Control of Slag and Inclusions in Traditional Japanese Iron- and Steelmaking Processes

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Synopsis
As inspiration in developing new iron and steel processes and products, three topics of traditional Japanese smelting and forging are presented.

(1) Iron sand, which is difficult to use in modern ironmaking, was used willingly instead of ironstone in traditional Japanese smelting. Titanium oxide was used to maintain slag fluidity and increase the carbon content of the product by decreasing the activity coefficient of Fe0 in the slag.
(2) Two types of operations were carried out with very similar furnaces. In one of the operations, mainly molten pig iron was produced, and in the other, steel blooms (higher than 50 wt% of the total product) and molten pig iron. The main controlling factors were the type of raw material used (titanium content of the iron sand) and the oxidizing potential of the furnace operation, which influenced the degree of iron carburization and decarburization.
(3) It is believed that traditional Japanese steel is the best material for making Japanese swords. This is confirmed by experiments conducted with different materials. In the case of traditional Japanese steel, being of non-uniform, or inhomogeneous composition in respect to carbon and inclusion content, the original material can be improved by forging and there is less contamination during forging. On the other hand, in the case of modern steel, the forging process negatively affects steel weldability by contamination. This means that the complex combination of material and steps of the total process is important for producing the desired product characteristics.

1. Introduction

The traditional Japanese iron and steel smelting techniques, called Tatara, originated in ancient China and Korea. The oldest smelting furnace excavated in Japan is from the mid-6TH century. In the earliest box-type furnace (Fig. 1-a), a horizontal cross section was oval (diameter about 45 cm)[1]. In the 8TH century, a shaft-type furnace (Fig. 1-b) appeared in eastern Japan, but in western Japan, the box-type furnace underwent improvement of both the bellows and the underground structure, resulting in the 17TH-century Tatara furnace shown in Fig. 1-c. The walls of the furnace (cross section about 1.3 × 3 m, height about 1.3 m) were made of earth. The slag notches were located along both of the smaller sides, while the tuyeres were on the longer sides. Using this furnace, about 10,000 tonnes of iron and steel were produced per year in Japan in the 18TH and 19TH centuries.

In traditional Japanese smelting, ironstone (magnetite) was used as a raw material, at first. But soon afterwards, iron sand was adopted instead of ironstone. As iron sand contains titanium oxide, it is not desirable in modern ironmaking processes. But why was iron sand willingly used instead of ironstone in traditional Japanese processes? This is the first subject to be discussed.

In the 17^{TH} century, the technological basis for producing the molten pig iron by the Tatara process was established. The pig iron produced was converted to medium- or low- carbon bloom material by a special blacksmithing technique called Ohkaji. This can be called the traditional Japanese indirect ironmaking system[2)]. But, from the beginning of the 19^{TH} century, the aim was to produce high-carbon blooms directly in the Tatara furnace. Its product, called Kera or Tama-Hagane, is thought to be the best material for making the Japanese sword, famous for both its function and aesthetic value. The direct production of blooms by the Tatara process and the knowledge of the skills of sword making have been kept strictly secret[3)]. The second subject to be discussed includes the characteristics of the process of direct steelmaking and its product as the material of Japanese sword making.

2. Why was iron sand used in traditional Japanese iron smelting?

2.1 Experiments

One of the authors, Haba, carried out 34 smelting experiments with a reconstructed traditional furnace using iron stone and iron sand (goethite) as raw materials in order to compare the operations and the products[4)]. The other conditions were the same as those of traditional smelting (Table 1). 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 deslagging operation was difficult, at a temperature of about 1350°C. However, in the operation using iron sand, which contains titanium oxide (Table 1- a), deslagging was easy at about 1350 °C. It was also observed that the carbon content of the product increased with the content of titania in the iron sand.

These experiments made it clear that iron sand was chosen as the raw material because it made the operations for the production of molten pig iron for the conditions of traditional Japanese smelting easier.

2.2 The function of titanium oxide in traditional smelting

Ordinarily, it is thought that titanium oxide increases the viscosity of the molten slag. Under high reducing conditions, this is true, but under oxidizing or low reducing conditions, the influence of titanium oxide on viscosity is not so clear. Ohho[5)] showed that viscosity decreases with TiO_2 in the CaO-SiO_2-TiO_2 slag system at 1500 °C. This complex influence of titanium oxide on viscosity can be explained by considering that titanium oxide increases viscosity remarkably when Ti0 is formed under high reducing conditions.

On the other hand, TiO_2 decreases the activity coefficient of FeO in molten slag and maintains a high FeO content in the furnace. FeO in the slag plays the leading part in making the slag fluid.

Fig. 2 illustrates the carburization and decarburization of iron in the traditional smelting furnace. Titanium oxide in the iron sand promotes the carburization of iron in zones (*B*) and (*C*) by accelerating the separation of iron from gangue and by suppressing carburization in zone (*D*) by decreasing the activity coefficient of FeO in the molten slag. Therefore, the carbon content of the product is thought to increase with the titanium oxide content of the iron sand.

3. The processes and products of traditional Japanese direct steelmaking

In the 17^{TH} century, large steel blooms were occasionally formed in Tatara furnaces. As such blooms could not be cracked until the middle of the 18^{TH} century, an operation preventing the formation of these large steel blocks was used. In the process, more than 90 wt% of the product was pig iron[6)]. In about 1850, a process which could crack the blocks was developed[6)]. From the beginning of the 19^{TH} century, as the demand for steel increased, the amount of steel-bloom production increased. And in the middle of the 19^{TH} century, the direct steelmaking process was established, rendering a product of at least 50 wt% steel (the rest was pig iron).

The typical direct steelmaking procedure is illustrated in Fig. 4[7)]. In the first stage, molten slag and molten pig iron were produced. In the operation for producing mainly pig iron, the existing conditions remained unaltered to the next stage, but in the direct steelmaking operation, the conditions were strictly modified for each stage by the instruction of an experienced leader. A comparison of the operating conditions of the two processes[7)] is shown in Table 3. The main controlling factors seem to be the type of raw material (Ti content of the iron sand) and the oxidizing potential in the furnace, which influenced the degree of iron carburization and decarburization.

The formation of the steel bloom took time (Fig. 3), and after 3 days, the steel block was taken out. This block was cracked and the product sections were visually examined by a master of the skill and sorted by quality. Tama-Hagane refers to the parts of first-class steel quality, and Ohwarisita to those of inferior quality. An example of the steel product's chemical composition is shown in Table 3[7)].

Many theories of the mechanisms of direct steelmaking in the Tatara furnace have been presented: (1) the dissolution of molten pig iron[8)], (2) the decarburization of molten pig iron by unreduced iron sand, molten slag or oxidizing atmosphere[9,10)] (Fig. 2), and (3) the sintering of reduced iron[9)].

In order to verify these, the distribution of P at three points in a sample of a steel block (80×50×40mm) was investigated by EPMA (Fig. 4). The results were divided into three categories: dendritic, partly dendritic and non-dendritic. This means that the directly produced steel by the Tatara process was a mixture of steel which goes through a state of molten pig iron and steel which does not There is a technical tradition that excellent traditional steel can be produced when a large amount of molten pig iron is produced. We can suppose that the first-class steel part was produced with molten pig iron, and the inferior part was produced by the sintering of reduced iron.

4. Traditional steel sword making

4.1 The sword making process and the necessary conditions

It is important that the steel material used for Japanese swords is free of large non-metallic inclusions and has a homogeneous inclusion distribution. At the same time, as an aesthetic factor, moderate non-uniformity, especially the right deviation in carbon content (not compromising the perfection of the function of the sword) is necessary[11)]. The sword making process[12)] is outlined as follows (Fig. 5):

(*A*) Tama-Heshi: Forging to a flat plate
(*B*) Tsumi-Wakashi: Welding of the piled plate by light hammering at a high temperature
(*C*) Orikaeshi-Tanren: Straight forging and folding - folding 5 times for Shita-Gitae as an early stage of forging, and folding 7 times for Age-Gitae as a latter forging stage

(*D*) Sunobe: Straight forging
(*E*) - (*H*): Final forging to make sword shape - quenching, grinding and polishing.

During this process, good weldability in (*B*) and (*C*) is important.

4.2 Experiments

In order to clarify the characteristics of the traditional Japanese sword material Tama-Hagane, the behavior of eight kinds of iron and steel materials in the sword making process was compared[12].

The tested raw materials and their chemical compositions are shown in Table 5.

¥Tama-Hagane:	the first-class steel produced by the Nittohho Tatara process
¥Wazuku:	pig iron by the Tatara process
¥Ohkaji-Material (2):	steel, decarburized Wazuku
¥Ohwari-Sita:	inferior steel part sorted from a steel bloom
¥Kotetsu:	ancient Japanese iron collected during repair of the Himeji castle
¥Electrolytic pure iron	
¥Cutlery steel:	product of a modern steelmaking process

The microstructure and chemical composition of the raw materials are not uniform, except for the modern steel material.

Before the Tama-Heshi stage (Fig. 5-(*A*)), the carbon contents of the Wazuku, Kotetsu and electrolytic pure iron materials were controlled using a special blacksmithing technique called Oroshi. All samples were forged by one sword smith, and test specimens were taken at each stage.

4.3 Experiment results

Distribution of carbon content

Fig. 6 shows the change in the deviation of carbon content in the forging process by EPMA analysis. The range of deviation becomes narrower, especially in the first stage of forging, and converges to a small range sufficient for practical use in the stage of Age-Gitate or Snobe.

Non-metallic inclusions

The average inclusion size decreased uniformly in every material by elongation and tearing off with the process of forging. But, two types of changes were observed in inclusion behavior during forging (Fig. 7). The inclusions of the Tama-Hagane material (for which a high carbon content is maintained throughout the forging process) were mainly fayalite inclusions (Fe_2SiO_4) in a thin, elongated and torn state, and contained titanium or vanadium oxides, which results from iron sand and some quantity of silicate, and less contamination by forging.

In the Ohkaji materials (for which the carbon content is kept at a low level through the forging process), contamination by FeO and Al_2O_3 during forging was noted.

The inclusions found in the electrolytic iron were mainly FeO, Al_20_3, SiO_2 and other silicates, which are introduced in the forging process. The cutlery steel contained silicate, which was contained in the raw materials.

Weldability

After polishing the specimens (7 mm diameter, 3.5mm length) cut from Shita-Gitae material, the polished surfaces were attached and heated in an Ar atmosphere at 800°C for 5 minutes. The samples were compressed to 30 % displacement at the strain rate of 1 and air cooled.

From the experiment, it was found that the Tama-Hagane material showed little peeling and a good weld. Both the welds of the electrolytic iron and cutlery steel materials were insufficient with noticeable tears as a result of contamination.

5. Consideration

5.1 Effective use of titanium oxide in iron sand

Ironmaking in most of the world consisted of producing molten pig iron by increasing the temperature and promoting the reduction of iron oxide using a high-shaft furnace and adding CaO as flux. However, in traditional Japanese ironmaking, titanium oxide in iron sand was effectively used for decreasing the activity coefficient of iron oxide in the slag. This made it possible for both a fluid slag high in FeO content and a high-carbon product to coexist at a moderate temperature. The process does not seem to have been an economical one, because the yield of iron was low (30-50%). But, the proportional cost of the iron sand was about 15 % of the total cost and that of charcoal was higher than 45%[7]. We can, therefore, understand that the best alternative was to use iron sand for reducing the total cost at the time.

5.2 Combination of each process

The modern process has been developed by dividing the process into many parts and optimizing each part. But, modern steel, which is clean and homogeneous, is inferior to the traditional Tama-Hagane steel, which is inhomogeneous in carbon and inclusion content, as a material for the making of Japanese swords. In the process of the former, the steel was contaminated during forging, but in the process of the latter, the steel was improved by forging and the degree of contamination was less. The inhomogeneity of carbon content in the traditional steel may play an important role in decreasing inclusion content and preventing contamination by partially melting during forging. This suggests that not only optimization of each step, but also optimization of the combination of the steps of the process is important.

6. Summary

It seems that the development of iron and steelmaking during the 20TH century has left little space for substantial change. But, if the conditions governing current iron and steelmaking (e.g. resources, energy, environmental regulation and product demand) change, this may no longer be the case. Here, consideration of traditional Japanese iron and steelmaking processes, which, about 100-300 years ago, developed separately from their counterparts used in most of the rest of the world, may inspire new concepts.

References

1) Mitsuru.Tete; Proceedings of The Fourth International Conference on the beginning of the Use of Metals and Alloys p, 15, (May 1998), Matsue.
2) Takuo Sizuki; ibid.p.207
3) Yoshinori Anazawa; ibid, p.21
4) Mutsumi Haba; ibid, p.29
5) Kazumi Ohno; Tetsu-to-Hagane (1965) p.110
6) Ichiro Takahashi; Ferum 1(1996) 11.P.46 (ISIJ)

7) Jukichi Kozuka; Tetsu-to-Hagane (1966) p.1763
8) Kuniichi Tawara; Japanese traditional smelting of iron and steel with iron sand (Maruzen) (1933)
9) Kingo Kiyonaga; Materia Japan 33(1994) 1. p.1453
10) Kazuhiro Nagata; Private Communication
11) Takuo Onoue; Private Communication
12) Tamiya Kishida, Yutaka Sato, Yoshiyuki Murakawa and et.al; Proceedings of The Fourth International Conference on the Beginning of the Use of Metals and Alloys p.41 (May 1998), Matsue.

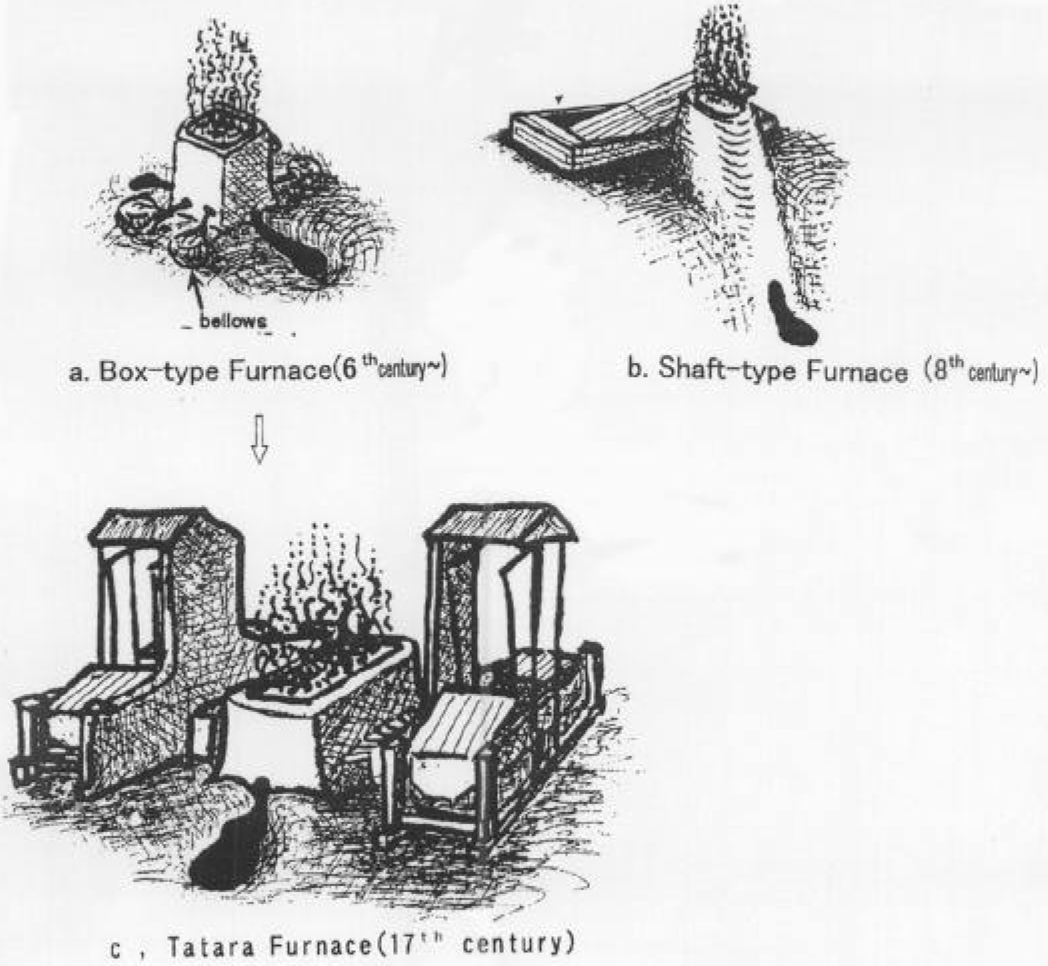
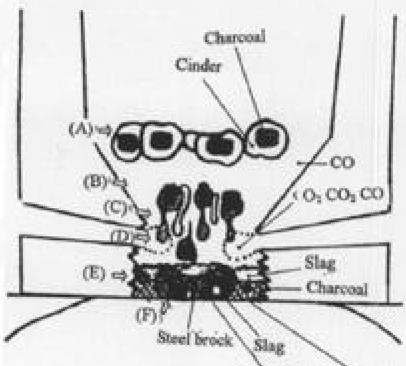


Fig. 1. Image of Japanese traditionl smelting furnaces



Position	Tendency
(A) Fusion point	Carburization or DeCarburization
(B) Adhesion point between charcoal and iron	Carburization
(C) Directly over tuyeres	Carburization
(D) In front of tuyeres	Decarburization
(E) In liquid slag	Decarburization
(F) Adhesion point between bed-charcoal and iron	Carburization

Fig.2 Shematic drawing of reaction between carbon and iron in smelting furnace

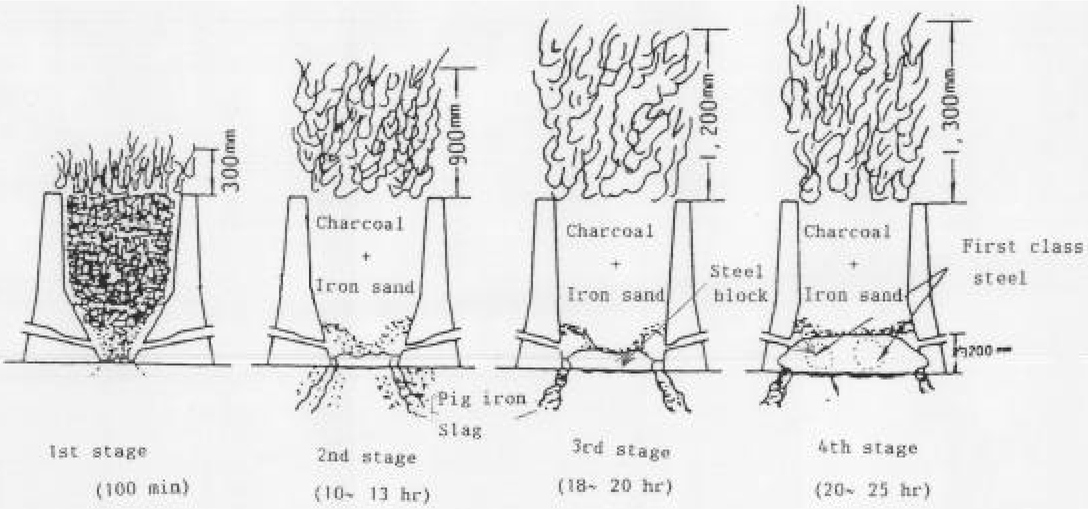


Fig.3 Operation of Tatara for direct steel making

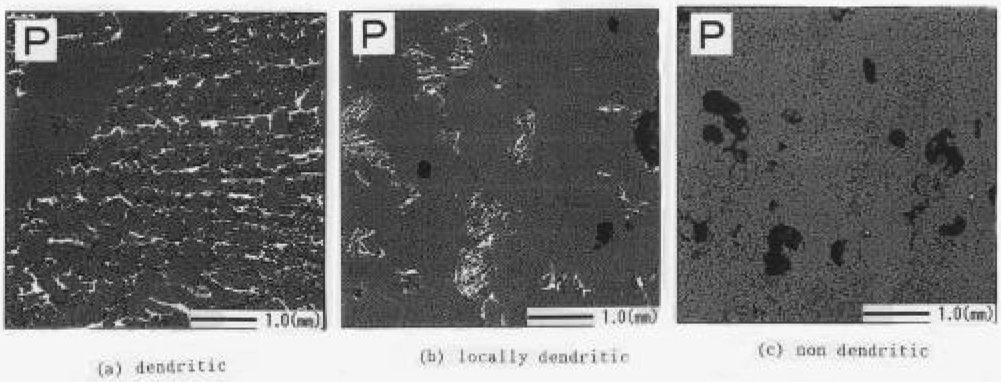


Fig.4 Various type of P distribution by EPMA in steel block by EPMA

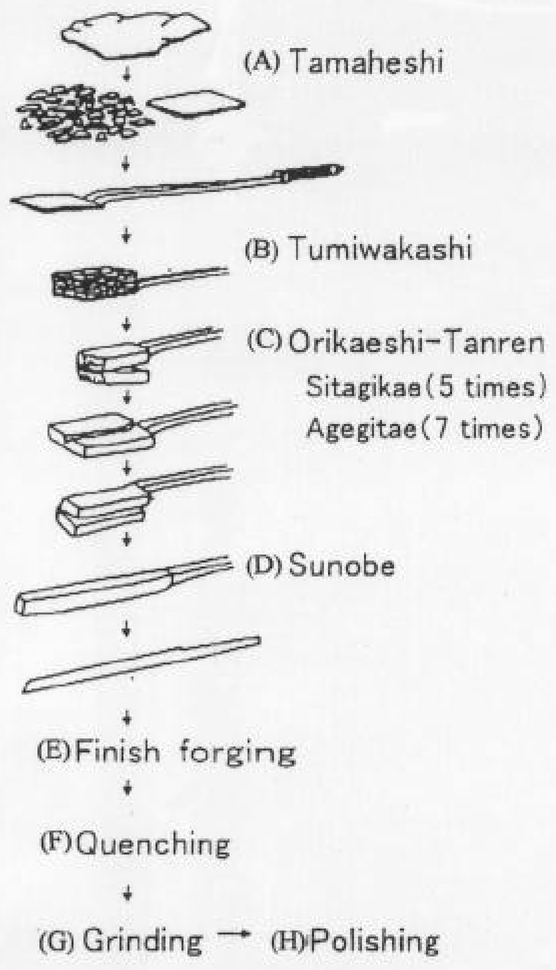


Fig. 5 Outline of swordmaking process

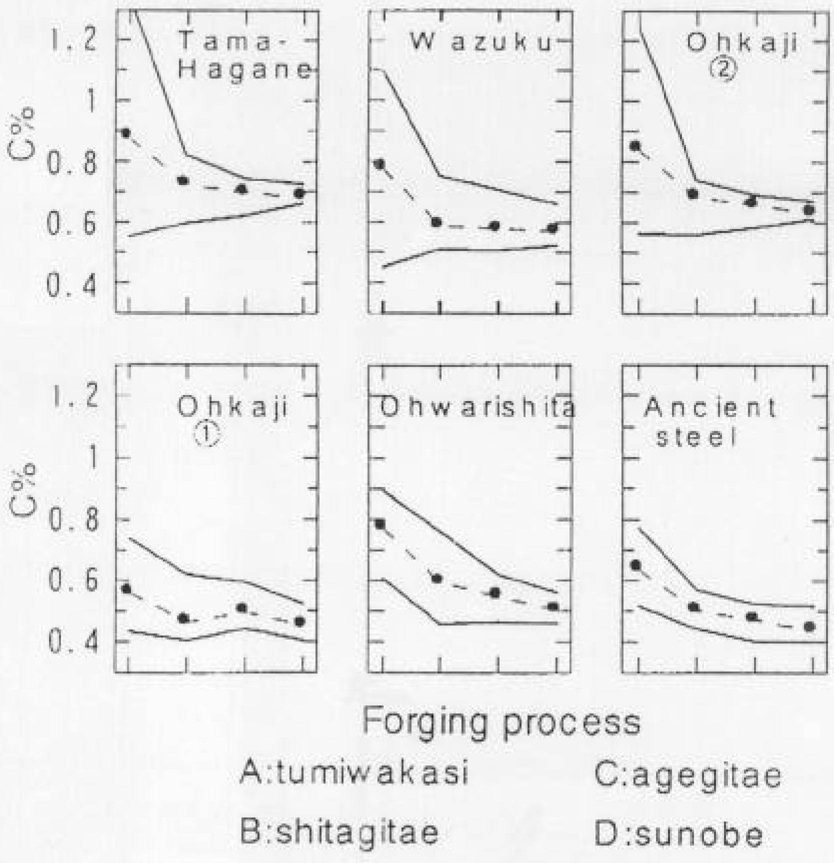


Fig. 6 Change of carbon contents distribution during forging

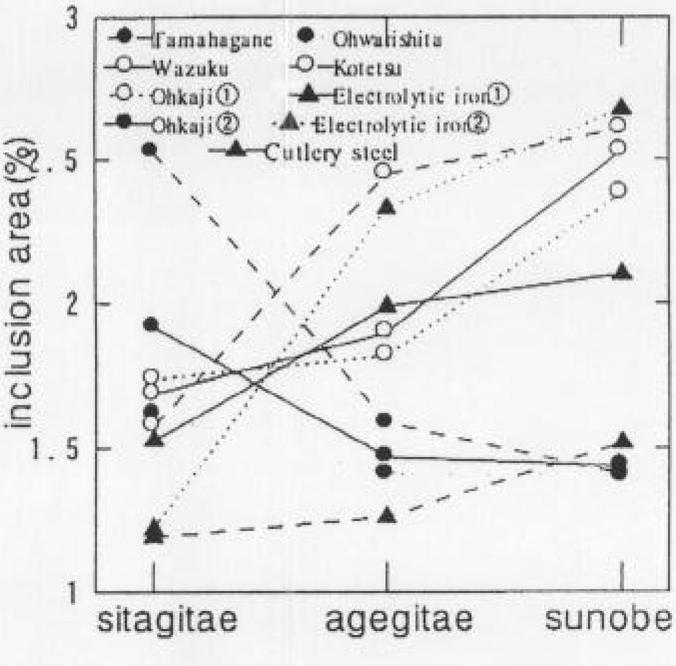


Fig. 7 Change of inclusion area during forging

Table 1 Common condition in Japanese traditional smelting

Reductant	Charcoal
Added flux	Nothing
Wall of Furnace	Soil (Consumable)

Table 2 Example of composition of iron sand (wt%)

	T.Fe	TiO_2	FeO	Fe_2O_3	SiO_2	Al_2O_3	CaO	MgO	P	S	V_2O_3
A Akome-1	54.56	6.82	18.48	51.08	14.90	4.98	1.60	1.74	0.032	0.036	-
B Akome-2	52.07	5.32	19.55	52.71	14.50	4.30	2.68	0.94	0.095	0.026	0.370
C Masa -1	59.00	1.27	24.72	64.45	8.40	2.34	2.34	1.54	0.064	0.009	0.258
D Masa -2	59.98	1.54	20.98	62.45	10.02	1.62	1.62	1.27	0.060	0.023	0.234

Table 3 Example of composition of products by Tatara process (wt%)

	C	Si	Mn	P	S	Ti	V	0
Pig iron -1	3.12	0.37	Tr	0.046	0.023	-	0.02	0.0131
-2	3.44	0.11	Tr	0.043	0.022	-	0.02	0.0176
Tama-Hagane -1	1.42	Tr	Tr	0.013	0.007	0.004	0.02	0.0115
-2	1.17	0.02	0.02	0.032	0.008	0.004	0.02	0.0267

	T.Fe	FeO	Fe_2O_3	SiO_2	MnO	Al_2O_3	Ca0	P	TiO_2
Slag -1	34.07	36.68	7.07	27.66	1.63	6.08	2.29	0.12	11.43
-2	49.52	58.85	5.40	22.52	1.23	5.40	0.18	0.02	5.10
-3	27.20	30.76	4.62	41.30	1.16	9.21	1.49	0.03	9.51

Table 4 Comparison of operational condition of iron making and direct steel making

	Iron making	Direct steel making
Sort of iron sand	Akome(TiO_2; 5~12%)	Masa(TiO_2; 1~2%)
Traveling time of iron sand	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	About 80 hours	About 55 hours

Table 5 Samples for swordmaking experiment (wt%)

Materials	Raw Materials								Snobe
	C	Si	Mn	P	S	V	Al	Ti	C
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 Material (A)	1.04	0.025	0.015	0.073	0.0088	0.040	0.006	0.010	0.37
Ohkaji Material (B)	1.00	0.025	0.005	0.046	0.0097	0.006	0.016	0.011	0.63
Ohwarisita	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
Electrolic 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