The 2006 international symposium on Ancient Metallurgical Experiments

In September 2006, the archaeological open air museum in Eindhoven (NL) hosted an international workshop, the 3rd in a row after 2 previous ones at locations in Germany. 90 craftspeople, artists, experimental archaeologists and metallurgists from 15 countries gathered for a full week of (iron) smelting (15 furnaces) and (bronze) casting (5 teams). In the evenings, presentations were given while the light night from the furnaces lit the sky. Blacksmiths were forging axes and art. 2,500 kilos of charcoal were consumed during the week. Another result of this workshop were the numerous new contacts which led to annual follow up workshops. The paper by Dr Espelund as presented here was given in an older version on one of the evenings in Eindhoven, the Weker et al. paper is based on experiments carried out during that week.

Experimental metallurgical process in a slag pit bloomery furnace

The article describes attempts to recreate iron smelting process used in the Poland region during the period of the Roman Empire, using archaeological evidence from Holy Cross-Mountains.

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1. Introduction

For the past 40 years many major Polish research centres have undertaken studies on the oldest traces of metallurgy found in Poland. Two large centres of ancient metallurgy were found. They functioned in the period heavily influenced by the Romans. Those two centres were:

- the Masovian centre
- the Holy Cross-Mountains centre

Apart from these two centres, there were smaller metallurgical areas in Poland, located in Silesia, in the region of Opole, Brzeg and Wołów. The first study of slag in the region of the Holy Cross mountains was initiated by Professor M. Radwan and systematic archaeological investigation began in 1955 [1]. The study of collected materials (slag blocks, parts of wall of the smelting furnaces), the steel made at that period, numerous experiments and the modern metallurgical knowledge have enabled the reconstruction of the furnaces and the metallurgical process itself dating back to the period under the Roman influence [1,2].

The furnace typical of Holy Cross-Mountains region: the slag-pit furnaces had a cylindrical shaft which was 1.2 to 1.5 m high and an internal base diameter of approximately 0.5 m. The shaft of the furnace was built over a pit in the ground.

The shaft was used to supply in turns portions of iron ore and charcoal which acted as the fuel. As the charcoal was burning, the subsequent layers of the charge moved down into the zones of higher temperatures. The result of this was the disintegration of the minerals and iron compounds which the ore contained.

$$FeCO_3 \rightarrow FeO + CO_2$$

Then the ferric oxides undergo gradual reduction to pure iron.

$$Fe_2O_3 \rightarrow Fe_3O_4 \rightarrow FeO \rightarrow Fe$$

Carbon and carbon monoxide which were produced during the combustion of charcoal served as the reducing agents. The following reactions took place simultaneously:

$$FeO + CO \rightarrow Fe + CO_2$$

$$CO_2 + C \rightarrow 2CO$$

(so called *Boudouard's reaction*)

The molecules of reduced iron stick together creating at first small spherical particles and then a spongy lump of iron (also known as iron bloom).

The low temperatures in the furnace (approx. 1100 - 1300 °C) do not allow the melting of the metal. In these temperature slag can be melted and may flow down and fill the underground part of the furnace. The iron bloom would be suspended, and then stick to the walls of the furnace and stay at the level of the nozzles. After breaking off of the upper part of the furnace the iron bloom was removed and subjected to further treatment. By means of delicately removing pieces of slag and forging, the contamination produced during the process could be successfully removed. The final product came in the form of a compact piece of iron which could be used for manufacturing other products. This model was widely accepted and has been applied to this day [2].

Many experimental processes were carried out on the basis of this model. Despite this, the efforts to get the slag block and the iron bloom comparable to those found in the archaeological excavations did not succeed fully. In the experiments in which different kinds of ore were used the researchers succeeded in obtaining pieces of metal that were of different sizes. Yet after getting them out of the furnace it turned out that they contained carbon to an extent that forging was impossible. The pieces had the characteristics of cast iron. Not even once did the researchers succeed in obtaining a piece of slag that would have a smooth surface and that could be separated from the iron bloom just like it was done in ancient times. The observations of the series of experiments and their results lead us to claim that despite the efforts made by many researchers and the

■ **Fig. 2** View of the second bloomery furnace

■ **Fig. 1** View of the first bloomery furnace

knowledge of modern metallurgy the above mentioned model is not sufficient. During the experiments many questions arose to which the answers have not been found. The results of those experiments are still to a large extent a matter of coincidence.

During the symposium, Ancient Metallurgical Experiments in Eindhoven the researchers carried out two smelting processes. The furnaces were based when it came to shape and dimensions on the ancient type of furnace typical of the Holy-Cross Mountains region (a shaft furnace with a pit). The controlled process parameters included the draught of air, weighted amounts of ore and charcoal and time.

The products of chemical reactions - slag and pieces of metal that were obtained during the experimental melting process were then analyzed in the laboratories of AGH University of Science and Technology in Kraków (Poland).

2. The course of the melting

2.1. The construction of the ancient furnace

The two bloomery furnaces were building according to models reconstructed by M. Radwan and K. Bielenin on the basis of their long archaeological and technical experience [1]. The dimensions and the shape of both furnaces were similar. They differed in terms of height and the construction of the pit. The way they looked is presented in fig. 1 and 2.

The pit of the furnace was dug in the ground. Its diameter in both cases was 35 cm and its depth 30 cm. Its internal surface was smoothed using clay dissolved in water. The shaft part situated above the pit was made from previously prepared and dried bricks. They were formed from a mixture of clay and straw. The bricks had the shape of cuboids and their dimensions were 15 × 11 × 8 cm. During the building process the shape of the bricks was adjusted by means of cutting the bricks' edges in order to obtain a circle.

Clay dissolved in water served as the mortar. The height of the shaft was 100 cm and in the case of the second furnace 110 cm. The internal diameter on the level of the pit was 35 cm and in the upper part 20 cm in the case of the first furnace and 18 cm in the case of the second. At a height of 12 cm above the ground level, two holes were made on opposite sides of the furnace. They served to suck in air during the process of drying the furnace.

When the drying was complete and before the bloomery process actually began the holes were covered. A tuyére, a steel pipe with a diameter of 5 cm was put in one of the holes. Using blacksmith's bellows a draught of air was directed through the pipe. Pieces of beech wood were put vertically inside the pit. The pit filling it very tightly.

2.2. The course of experimental melting

The first furnace was dried with the use of charcoal and natural draught. As the charcoal burned down a new amount was added. The drying process took 20 hours. Afterwards the final warming of the shaft began. In order to do so the entire space inside the shaft was filled with charcoal and artificial draught was applied using the blacksmith's bellows. The intensity of the operating of the bellows was adjusted to the speed of the moving charcoal in the shaft of the furnace as well as to the external temperature (the colour) of the furnace walls. After about 6 hours a proper temperature needed to begin the smelting process was finally obtained. From this moment the iron ore was added.

Bog iron ore prepared by the organisers of this experiment was used. Before putting it into the furnace the ore had to be specially prepared. The preparing process consisted in putting it into the fire first. In order to do so a metal plate with the ore on it was put over the fire. After an hour's heating, the ore was crumbled with a hammer into pieces similar of walnut size.

Portions of iron ore and charcoal were inserted in turns. The portions were about 0.5 kg. Proportions of iron ore to charcoal 1:1. The time when a new portion was inserted depended on the speed of the processes taking place inside the furnace and the speed of settling of the materials (i.e. their level). The researchers tried to keep a steady level of the charge in the shaft part of the furnace so that it would not go down by more than 15-20 cm from the upper edge of the shaft. In total 38 kg of iron ore and 40 kg of charcoal was used. Once the adding of the ore was over, the artificial draught of air was stopped and the nozzle removed from the steel pipe of the furnace. At the same time a hole was made in the lower part of the shaft on the opposite side. From that time on the furnace worked using only the natural draught of air, until the charcoal had burned down totally.

After a 10 hour break to allow the furnace to cool, the process of dismantling began. When the upper and middle parts of the shaft were removed, one discovered a slag block weighing approximately 12 kg as well as pieces of iron weighing at together 4 kg. **Fig. 3** shows the achieved slag block.

The second smelting process was carried out in a furnace of different construction. There was a sort of a vault between the pit and the shaft. It was made of clay and wooden sticks which served as the structure. A hole of 15 cm in diameter was made in the middle part. **Fig. 4** shows such a vault.

The drying and warming of the pit and the shaft of the second furnace took place in a similar way to the first furnace and took approximately 36 hours. The way the ore was prepared was changed. Bigger portions were used this time (approx. 7.5 kg). They were added when the level of settling charcoal showed that the previous portion of ore was already in the zone of the highest temperatures. The portions of ore were added every 50-60 min. on average. In total 38 kg of ore and twice as much charcoal was used. The reduction process took 5 hours. Once the adding of ore was over, the artificial draught of air was stopped and the furnace worked using only the natural draught of air until the charcoal burnt down. At the end of the process the shaft was covered with a metal plate for 30 minutes. After a 5 hour break the dismantling of the furnace began. It turned out that there was only a small amount of slag in the pit and that the hole in the vault was sealed by slag. The rest of the slag was found in the lower part of the shaft. A clearly separated fraction of reduced iron i.e. an iron bloom was not found. Instead iron had formed as separate small spherical pieces that could be found in the mass of slag.

2.3. The obtained results

The experimental smelting processes that were carried out enabled the comparison of slightly different constructions of the furnace and different technologies of putting the charge into the furnace. During both processes the same bog iron ore and charcoal were used and the atmospheric conditions were the same. A detailed measurement of temperature, composition of chemical gases and the doses of air was not carried out.

The process parameters were controlled by means of observation only the external phenomena connected with the process taking place inside the furnace: the colour and type of the flame, physical changes of the external surface of the shaft, the speed of settling of the charge.

As can be inferred from the observations the temperature in both furnaces in the region of nozzles exceeded 1100 °C. The fact that the flame of the gases coming out of the shaft was blue meant that there was an excess of carbon monoxide (CO) inside the furnace. Such conditions enabled the process of reduction of ferric oxides to pure metal. During the entire process the speed of settling of the charge decreased only at the end which may suggest that the process could be prolonged.

After dismantling the first furnace it turned out that in the upper part of the slag block is a three-dimensional structure containing a lot of iron which did not have the characteristics of a homogenous iron bloom (**Fig. 5**). The small pieces of iron were very fragile – every attempt to forge them in cold temperature failed. The process did not live up to the expectations of the researchers. Nevertheless a bigger piece of iron was chosen to be forged again. It was forged on a metal anvil after it had been heated in the blacksmith's hearth. The operation of forging was carried out slowly, hammering the piece using little energy. After a multiple cycle of heating and forging a cube showing all the characteristics of metal was obtained (**Fig. 6**). The forged cube was then subjected to metallographic analysis. The obtained slag block and the iron structure (the iron bloom) confirm that the reduction zone of the ferric oxides was in the direct vicinity of the nozzles - slightly above the inlet for the air. In this zone the temperatures were the highest and the conditions for the reduction to take place were best.

During the melting in the second furnace only small spherical pieces of iron were obtained. They were unsuitable for forging and heating by means of simple blacksmith's methods. It turned out that the slag block had filled the lower part of the shaft and probably if the process had lasted longer the zone of charcoal combustion would have moved up the shaft. Such conditions would not be favourable for total reduction.

■ **Fig. 3** View of achieved slag block

■ **Fig. 4** View of the vault between the pit and the shaft

■ **Fig. 5** View of achieved slag block in II test

■ **Fig. 6** View of the small pieces of iron forging process

In the case of both furnaces the amount of obtained products (slag and iron) is clearly lower than the amount of the ore used. A more detailed study of materials used cannot be done. It would require a special preparation of the process. A general analysis suggests that a significant loss of iron takes pace due to its oxidation in the upper part of the shaft.

After removing the slag block and a total dismantling of the shaft, the pit was investigated. It turned out that in both cases there was only charcoal in the pit. In both cases the attempt to obtain a pit filled tightly with slag failed. During the process no metallurgical flux was used in order to liquify the slag. In both cases the visually measured temperature in the area of nozzles was similar. It means that the way of putting in the charge (small or big portions) does not influence the process in any major way.

The extent to which the heated ore was crumbled (pieces having dimensions of 2-3 cm) turned out to be appropriate. Small pieces of the charge make the flow of the gases harder. Big pieces, on the other hand, enable a quick flow of the gases. In both cases the possibility of the reduction of the iron ore is limited.

The presence of reduced molecules of metallic iron (even if in small amounts) means that the metallurgical process is similar to that taking place in the ancient furnace but to a smaller extent. The reconstruction of the entire process requires further research.

3. The results of metallographic investigations

3.1. The object and the range of research

Samples of the mass containing both iron and slag that were obtained during the experimental melting process in the first furnace constituted the object of research. A piece of the slag block served as one of the samples. The second sample was a piece of the slag block that was subjected to forging in high temperatures and heating in the blacksmith's hearth.

The sections of the samples were subjected to standard metallographic procedures. They were then observed under an optical microscope - AXIO Imager produced by Zeiss. The samples were also studied by means of a scanning microscope Hitachi 3500N with a Noran EDS analyser. Structural observations and chemical microanalysis concerned the iron phase and the slag phase. Slag was subjected to an X-ray diffraction phase analysis using a HZG4 diffractometer.

3.2. The results

Fig. 7 shows the microstructure of the sample containing iron and slag etched by means of Nital. A Ferritic structure is visible. It is coarse-grained and it contains nests of slag phase. The iron was strongly contaminated by non-metallic phases. The slag phase (**Fig. 8**) was not homogenous in terms of structure. In the grey background one could notice dispersed bright dendrites, having equal axes and powdery grains which had the morphology of an eutectic. The above mentioned ingredients could be found in large clusters.

The grey part was the phase from Fe-Si-O and the bright dendrites as well as bright grains were the phase from Fe-O. The above mentioned phases were identified using X-ray diffraction analyse show a fayalite matrix Fe_2SiO_4 and bright dendrites and grains wüstyt FeO. The presence of Fe_α was also noted. There was neither silica SiO_2 nor mullit $Al_6Si_2O_{13}$ which would be found in the authentic ancient slag.

Forging of the block containing metal and slag led to significant changes in the morphology of iron and slag phase. The network of iron clusters could be observed at the iron grain boundaries. The microstructure of forged, etched iron was shown in **Fig. 9 and 10**. A large diversity of structure in the sample section can be noticed. Ferritic structure, ferritic–perlitic structure, perlitic structure and martensitic structure can be seen. There is a structure resembling a triple phosphide eutectic $\alpha Fe\text{-}Fe_3P\text{-}Fe_3C$ at the grain boundaries.

3.3. Conclusions made on the basis of metallographic

What was obtained in the process of experimental melting of the iron and slag phases was a conglomerate. There were significant differences in the iron microstructure between the sample combining iron and slag and the sample obtained by means of forging and heating. This means that the products of chemical reactions taking place during the experimental smelting processes are not homogenous. Iron in the sample that did not undergo forging had ferritic structure with slag phase elements. Slag had the phase composition and a structure typical of ancient fayalite slag.

Multiple forging of the sample in high temperature together with heating it in the hearth resulted in partiall removal of the slag phase. Its remnants could be found in the spaces between iron grains. The forged sample is different in terms of phosphorus which separated the slag phase network and at the grain boundaries of iron phase. The microstructure of this last phase shows the local presence of carbon-enriched iron (to the eutectoidal composition). Martensitic structure came into being probably during a quick cooling of the sample once the forging was over.

The microstructure of the obtained iron in the forged sample is analogous to the microstructure of gromps – the remnants of ancient smelting results which were found during archaeological excavations. A detailed original description of the forming conditions of gromps can be found in the works of Z. Kędzierski and J. Stępiński [3].

4. Conclusion

Two smelting processes were carried out an experimental bloomery furnaces known as Holy Cross-Mountains slag-pit furnaces. Researchers attempted to reconstruct the technological process of iron smelting dating back to the Roman times. Thanks to making both smelting processes different in certain respects it became possible to see if structural features of the furnace and the technology of loading the charge can influence the process itself and its results. In both cases slag phase and small amounts of metal phase were obtained. Both the slag and metal mass differed, however, from the traces found during archaeological excavations. In both smelting processes the attempt to obtain a slag block that would fill the pit failed.

The metallographic investigation of the slag and metal phase shows that the products obtained during the smelting processes had chemical and morphological composition similar to ancient products. What can be inferred is that in the respect of metallurgic process the experimental smelting was quite similar to the ancient one. A complete reconstruction of the process requires, however, further experimental research.

Bibliography

1. *Bielenin K.*: Starozytne górnictwo i hutnictwo żelaza w Górach Swiętokrzyskich, 1974, Warszawa-Kraków, in Polish
2. *Pleiner R.*: Iron in archaeology, the European bloomery smelters, 2000, Praha
3. *Kędzierski Z., Stępiński J.*: Metallographic analysis of iron from the Roman period fund in Poland, 50 years of research on ancient metallurgy of the Swiętokrzyskie Mountain Region, 2006, Kielce, Poland, in Polish

Summary

Experimentelle metallurgische Prozesse in einem Schmelzofen mit Schlackengrube

Experimentalarchäologische Schmelzprozesse wurden mit der Absicht durchgeführt, einen Vergleich von leicht unterschiedlichen Ofenkonstruktionen und verschiedenen Methoden zur Platzierung der Ofenladung zu ermöglichen. Diese Arbeiten geschahen vor dem Hintergrund, die technologischen Prozesse zur Eisenherstellung während der Römischen Kaiserzeit zu rekonstruieren. Die kontrollierbaren Parameter bezogen sich dabei auf den Luftzug, die gewogenen Mengen von Erz und Holzkohle sowie die Zeitdauer. Die produzierte Schlacke und das Metall wurden anschließend im Labor analysiert. In beiden Öfen war die Menge der Produkte sehr gering; der Versuch , eine dicht mit Schlacke gefüllte Grube zu erzeugen, schlug fehl. Auf der anderen Seite zeigte die metallographische Untersuchung auf, dass die produzierte Schlacke und das Metall eine chemische und morphologische Zusammensetzung aufwiesen, welche den archäologischen Objekten weitestgehend entsprach.

Métallurgie expérimentale en haut-fourneau

Deux procédés de fusion du métal ont été expérimentées pour retrouver les techniques de l'époque romaine et voir si la structure même du fourneau et la façon de le charger en combustible influencent le processus de fusion et ses résultats. Dans les deux cas, des scories et des coulées de métal ont été obtenus et aucun ne présentait l'aspect ou la masse des traces trouvées sur les sites archéologiques.

L'analyse métallographique des scories et du métal ont pourtant démontré qu'ils avaient une composition morphologique et chimique similaire aux vestiges anciens, donc le procédé de fusion a bien dû être proche des techniques romaines, mais il est indispensable aujourd'hui de recommencer les expérimentations.

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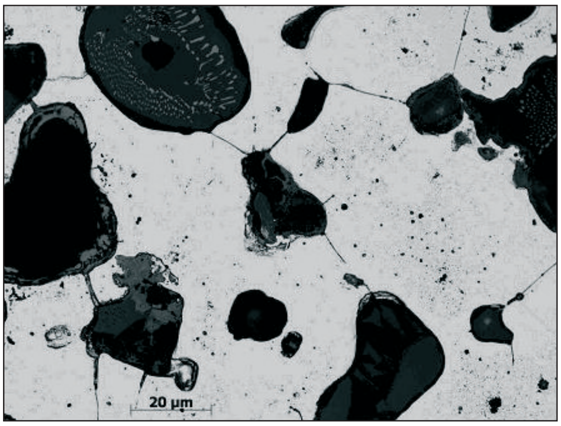
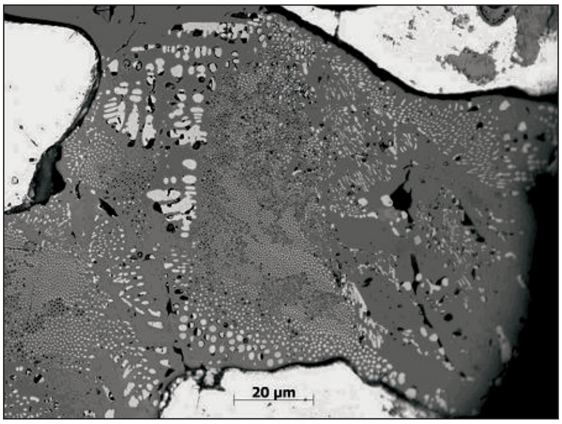
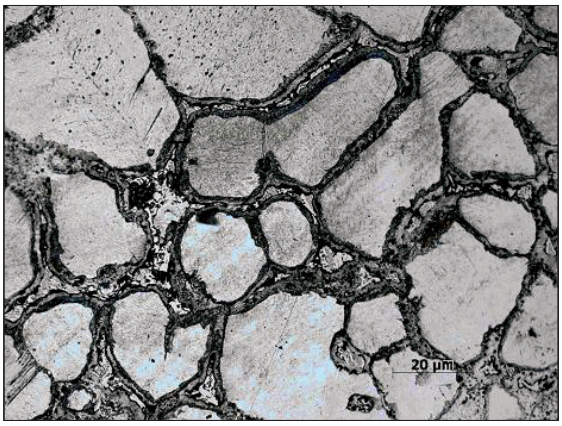


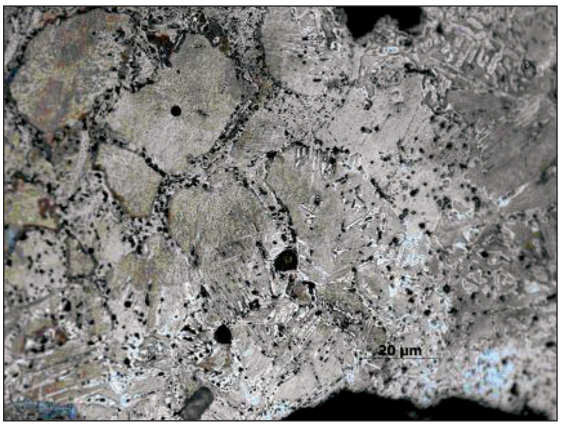
■ **Fig. 7** Microstructure of slag – iron mixture



■ **Fig. 8** Microstructure of slag phase



■ **Fig. 9** Microstructure of iron after forging



■ **Fig. 10** Microstructure of iron after forging