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CARBURISATION OF IRON IN ANCIENT INDIA

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Iron can be carburised by prolonged heating with charcoal or other organic matter at about 900°C. The absorption of carbon into iron is, however, very slow and it is very difficult for big objects. Moreover, efforts to carburise the whole of the object cause excessive carburisation of the edges resulting in brittleness.

The ancient smiths were wise enough to have developed the lamination technique as it could fulfill their requirements without having to carburise the whole blocks of iron. This technique was employed as early as 1200 B.C. for making iron implements. The technique was quite wide-spread. This technique imparted both toughness and flexibility due to the presence of carburised and wrought iron layers.

The carburisation process was well-developed around 600 B.C. Originally, the addition of carbon might not have been done deliberately but once its importance was realised, it might have become a popular technique.

Indian steel referred to as wootz was made by a process resembling the modern cementation or crucible process. Literary references indicate that the smiths had mastered this technique of making steel by carburisation of wrought iron in closed crucibles.

Bronson has sought to give credit of origin of wootz steel to some Arabian countries. This has been refuted by Prakash who has quoted from ancient Indian texts which give description of crucibles for making steels and classifications of iron and steel.

INTRODUCTION

The time when smiths were first able to introduce carbon into iron and control the working processes according to needs and specific uses must be very significant. Wrought iron derived from the bloomery hearth, used in antiquity, is soft and malleable while the introduction of carbon imparts its strength and hardness. Steel contains carbon which is much less than that in the cast iron. The process of making a perfect homogeneous steel must be difficult to control since the melting point of iron is 1540°C and it could not be liquefied as it must be extremely difficult to achieve this temperature.

How could have the carburisation of iron been achieved then? Iron can be carburised by prolonged heating above 900°C in contact with charcoal or other organic matter. Initially, the carburisation could have been unintentional during the iron smelting operation because of the increase in the ratio of fuel to ore. During reduction of iron ore to metallic iron in a fuel-fired furnace, carbon monoxide gas reduces the ore. If the fuel-ore ration increases above a certain level, the reduced iron from the ore dissolves carbon to the extent depending on the amount of fuel present. The product may contain carbon from nearly zero to four percent.

Alternatively, pure wrought iron recovered from the furnace can dissolve carbon if heated again to a temperature of above 740°C. This process can be controlled directly for the percentage of carbon to be added. At about 740°C, the reaction proceeds as follows:

$$3Fe + 2CO = Fe_3C + CO_2$$

Cementite (Fe_3C) immediately dissolves in the iron. Carbon monoxide gas transfers the carbon into iron and is the transfer agent except in molten systems. Carbon dioxide formed reacts with carbon in the fuel to form more carbon monoxide. The reactions are accelerated by the presence of small quantities of alkali carbonates. It is a slow process. The carburising temperatures above about 850°C are, therefore, used to obtain useful depth of carburisation[1].

Presence of iron carbide, Fe_3C confers increased hardness to the metal. Iron and iron carbide, also known as cementite are distributed in steel in layers. Presence of ferrite layers gives malleability to steel. The layered structure of steel is called pearlite.

The carburisation process seems to have been discovered accidently by the smiths during repeated and prolonged heatings of iron in the charcoal fire. The modern process involves heating of wrought iron or mild steel in a powdered mass of carbon-rich material and the surface of iron object is carburised to an extent desired by controlling the length of the treatment. The ancient smith by repeatedly heating the piece of wrought iron during forging in a charcoal fire and maintaining the temperature must have discovered this process.

The absorption of carbon into iron is, however, very slow and it is extremely difficult for big objects. Moreover, heating for sufficient time to carburise the whole of metal would carburise the edges excessively causing brittleness[2]. The ancient smiths must have probably understood the difficulties of diffusion of carbon into iron which resulted simultaneously in brittleness in high carbon steel. These difficulties might have led the ancient blacksmiths to develop a technique of introducing carbon into iron without melting it. This involved carburisation of several thin pieces of iron sheets, then piling them together into flat laminated slabs which possessed qualities equivalent to hard steel. This was the lamination technique which was adopted by our ancestors and used in the fabrication of iron tools and implements and weapons, etc. around the beginning of 1st millennium B.C.

First such objects on which metallographic studies were conducted were the iron objects recovered in the excavation at Dhatwa, an iron working site dated to 500-400 B.C. Samples were cut from the pointed end of a plough share, fragments of a knife and a chisel. Only the sample cut from ploughshare could withstand strain of polishing, the other two broke down.

Metallographic examination revealed that the ploughshare had a closely welded laminated structure. It could be inferred that the implements were made from the bloom in the three stages. First, the small bloom from the furnace was heated to redness in an open forgefire maintained at above 900°C with the help of a forced draught. During forging, the slag separated out and the surface of the metal was progressively carburised and casehardened. This imparted strength and hardness to the forged surface. The bloom was then beaten to a thin strip of metal. In the second step, a number of such strips were joined, one by one by welding, this form of welding in the present times is known as smith welding. Finally, the whole builtup mass of iron was further forged to shape it into a desirable and useful form. Hegde has suggested that the ancient Indian blacksmiths were probably well-versed in the technique of imparting hardness to the cutting edges by heating and quenching although he could not trace any such evidence due to cutting edges of the implements being heavily corroded[3].

Studies carried out by Agrawal, et. al. on iron objects from different excavation sites e.g., Komaranhalli and Tadakanhalli (1200 B.C. - 1000 B.C.) in Karṇāṭaka, Śṛṅgverapura (250 B.C. - 200 A.D.), Hulaskhera (700 B.C. - 700 A.D.), Bhāradvāja Āśrama (300 A.D.-600 A.D.), Jammu (600 B.C.-600 A.D.) and Kauśāmbī (PGW - 500 A.D.) from Uttar Pradesh have revealed the extensive use of lamination technique in ancient India. Very small cross-sections were taken from these artifacts, mainly implements, chisels, nails, knives, spears and spearheads. Artifacts from the early iron age (Circa 1000 B.C.) were mineralised with only some remanent metal in the core. Artifacts belonging to 600 B.C. - 700 A.D. had good metallic cores. Study of relic carbide in the mineralised metal at higher magnifications together with the micro-structure in the metal core helped in proper identification of fabrication technique. Examination of metal sections indicated that the metal was prepared by hammering and welding together the sheets of carburised and uncarburised iron. One sheet was carburised and the other was not. In this way, several sheets of iron and carburised iron could be piled alternately and forged into a flat slab. This technique of using alternate sheets of carburised and uncarburised iron provided an opportunity to the smith to save time, hardwork and materials required for carburisation of larger blocks of iron. Thus the effect of carburisation of few strips could be extended by forge welding these strips into soft wrought iron.

The artifacts from megalithic site Komaranhalli were in advanced state of corrosion and no metal core was present in many of the objects. Examination of a spear-head showed that the carburised layers had carbon content of 0.2-0.3%, whereas the uncarburised layers had a carbon content of about ~0.05%. The average micro-hardness of the carburised and uncarburised layers was 125 and 100 respectively. By using high magnification, relic carbide was detected. The artifact was fabricated by hammering and welding together the sheets of carburised and uncarburised iron sheets. There were no signs of tempering or heat-treatment. Other objects e.g., swords, spearheads and daggers were also found to be fabricated by the lamination technique.

Tadakanhalli is a megalithic site near Komaranhalli and is contemporary to Hallur which is dated to circa 1000 B.C. The objects from Tadakanhalli had been fabricated by the lamination technique. Ten objects consisting of spearheads, daggers, swords, chisel and axes were examined. In an iron axe, banded structure consisting of three carburised layers and four uncarburised layers was seen. The examination of darker and lighter colour layers revealed that the carbon content was 0.2 - 0.3% and 0.04 - 0.05% respectively in the carburised and uncarburised layers. There was no indication of the axe having been quenched or tempered.

Examination of artifacts from Śṛṅgverapura, Hulaskhera and Bhāradvāja Āśrama in Uttar Pradesh also showed carbon content in the range of 0.2-0.3% and micro-hardness of the carburised and uncarburised layers in the range of 125-135 and 100-105 respectively. Studies conducted on these artifacts showed that no attempt had been made for hardening and heat-treatment.

Examination of micro-structure of a section taken from a spearhead from Kauśāmbī revealed a piled structure produced by two layers in the metal. One layer was carburised and having a very low carbon content and the other was uncarburised, mainly of wrought iron having a ferrite structure. A section from the pointed end of the same spearhead revealed an air cooled dispersed pearlitic structure having carbon content 0.6% and microhardness of 216. Examination of other objects showed that the lamination technique was used in fabrication of weapons and tools and that smiths had good knowledge of carburisation with the ability to produce homogeneous medium and high carbon steel around 400-500 A.D.

The investigations of iron artifacts from Karṇāṭaka and Uttar Pradesh have shown that these artifacts were made by the lamination technique. Smiths used the metal which was extracted by the direct process from the iron ore. Pure iron was made from the bloomery iron by repeated forging at high temperature (1000-1100°C). Iron sheets were then carburised by directly putting them into charcoal fire for sometime. The process is known as cementation, carburisation or case hardening. Excess of carbon would result in brittleness. The excess carbon was removed by repeated heating and hammering. All the metal pieces of carburised and uncarburised iron were placed together, forged and folded. The final folding of the pile was done in such a manner that the carburised layer must appear on the surface as well as inside the pile alongwith the uncarburised layer. These small piles were then joined together and worked below the critical temperature to impart shape to the object. The presence of carbon makes the object tough whereas pure iron gives good flexibility to withstand the sudden impact. The technique thus enabled fabrication of the objects which had both internal and external strength.

An axe from Jajmau in Uttar Pradesh had high carbon content and it was found to be quenched, tempered and cooled in air besides the laminated structure to give it extra toughness at the edge. The micro-structure showed that the axe was made up of two high carbon layers sandwiching a low carbon layer. The high carbon layer had a tempered structure near the cutting edge while the low carbon layer had a pearlitic structure. The axe was thus tempered after forging the layers. The examination of some more objects e.g. arrowheads, spearheads, nails, daggers, blades and sickels from this site pointed to the wide use of lamination technique at this site[4].

Chemical and spectroscopic examination on six objects from Rājghāt excavations (600-400 B.C.) carried out by Bhāradvāja revealed that these objects had the carbon content 0.12 to 0.42%. The low percentage of carbon suggests that these objects might have been naturally carburised during forging. One object contained 1.1% carbon and was apparently intentionally carburised[5].

Metallographic studies were carried out by Hari Narain, et. al. on four iron objects belonging to 600 B.C.-300 A.D. from Atranjikhera in Uttar Pradesh. Micro-structure in two implements consisted mainly of pearlite with some ferrite. These were completely mineralised with very less residual metal. Ferrite structure with pearlite and relic carbide structure homogeneously distributed in the residual and mineralised metal was revealed in the celt. Carbon content was around 0.5 to 0.6%. The artifact was forged from carburised wrought iron and after getting the desired shape was rapidly cooled in air. In the chisel heterogeneous carbon concentration (pearlite with ferrite streaks and mainly pearlite) of 0.5-0.8% in the residual metal was found. There was heavy carburisation near the cutting edge.

The micro-structures of house-hold artifacts showed mainly ferrite structure with a trace of carbon. The carbon content was 0.04 - 0.09%. The small amount of relic carbide in one object indicated that attempts were made to carburise the wrought iron to some extent[6].

It can be seen that the lamination technique was employed as early as 1200 B.C. for making iron implements. Since the implements made by this technique have been discovered from different parts of India, it can be said that this technique was quite wide-spread. This technique, however, seems to have been much earlier in vogue in the South. The technique enables the fabrication of objects which were tough and offered flexibility as well due to the presence of carburised and wrought iron layers. The technique involved introduction of carbon inside iron without achieving high temperatures and without keeping iron in the hearths for longer periods. After the introduction of carbon, several sheets of wrought iron and carburised iron could be laminated into one place for the fabrication of the artifacts. We can say that the smiths were intelligent enough to have understood the difficulties of carburisation of larger blocks of iron and to have developed this technique. The technique was cost-effective as the whole of iron was not required to be carburised.

The technique of carburisation was fully developed by 600 B.C. as is evident from the examination of objects of this period from Rājghāt and Atranjikhera.

After the ancient blacksmiths had understood the effect of alloying of carbon with iron, he must also have gradually understood that the iron-carbon alloys formed by the carburisation process were not homogeneous and did not possess the qualities required of them due to the presence of cementite needles and slag inclusions. These shortcomings would have been minimised with the advent of wootz steel making process[7].

Wootz Steel: There are reference in literature of the excellent quality of Indian steel as early as 4th Century B.C. Ktesias, who was in the court of Persia in the 5th Century B.C. mentions two remarkable swords of Indian steel presented to him by the king of Persia and his mother. Numerous stone inscriptions of Emperor Aśoka suggest the use of fine and sharp steel implements in India in the 4th Century B.C. Quintus Curtius mentions that Alexandar the great, after defeating King Porus (326 B.C.) received from him a gift of 100 talents or 30 lbs. of steel.

It can not be said with certainty as to when and where the wootz steel was first made in India. It was perhaps produced at several places like Mysore, Salem and Hyderabad. Cakes of wootz were exported for the manufacture of Damascus blades. This had unique properties of hardness and malleability. The Damascus swords attained reputation for their flexibility, strength, sharpness and beautiful patterns on the surface of the sword[8,9].

Tylecote says that an interesting Indian development was the melting of steel in crucibles in which a homogenous high-carbon steel with carbon content ranging from 1 to 1.6% was said to have been produced by the cementation process. He believes that this steel, later in the medieval periods was known as wootz steel. This was also known as Damascus steel because it was exported to the West Asian countries and from there to the West[10].

Wootz steel was produced in ancient India by a process resembling the modern cementation or crucible process. However, the process had two peculiarities - the carbon was introduced into the crucibles in the form of wood and wootz steel so produced had 'damask' pattern. Wrought iron was first made by direct reduction of magnetite ore. Wootz was produced from carburisation of wrought iron which was heated in closed crucibles with dry wood chips, stems and leaves of plants over charcoal fire maintained by blowing air with large bellows. The operation took 4-5 hours to complete. Steels so obtained was heated again so that the excess of carbon was burnt off. Sometimes, the hot metal was quenched by pouring hot water over it. Steel was produced by this traditional method at some places even in the 18th and beginning of the 19th century. This gives credence to the use of crucible process in early periods, as Ray believes, that this process was the continuation of the original process which was acquired by the ancient smiths even before the beginning of the Christian era[11].

Bronson has sought to give the credit of origin of wootz technology to some Arabian countries in view of lack of any concrete evidence on the basis of metallographical studies of Indian wootz steel. He does not agree that Indian steel was made in crucibles during the early periods or that it had outstanding quality. Bronson argues that no Indian steel that might have been made in some way other than by the carburisation or case-hardening of low-carbon bloom and by pile-welding of layers of iron thus hardened have yet been identified. He says that there are no evidences to suggest that crucibles were used in steel-making before the late first millennium A.D. and there are no reasons to think that crucibles were used in India even during this period[12]. Prakash has taken strong exception to this and quoted from his research on Āyurvedic medicine that Caraka, the father of Indian surgery and Āyurvedic medicine (Ca 700 B.C.) designed over 20 types of surgical instruments and gave details of carburising and hardening procedures for each. He has also quoted from Rasaratnasamucchaya which gives a detailed description of composition of a crucible and of classifications of iron and steel. Prakash, however, agrees that archaeological evidence to support this has not been found and written records on the making of wootz steel are not available, mainly because this art passed from artisans as family secrets learned by practice only[13].

Hegde has also pointed out that there are no evidences of homogenous steel being found from any of the ancient Indian sites excavated and examined so far[14].

Bronson has said that crucible steel making might have originated in India but literary descriptions available would ascribe wootz process to Arabs and Persians. Until more archaeological and metallurgical date become available, Bronson has preferred to reserve the judgement about the antiquity of wootz[15].

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