of the mould are maintained above the solidification temperature of the cast metal and cooling of the ingot is conducted outside of the mould, solidification of the ingot avoids the mould wall and proceeds without any opportunity for equiaxed crystals to form and separate. If the temperature distribution within the mould is chosen skilfully, it is possible to produce a solidification interface shaped as if projection out within the mould. When this is done, the center part of the ingot solidifies first, and only the thin surface layer solidifies finally immediately outside of the mould.

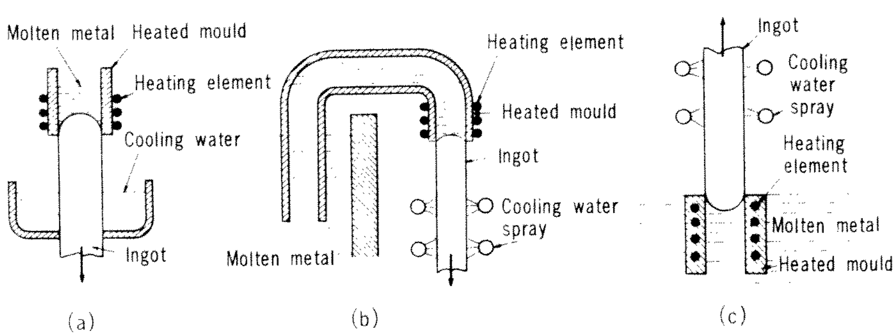
This completely prevents crystals from forming and separating on the mould wall, so the ingots consists of a perfect unidirectionally solidified structure having absolutely no equiaxed crystals. Under this method, there is no opportunity for fresh crystals to form, and the crystals decline in number as they grow by competing against each other, so it is extremely easy to obtain ingots composed of single crystals. Since there is no friction between the mould wall and the ingot, the surface of the ingot is smooth and beautiful. Materials with various complex sectional forms, including of course a round or square form, can be obtained by direct casting. It is also possible to produce plates and tubes continuously from hard and brittle alloys, which is difficult by plastic working.

Looking at Fig. 3.41(b), no doubt the reader will think that the molten metal would burst forth from the bottom of the mould, and that it would be quite impossible to carry out this process. This figure merely demonstrates the principle of the process, and in order to actually use this in continuous casting, measures must be taken to ensure that the molten metal does not gush out from the mould exit. That is, a method must be used which reduces the molten metal pressure at the exit end of the mould to almost zero.

It was six years ago in 1978 that I thought of this O.C.C. process. I devised this in order to cast specular ingots continuously without any friction with the mould.

Firstly I wound nichrome wire around a heat-proof glass tube with an inner diameter of 10 mm, as in Fig. 3.42(a), and made a heated mould, and placed this vertically and set an iron dummy bar in its bottom end. For water cooling I opened a hole in the bottom of a plastic bowl and dropped the dummy bar though it and supplied water inside this bowl. Gradually pouring the Sn melted in the mould, I lowered the dummy bar. Since I used a jack used when cars break down as the apparatus for lowering the dummy bar, the center of the dummy bar would not easily remain fixed, and though a specular ingot was somehow obtained for the first 50 mm or so at the most, the molten metal soon burst out from the bottom of the mould. I had no funds to make any grand equipment, and manufacturing on a commercial scale seemed extremely difficult, so I soon abandoned the experiment. I did apply for a patent, but I had no thought of paying money and registering it even if a patent was obtained in the future.

When I attended the Conference on the Solidification of Metals at the University of Warwick in Coventry in 1980, I was listening to an address



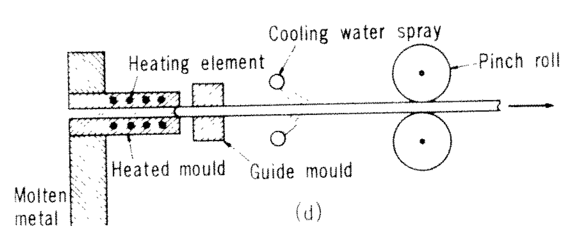


Fig. 3.42 Casting method utilizing the O.C.C. principle. (a) Downwards continuous casting; (b) Siphon-type downwards casting; (c) Upwards continuous casting; (d) Horizontal continuous casting

by Professor M.C. Flemings of MIT on "Process modelling and process development". This included an introduction of the continuous casting process for iron developed by Dr. Eisuke Niiyama and his colleagues at Hitachi, Ltd. This was a continuous casting process using a steel belt and wheels with a groove shape on the periphery, and refined and applied to the continuous casting of iron the Properzi process and SCR process, which had long been used in the continuous casting of aluminum and copper as methods of continuously casting ingots with a beautiful surface without any friction with the mould. Listening to this address, I recalled the O.C.C. experiment that I had abandoned five years earlier.

After the address I told Dr. Niiyama of my O.C.C. idea. He strongly urged me "That's interesting. It may be of use for something, even if not for the continuous casting of iron, which has a high melting point, so don't give up. You should definitely keep at it." I returned to Japan and immediately began to consider methods in which breakout would not occur.

"Why does breakout occur? Because the pressure of the molten metal is exerted on the exit end of the mould. So if the pressure of the molten metal at the mould exit is zero, continuous casting without breakout would be possible." I immediately formed the molten metal feed pipe connecting the holding furnace and the heated mould into a siphon shape, as in Fig. 3.42(b), and ensured that the surface of the molten metal in the holding furnace and the lower end of the heated mould were maintained at the same level. I succeeded in continuously casting bar and tube ingots downwards. However, this process also has its defects. Making the molten metal feed pipe is very difficult in itself, and cleaning inside it is extremely difficult. In addition, there is no place in the mould for gas released during solidification to escape.

In my next experiment I immersed the mould to the surface of the molten metal in the holding furnace, as in Fig. 3.42(c), and held the upper end of the mould at the level of the surface of the molten metal, and after bringing a dummy bar into contact with the molten metal in the mould, I drew it upwards. With this method there is absolutely no risk of breakout. Nevertheless there was the problem that even if cooling by gas is possible, water cooling is difficult for industrialization.

The method I finally arrived at was to extract the ingots sideways from a mould set horizontally at a level directly below the surface of the molten metal in the holding furnace, as in Fig. 3.42(d). With this method water can be used as the coolant and it is easy to cast thin plates or wire rods continuously. Moreover, if a top-open mould is used, the gas released by the solidification interface can escape, and the location of the solidification interface within the mould can be observed, thus greatly facilitating operations.

The ingots produced by this process can be continuously cast to produce plate or bar ingots that all have a smooth and beautiful surface and only a unidirectionally solidified structure with no central segregation. In the past only short single crystals could be obtained, but this enables single crystals unlimited in length to be easily produced. Single crystal wires and plates and tubes can be obtained easily. This is one instance of what I said at the outset that "Application of the separation theory will enable the development of new casting techniques that will make it possible to supply materials suited to contemporary demands."

Instead of reading theses, I adopted the method of directly observing with frank eyes actual solidification phenomena in metal, without any preconceptions whatsoever. When I find a new phenomenon I immediately think it over myself and put together a theory, and try this out directly on specialists who have presented solidification theories in the past and see their reaction. Towards this end I have travelled not only to Europe and the United States, but even as far as Australia. I have attempted to learn the details of the circumstances under which these people carried out their research and how they performed their experiments and built up their theories, not from theses but from the authors themselves.

One method I have used in my research is to simplify as much as possible apparently complex phenomena, and then to observe them. This is because I thought that combining the several pieces of knowledge obtained would be extremely useful in the actual control of cast structures. For example, as I stated when discussing cast iron, in order to understand the apparently complex phenomenon of cast iron solidification, I adopted the method of not immediately using cast iron, but firstly regarding this as one eutectic alloy and using other simple eutectic alloys to investigate solidification phenomena common to all eutectic alloys. Based on this I then attempted to elucidate the complex phenomenon of cast iron solidification.

Another research method I have used is to firstly carry out macroscopic observations and then to observe on a microscopic basis. Before one knows whether the object is a cow or a horse, there is no point in looking only at the pores of the skin just because one has a microscope. I have adopted the method of first observing phenomena with the naked eye, and then if necessary I use a magnifying glass and then a microscope or electronic microscope if further local magnification is necessary. This is because I thought that discussing microscopic segregation within dendrites before a macroscopic understanding of the solidification phenomenon is obtained would not lead to the birth of any new techniques for solidification structure control.

Upon discovering under the microscope alien matter that differs from the surrounding matter, some people immediately jump to the conclusion that it nucleated at that site. Scholars formulate a theory to explain this, and finally this takes root as if it were Scripture. I have endeavoured not to be influenced by such "theories", and have tried not to apprehend solidified structures in one plane, but in terms of the whole casting, including the surface which is the cooled end. The reason that I observed the solidification of an ammonium chloride model in a transparent glass container was also because I tried to grasp the whole picture of the solidification of the metal in the mould in macroscopic terms.

In the course of my research into solidification I have often been remonstrated with – "Why don't you use numerical formulae?" I was afraid that if I got carried away by making numerical formulae and inserted plausible-sounding hypotheses to no purpose without clearly grasping the phenomena, I would rather end up by merely giving listeners the impression that solidification is a difficult matter.

I have often heard people say "Using only data that suits one's purposes results in such an explanation." On each occasion I have told myself "If my theory is correct, a day will surely come when they will accept it. It takes time for them to understand it." With my success in developing the O.C.C. process for the continuous casting of ingots in which single crystals are stretched out without limit, I feel that I have finally completed the sketch of the large picture of metal solidification that I have been drawing over the past two decades. I conclude here in the hope that readers will add beautiful colours to this sketch of mine and finish it off into a splendid painting.