Control of slag and inclusions in traditional Japanese iron- and steelmaking

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In traditional Japanese smelting and forging, three characteristic technologies can be identified that may provide a basis for a new process concept in the production of iron and steel. First, ironsand, the use of which is problematic in modern ironmaking, was used instead of ironstone. The titanium oxide present was effective for keeping the slag fluid and increasing the carbon content of the product, thereby decreasing the activity coefficient of FeO in the slag. Second, two types of operation were carried out with almost the same type of furnace. In one of the operations, mainly molten pig iron was produced and, in the other, steel bloom ($>$50 wt-% of total product) and molten pig iron. The main controlling factors were the type of raw material (titanium content of ironsand) and the oxidising potential in the furnace, which influenced the degree of carburisation and decarburisation of the iron. Third, it is thought that traditional Japanese steel is the best material for Japanese swordmaking. This was confirmed by an experiment with different materials. In the case of traditional Japanese steel, both the homogeneity of the carbon distribution and the inclusion content in the original material can be improved by the forging process, since the degree of contamination during forging is less. With modern steel, weldability in forging is adversely affected by contamination during forging. This means that the complex combination of material and total process is important for producing the particular product. I&S/1690

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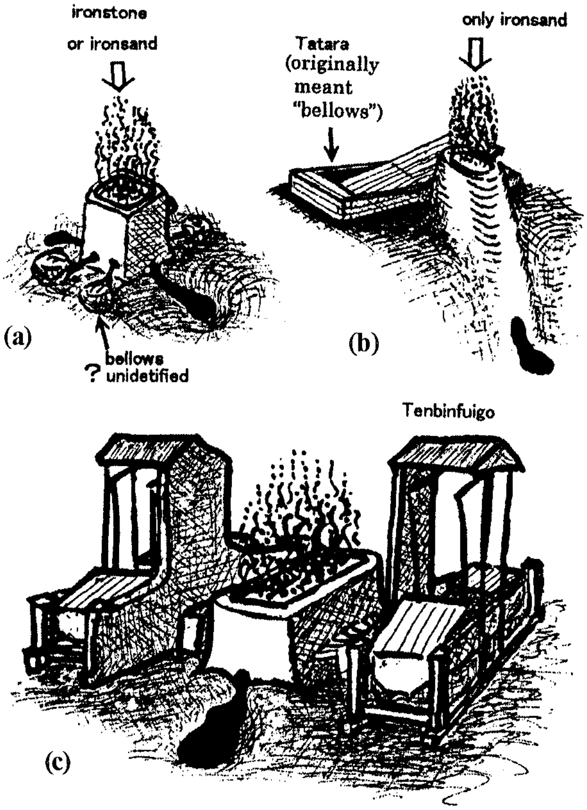
INTRODUCTION

The traditional Japanese iron and steel smelting techniques known as Tatara originated in ancient China and ancient Korea. The conditions of smelting that were commonly used are: charcoal reducatant, no added flux, and furnace walls of soil (consumable). The oldest smelting furnace discovered in Japan dates from the middle of the sixth century. In the earliest box type furnace (Fig. 1*a*), the horizontal section was oval in shape and about 450 mm in diameter.[1] During the eighth century, the shaft type furnace (Fig. 1*b*) appeared in eastern Japan. However, in western Japan, the box type furnace underwent a number of improvements in both the bellows and the underground structure. During the seventeenth century, the box type furnace developed into the Tatara furnace, shown in Fig. 1*c*. The wall of the furnace was about 1·3 × 3 m in section and about 1·3 m in height, and made of soil. The slag notches were made along the short sides, while the tuyeres were placed on the long sides. Using this process, approximately 10^4 t of iron and steel were produced each year in Japan during the eighteenth and nineteenth centuries.

Traditionally, ironstone (magnetite) was used as a raw material for the smelting process in Japan. However, ironsand was eventually adopted in its place. Because ironsand contains titanium oxide, it is considered unsuitable for use in modern ironmaking. The first question to be addressed in this paper will be the reason why ironsand was used instead of ironstone in traditional Japanese smelting.

In the seventeenth century, the technology for producing molten pig iron by using the Tatara process was established. At that time, pig iron was converted to middle or low carbon bloom by using a special blacksmithing technique called Ohkaji. This can be called the traditional Japanese indirect ironmaking system.[2] However, from the beginning



a box type furnace (6th century ×); *b* shaft type furnace (8th century ×); *c* permanent Tatara (17th century ×)

1 Japanese traditional smelting furnaces

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of the nineteenth century, the aim was to produce high carbon bloom directly in the Tatara furnace. Its product (known as Kera or Tama-Hagane) is considered to be the best material for making Japanese swords, which are famous for both their performance and their aesthetic value. The technology and methods used in direct bloom making by the Tatara process, as well as swordmaking, have been kept a closely guarded secret.[3] The second subject to be addressed will be the particular characteristics of the direct steelmaking process, and the role of its product in Japanese swordmaking.

WHY IRONSAND WAS USED IN TRADITIONAL JAPANESE IRON SMELTING

Experimental

One of the present authors carried out 34 smelting experiments using a reconstructed traditional furnace, with ironstone (goethite) and ironsand as the raw materials, to compare the operation and the product.[4] The other conditions chosen were the same as those commonly used in traditional smelting cited above.

In the operation with ironstone, the reduction of iron oxide itself was not difficult, and low carbon iron was obtained. But the fluidity of the slag was poor, and the 'slag-off' operation was difficult, once the temperature reached about 1350°C.

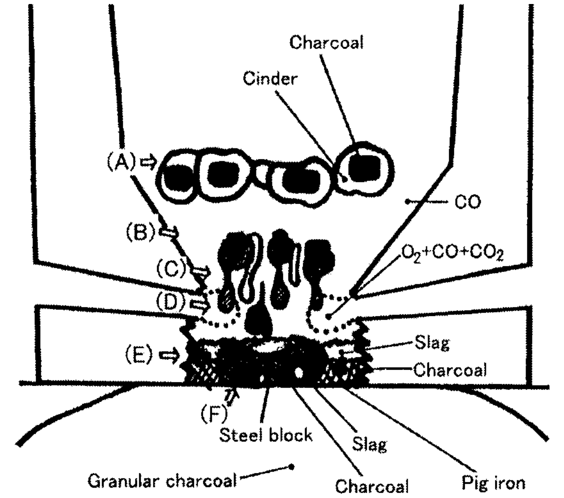
On the other hand, in the operation with ironsand, which contains titanium oxide (*see* Table 1), the 'slag-off' operation was easy at about 1350°C. It was also discovered that the carbon content of the product increased with the titanium oxide content of the ironsand.

It was apparent from these experiments that ironsand was chosen as the raw material in traditional Japanese smelting because it led to easier operation for the production of molten pig iron.

Function of titanium oxide in traditional smelting

Generally, it is thought that titanium oxide additions increase the viscosity of molten slag. Under high reducing conditions, it is true, but under oxidising or low reducing conditions, the influence of titanium oxide on viscosity is not clear. Ohno[5] showed that viscosity decreased with an increase of TiO_2 content in the slag system $CaO–SiO_2–TiO_2$ at 1500°C. This complex influence of titanium oxide on viscosity can be explained by considering that titanium oxide increases viscosity dramatically when TiO is formed under high reducing conditions.

On the other hand, TiO_2 decreases the activity coefficient of FeO in molten slag and keeps the FeO content at a high level in the furnace. FeO in the slag plays the leading role in making the slag fluid. The reaction zones for carburisation and decarburisation of iron in the traditional smelting furnace are shown in Fig. 2. Titanium oxide in the ironsand promotes carburisation of the iron in zones B and C by accelerating the separation of iron from gangue and by suppressing the carburisation in zone D by decreasing the activity coefficient of FeO in molten slag. Therefore, the carbon content of the product is thought to increase with titanium oxide content of the ironsand.



Zone	Tendency
A: Fusion point	Carburisation and decarburisation
B: Adhesion point between charcoal and iron	Carburisation
C: Directly over tuyeres	Carburisation
D: In front of tuyeres	Decarburisation
E: In liquid slag	Decarburisation
F: Adhesion point between bed charcoal and iron	Carburisation

2 Schematic diagram of reaction between carbon and iron in smelting furnace

PROCESS AND PRODUCT OF TRADITIONAL JAPANESE DIRECT STEELMAKING

Before the middle of the eighteenth century, there was no way of cracking large steel blocks, so an operational system capable of preventing the formation of large steel blooms in the Tatara furnaces was chosen. At that time, more than 90 wt-% of the product was pig iron.[6] Around 1750, a method by which steel blocks could be cracked was developed.[6] As the demand for the steel increased, from the beginning of the nineteenth century it was decided to increase the amount of steel bloom produced. Development of the direct steelmaking process continued until the mid-nineteenth century. According to the direct steelmaking process, the ratio of steel product should be about 50 wt-%, the rest being pig iron. An example of the chemical compositions of products from the Tatara process is given in Table 2.[7]

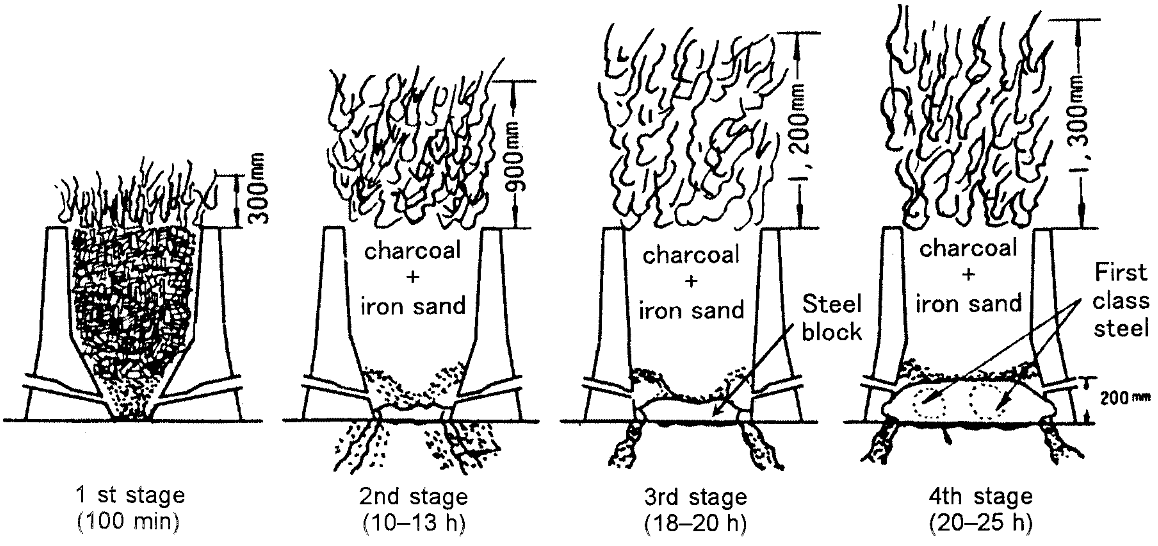
The typical pattern of operation for direct steelmaking by the Tatara process is shown in Fig. 3.[7] The conditions of operation were changed for each stage on the orders of an experienced team leader. The main controlling factors seem to be the type of raw material (titanium content of ironsand) and the oxidising potential in the furnace, which influenced the extent of carburisation and decarburisation of iron. A comparison of the conditions of operation for ironmaking and direct steelmaking[7] is given in Table 3.

In direct steelmaking, the conditions of operation in the first and second stages are similar to those of ironmaking. The main products were molten slag and molten pig iron during the early stages. By changing the condition of operation, steel bloom formed with time following the third

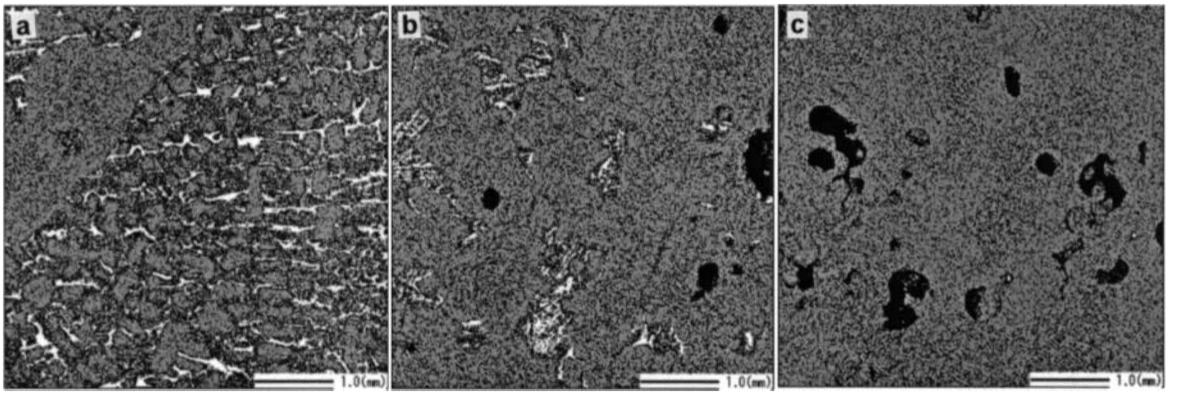
Table 1 Example compositions of ironsand, wt-%*

	T.Fe	TiO_2	FeO	Fe_2O_3	SiO_2	Al_2O_3	CaO	MgO	P	S	V_2O_3
Akome 1	54·56	6·82	18·48	51·08	14·90	4·98	1·60	1·74	0·032	0·036	...
Akome 2	52·07	5·32	19·55	52·71	14·50	4·30	2·68	0·94	0·095	0·026	0·370
Masa 1	59·00	1·27	24·72	64·45	8·40	2·34	2·34	1·54	0·064	0·009	0·258
Masa 2	59·98	1·54	20·98	62·45	10·02	1·62	1·62	1·27	0·060	0·023	0·234

*T.Fe is total Fe content.



3 Operation of Tatara furnace for direct steelmaking



a dendritic; *b* locally dendritic; *c* non-dendritic

4 Various types of P distribution in steel block, determined by EPMA

stage (Fig. 3). After 3 days, the steel block was removed after breaking the furnace. This block was cracked and the product sorted through observation of each section. First class steel product is known as Tama-Hagane, while the inferior product is referred to as Ohwari-Sita.

Many theories have been presented concerning the mechanism for direct steelmaking in the Tatara furnace: (i) the crystallisation of steel from molten pig iron,[8] (ii) the decarburisation of molten pig iron by reaction with unreduced ironsand, molten slag, or the oxidising atmosphere,[9,10] and (iii) the sintering of reduced iron.[9]

To verify this, the distribution of phosphorus at three locations in the sample of steel block (80 × 50 × 40 mm) was investigated by electron probe microanalysis (EPMA) (*see* Fig. 4). The patterns could be divided into three types: dendritic, partly dendritic, and non-dendritic. This result shows that the steel directly produced in the Tatara process is a mixture of steel produced through the state of molten pig iron, and that which was not. There is a technical tradition that high quality traditional steel can be made when a large amount of molten pig iron is produced.[6] Therefore, it can be concluded that the first class steel was produced through molten pig iron, while the inferior steel was a product of the sintering of reduced iron.

CHARACTERISTICS OF SWORDMAKING WITH TRADITIONAL JAPANESE STEEL

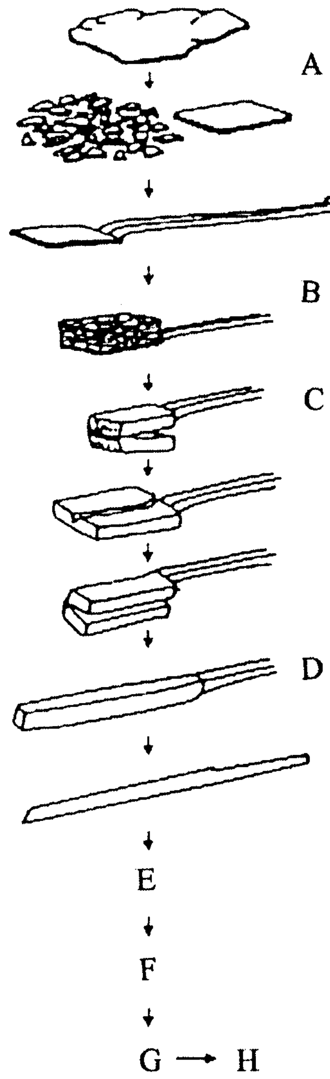
For good performance, the Japanese sword must be free from large non-metallic inclusions and the inhomogeneous distribution of inclusions. At the same time, for enhancing the aesthetic value, a moderate non-uniformity of carbon content is required.[11]

An outline of the stages of the swordmaking process[12] is as follows (*see* Fig. 5 also): Tama-Heshi (forging to flat plate, A); Tsumi-Wakashi (welding of piled plate through

Table 2 Example of composition of products* from Tatara process, wt-%

Product	C	Si	Mn	P	S	Ti	V	O	T.Fe	FeO	Fe_2O_3	SiO_2	MnO	Al_2O_3	CaO	P	TiO_2
Pig iron 1	3·12	0·37	trace	0·046	0·023	...	0·02	0·0131	...	...	...	...	...	...	...	...	...
Pig iron 2	3·44	0·11	trace	0·043	0·022	...	0·02	0·0176	...	...	...	...	...	...	...	...	...
T-H 1	1·42	trace	trace	0·013	0·007	0·004	0·02	0·0115	...	...	...	...	...	...	...	...	...
T-H 2	1·17	0·02	0·02	0·032	0·008	0·004	0·02	0·0267	...	...	...	...	...	...	...	...	...
Slag 1	...	...	...	...	...	...	...	...	34·07	36·68	7·07	27·66	1·63	6·08	2·29	0·12	11·43
Slag 2	...	...	...	...	...	...	...	...	49·52	58·85	5·40	22·52	1·23	5·40	0·18	0·02	5·10
Slag 3	...	...	...	...	...	...	...	...	27·20	30·76	4·62	41·30	1·16	9·21	1·49	0·03	9·51

*T-H is Tama-Hagane.



stage A: Tama-Heshi (forging to flat plate); stage B: Tsumi-Wakashi (welding of piled plate through light hammering at high temperature); stage C: Orikaeshi-Tanren (straight forging and folding – fold five times for Shita-Gitae as an early stage of forging, and seven times for Age-Gitae as a latter stage); stage D: Sunobe (straight forging); stages E–H: finish forging to sword shape, quenching, grinding, and polishing

5 Outline of stages of swordmaking process: good weldability is important at stages B and C

Table 3 Comparison conditions of operation for ironmaking and direct steelmaking

Condition	Ironmaking	Direct steelmaking
Type of ironsand	Akome (TiO_2: 5–12%)	Masa (TiO_2: 1–2%)
Travelling time of ironsand	A few hours	A little shorter
Air blowing through tuyere	Soft blowing	A little harder
Temperature	...	Be aware of upper limit
Erosion of wall	Moderate	Severe
Operation time	~80 h	~55 h

light hammering at high temperature, B); Orikaeshi-Tanren (straight forging and folding – fold five times for Shita-Gitae as an early stage of forging, and seven times for Age-Gitae as a latter stage, C); Sunobe (straight forging, D); finish forging to sword shape, quenching, grinding, and polishing (E–H). During the swordmaking process, good weldability is important at stages B and C.

To clarify the characteristics of Tama-Hagane as a material for Japanese swordmaking, Kishida *et al.*[12] compared the behaviour of eight types of iron and steel material during the swordmaking process. Their work is summarised below

Raw materials for experiments

The raw materials used were (*see* Table 4 for chemical compositions): Tama-Hagane (first class steel produced by the Nittohho Tatara process); Wazuku (pig iron from the Tatara process); Ohkaji material (two types, produced by decarburising Wazuku); Ohwari-Sita (inferior steel product sorted from a steel bloom); Kotetsu (Japanese ancient iron collected during the repair of Himeji castle); and electrolytic pure iron and cutlery steel (products of modern steelmaking).

The microstructures and chemical compositions of the raw materials are not uniform except for the modern steelmaking products (i.e. electrolytic iron and cutlery steel) before the Tama-Heshi (Fig. 5, stage A); the carbon contents of Wazuku, Kotetsu, and electrolytic pure iron were controlled by a special blacksmithing technique called Oroshi. All samples were forged by one swordsmith, and test specimens were taken at each stage from A to D illustrated in Fig. 5.

Results of experiments

Distribution of carbon content

The change of carbon content in the forging process as determined by EPMA analysis is shown in Fig. 6. The range of deviation becomes narrower, especially between stage B (Tsumi-Wakashi) and stage C/S (Shita-Gitae), with the range becoming small (sufficient for practical use) at the stage of Age-Gitae (C/Ag) or Sunobe (D).

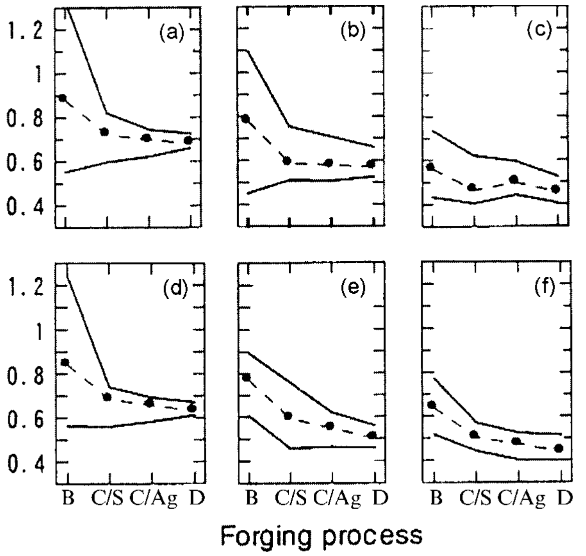
Non-metallic inclusions

The inclusion size decreases monotonically in every material by elongation and tearing during the forging process. However, two types of behaviour are noteworthy and can be observed in the change of inclusion area during forging (Fig. 7). The inclusions of Tama-Hagane (in which carbon content is kept at a high level throughout the forging process) are mainly fayalite (Fe_2SiO_4) of thinly elongated and torn state, containing titanium or vanadium oxides, which come from ironsand and some silicate. They are less prone to contamination during forging. In the Ohkaji material 1, in which the carbon content is kept low level throughout the forging process, contamination by FeO and Al_2O_3 during forging is more noticeable. The inclusions in electrolytic iron are mainly FeO, Al_2O_3, SiO_2, and other silicates, which are introduced during the forging process. Cutlery steel contains silicate, which was contained in the raw materials.

Table 4 Compositions of materials used in swordmaking experiments, wt-%

Material	C	Si	Mn	P	S	V	Al	Ti	C_{Sunobe}*
Tama-Hagane	1·31	0·018	0·006	0·042	0·0066	0·015	0·003	0·003	0·69
Wazuku	2·06	0·017	0·011	0·081	0·0164	0·021	0·002	0·003	0·57
Ohkaji 1	1·04	0·025	0·015	0·073	0·0088	0·040	0·006	0·010	0·37
Ohkaji 2	1·00	0·025	0·005	0·046	0·0097	0·006	0·016	0·011	0·63
Ohwari-Sita	0·61	0·022	0·007	0·067	0·0075	0·001	0·019	0·004	0·45
Kotetsu	0·14	0·024	0·003	0·051	0·0030	0·002	0·037	0·008	0·37
Electrolytic iron	0·0015	0·0005	0·0001	0·003	0·0006	...	0·001	...	0·50
Cutlery steel	1·12	0·180	0·240	0·017	0·0040	0·004	0·001	0·002	0·89

*C_{Sunobe} is C content of Sunobe material (stage D of forging process).



a Tama-Hagane; *b* Wazuku; *c* Ohkaji 1; *d* Ohkaji 2; *e* Ohwari-Sita; *f* Kotetsu (ancient steel)

6 Change of carbon content during forging: B, C/S, C/Ag, and D are respectively Tsumi-Wakashi, Shita-Gitae, Age-Gitae, and Sunobe stages of forging process (as illustrated in Fig. 5) and solid lines represent maximum and minimum values

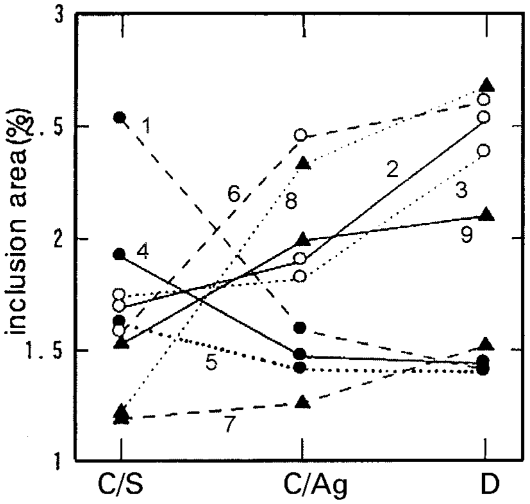
Weldability

Specimens (7 mm in diameter and 3·5 mm in length) cut from materials in the Shita-Gitae state were polished and the polished surfaces were stacked and heated in argon atmosphere at 800°C for 5 min. Then the specimens were compressed to 30% displacement, and air cooled. The Tama-Hagane specimen showed hardly any peeling and good welding. Both electrolytic iron and cutlery steel gave insufficient welding with noticeable tears, as a result of contamination.

DISCUSSION

Effective use of titanium oxide in ironsand

The main route for the development of ironmaking around the world was achieved by: (i) producing molten pig iron stably by increasing the operating temperature, (ii) promoting the reduction of iron oxide by using a high shaft furnace, and (iii) adding CaO as flux. However, in traditional Japanese ironmaking, the titanium oxide in ironsand was used effectively for decreasing the activity coefficient of FeO in the molten slag. It was possible to derive a fluid slag and a high carbon molten metal at moderate temperatures. This may seem to be uneconomical, because the yield of iron was low (30–50%). However, the cost fraction of total for ironsand was only about 15%, whereas for charcoal it exceeded 45%.[7] It can therefore be appreciated that ironsand was the best choice as the raw material from the standpoint of production costs at that time.



1: Tama-Hagane; 2: Wazuku; 3: Ohkaji 1; 4: Ohkaji 2; 5: Ohwari-Sita; 6: Kotetsu; 7: electrolytic iron 1; 8: electrolytic iron 2; 9: cutlery steel

7 Change of inclusion area during forging: C/S, C/Ag, and D are respectively Shita-Gitae, Age-Gitae, and Sunobe stages of forging process

Combination of process stages

As a material for Japanese swordmaking, modern steel, which is clean and homogeneous, is inferior to Tama-Hagane, the traditional steel. This is because the former steel is contaminated during forging, whereas the latter is improved by forging with less contamination. Tama-Hagane is inhomogeneous in carbon content and this may play an important role during traditional swordmaking in decreasing the inclusions and preventing contamination, as a consequence of partial melting during the forging process.

Modern technologies have been developed by dividing the process into several stages and optimising each stage. However, the factors mentioned above suggest that not only optimisation of each stage, but also the combinations constituting the total process are important.

SUMMARY

Considering the remarkable progress in technologies in iron- and steelmaking during the twentieth century, it seems that there is little room for essential improvement. However, to cope with the change of conditions surrounding iron- and steelmaking, e.g. resources, energy, environment regulation, and demand for the product, the optimum combination of process stages must be pursued. It is expected that the technologies of traditional Japanese iron- and steelmaking, which were developed separately from those in the rest of the world about 100–300 years ago, may suggest a new process concept for the future.

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