The metallurgy of Chinese cast iron statuary

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ABSTRACT: A group of 19 Chinese cast iron objects, mainly statuary and ritual objects, dating from between the 8th and 19th centuries AD, has been subjected to microstructural and compositional analysis. The objects were found to represent a range of cast iron types, including white cast irons with both divorced eutectic and ledeburitic microstructures, ferritic and pearlitic grey cast irons, and mottled cast iron. The microstructure of each object was found to be explicable by the chemical composition of the iron, especially its silicon, phosphorus and sulphur contents. The different chemical compositions can be accounted for by different smelting conditions, such as the use of coke or charcoal as furnace fuel, and the smelting of iron ore from different ore bodies. The sulphur contents, while not providing definitive evidence, suggest that at least some of the cast irons were smelted using coal or coke rather than charcoal. Where it was possible to tell, specifically for the white and mottled irons, the objects had been cast using the piece mould process. This work increases appreciably the corpus of analytical results for Chinese iron of this time period.

Introduction

Cast iron, the carbon-rich liquid product of iron smelting furnaces, is often thought of as a relatively recent material in comparison with wrought iron, its lower-carbon ferrous cousin whose first appearance dates from at least as far back as the mid-second millennium BC. In a Western context this is probably justified although sporadic finds of cast iron, possibly discards, have been found at a few early sites (see, *eg*, Burton Brown 1950) including some early bloomery iron smelting sites. It is now believed that in Medieval Europe there is likely to have been a gradual development that culminated in the *stuckofen* and ultimately, *c* 1000 AD, in the blast furnace (see, *eg* Pleiner 2000). For the most part, the cast iron (pig iron) product of early European blast furnaces was intended for conversion into wrought iron by a fining process, with the manufacture of cast objects a secondary and significantly later development. However in China liquid iron was being produced from blast furnaces very much earlier, certainly no later than the mid-first millennium BC and possibly as early as the 9th century BC (see *eg* Wagner 1993, Han Rubin 1996, Craddock 2003). Although it appears likely that some pig iron was being converted into wrought iron from early times, there is no question that functional castings were also being produced, so that by no later than the third century BC the casting of iron objects was a major industry in China (Bronson 1999). The rapid development of such an industry was undoubtedly aided by China's long experience with the production of high quality bronze castings, which dates back to the first half of the second millennium BC. This expertise could be readily applied to cast iron, which melts at temperatures not far above that of bronze.

Chinese iron castings were made for ceremonial and utilitarian purposes. Examples include agricultural implements (hoes, spades, plough-shares), weaponry (spear heads, daggers), salt evaporation pans, coinage, hardware (fittings, locks), tools (axes, hammers, saws), stoves, cooking vessels (pots, cauldrons, woks), lamps, bells, ornaments and structural segments of pagodas (see, *eg*, Hartwell 1962, 1966, Rostoker *et al* 1983, 1984, Wagner 1993, 2000, Bronson 1999, Craddock *et al* 2003, Wayman and Wang 2003, Chen Jianli *et al* 2003, Wagner unpublished manuscript). A major application was for statuary, which ranged from tiny objects to monumental statues weighing tens of thousands of kilograms. The present contribution deals with small cast iron statuary and other ritual objects.

Many workers have considered the role of iron in life in early China (see, *eg* Sung Ying-hsing (trans)1966, Bredt 1985, Rawson 1993, Till and Swart 1993, Paludan 1994). Iron in China was seen as a symbol of strength, determination, integrity and justice. Ann Paludan (1994) argues that in classical Chinese sculpture the choice of material was often as important as the choice of subject in conveying a message or achieving a desired result. A bronze object suggested power, jade implied purity and stone was associated with the tomb and immortality. Iron was a latecomer in the sculptural field and its particular associations are less easy to decipher. Furthermore, statuary, including statues of animals such as those represented among the objects analysed here, had symbolic importance as well. For example, since the earliest times, oxen had been associated with water and had come to symbolize water control. An ox was the most powerful beast at Chinese man's disposal. Its strength was invaluable in clearing the ditches and canals on which the water system for irrigating and food production depended. Iron statues of oxen or buffalo were therefore placed on the banks of lakes and rivers to prevent floods or in front of Daoist temples associated with water deities. The role of the famous iron oxen at Pujin bridge, dated to the Tang dynasty, are therefore not merely practical: their weight provided the necessary anchor for the chains, but such anchors in the form of oxen would also help to control the river. Chinese attitudes towards statuary were dominated by the belief that a statue contained the power to influence its surroundings. By making a statue, one brought into existence not only the actual powers of the subject but also its symbolic or assumed powers.

By the Tang and Song periods (AD 618–1279) master ironworkers were very skilled at founding massive iron objects using clay moulds. These castings include large Buddhist statues and bells and even pagodas and buildings. The vast production of the Song iron industry was probably prompted by pressure from peoples to the north, which meant a great increase in the armaments industry and an expansion of agriculture to maintain the economic base necessary for the raising and supporting of a large army.

A few large Buddhist iron figures and monuments have survived from the Tang and Five Dynasties (AD 907–960) periods but it was not until the 11th and 12th centuries that practical considerations led to an accelerating use of iron as a substitute for bronze. A dramatic increase in trade during these centuries meant that the demand for coinage was exhausting the supply of bronze. At the same time, improvements in smelting techniques meant that iron of reliable quality was easier to produce. This led to the use of iron for subjects such as temple guardians, which would have traditionally been cast in bronze.

Ceramic replicas in miniature of the essentials needed to accompany a dead person to the afterlife had commonly been placed in tombs from the Zhou dynasty onwards. The Chinese tended to see the afterlife as a continuation of their life on earth and so, if you had needed oxen, horses, and other animals in this life, then you would continue to need them and you took them with you in model form. Crude castings of animals in iron seem to have been common in the Song and Yuan periods, though many of these models may have been covered in gesso then painted to hide the traces of mould joints in the metal objects.

It would appear that many moulds were meant to be carefully disassembled rather than broken from the finished casting, which raises the possibility that the *luohan* and guardian figures may have been produced in series like ploughs and agricultural implements. Many of these iron figures would seem to have been made in sets. These *luohans*, judges of hell, guardian kings, were normally arranged around the main cult image or in the subsidiary halls of a Buddhist temple. Smaller religious figures were probably used in the domestic setting on a family altar in the home.

Although there were symbolic reasons for the use of different materials, different applications called for different physical and mechanical properties, and hence different microstructures, and the early Chinese ironworkers were able to produce different types of cast iron to fulfil the service requirements. These types included white cast iron (white iron) and grey cast iron (grey iron) as well as the intermediate state, referred to as mottled cast iron (mottled iron). Cast iron types will be explained in the Discussion section below. Chinese ironworkers also learned how to process cast iron so as to improve its properties, producing malleable cast iron and even steel by various specific heat treatments.

In the present work a selection of 19 cast iron objects from the collections of the Department of Oriental Antiquities, British Museum, has been subjected to chemical and metallographic analysis. These include Buddhist statuary as well as miscellaneous figures and ceremonial objects. A selection of these is shown in Figures 1 and 2. The dates of these objects were assigned

Figure 1: Cast iron figures (left: Buddha 1993.11-11.1; centre: standing horse 1994.1-29.4; right: standing ox 1994.1-29.5). The Buddha is 220mm in height.

Figure 2: Cast iron figures (left: crucible [not analysed], centre: Bodhisattva 1994.1-29.6; right: figure with pot belly 1988.5-19.3). The Bodhisattva is 280mm in height.

using stylistic criteria, for example roughly-cast animals on square plinths, similar to the horse (1993.7-14.1) and the ox (1993.8-4.1), have been found in tombs of the Yuan period (AD 1279–1368), so these two objects have been tentatively assigned to that period. Using this type of argument, 11 of the 19 objects are dated to the Ming and Qing dynasties *ie* from the 15th to the 19th centuries AD, the dates of the other eight being earlier, between the 8th and 13th centuries AD (Tang, Song, Jin, Yuan dynasties). The details of the objects studied are provided in Table 1. The study was carried out in conjunction with a parallel programme of analysis of 11th–13th century Chinese cast iron coins that has been reported separately (Wayman and Wang 2003).

Despite numerous studies of earlier Chinese ironwork, there have been few previously reported metallurgical examinations of Chinese cast irons of this later time period, and even fewer where microstructures were characterized. Among the latter are those of Pinel *et al* (1938), Henger (1970), Rostoker *et al* (1984), Moffatt *et al* (2001) and Chakrapani (2002). The results of other workers, mainly on earlier cast irons, have been compiled in Rostoker and Bronson (1990), Wagner (1993),

Table 1: Objects analysed

Object	Reg. no.	Date	Dynasty	height (mm)	casting type	comments
plaque	1992.6-13.1	AD 718	Tang	475	-	-
standing ox	1994.1-29.5	9-10thC	Tang/Liao	105	solid	-
head of Bodhisattva	1943.2-15.1	9-10th C	late Tang	560	hollow	-
warrior (standing on base)	1993.8-9.1	AD 1098	N. Song	330	hollow	-
head of Bodhisattva	1922.11-17.1	12-13th C	Jin	600	hollow	-
standing horse	1994.1-29.4	12-13th C	Jin	190	hollow	-
horse (on plinth)	1993.7-14.1	13-14th C	Yuan	145	hollow	-
ox (standing on frame)	1993.8-4.1	13-14th C	Yuan	225	hollow	-
seated Luohan	1990.4-13.1	AD 1494	Ming	1070	hollow	-
cat (seated)	1993.7-14.2	15-16th C	Ming	105	hollow	-
standing figure	1990.5-26.1	16th C	Ming	355	hollow	-
Buddha (seated, crowned)	1993.11-11.1	16th C	Ming	220	hollow	traces of gilding
altar vase	1982.2-13.1	AD 1579	Ming (Wanli)	520	-	-
jade maiden	1989.10-10.1	16-17th C	Ming	590	hollow	-
figure of Bodhisattva	1994.1-29.6	16-17th C	Ming	280	hollow	-
judge (seated)	1993.7-30.1	17th C	Ming/Qing	550	hollow	traces of gilding and red pigment
figure with pot belly	1988.5-19.3	17-18th C	Qing	200	near solid	-
incense burner	1990.11-22.1	18-19th C	Qing	320	-	-
incense burner	1988.5-19.2	AD 1848	Qing	280	-	-

Bronson (1999) and Wagner (2000), and research work continues (see *eg* Chen Jianli *et al* 2003). More such investigations are needed to increase our understanding of the singular developmental history of Chinese iron technologies, both smelting and casting. The study reported here is a contribution to this process.

Experimental procedures

Samples of the order of a few cubic millimetres in size were cut from each of the objects and examined using optical and scanning electron metallography in order to characterize their microstructures. Elemental compositions were obtained by energy dispersive X-ray analysis (EDX) in the scanning electron microscope (SEM). With this technique both bulk compositions and the compositions of individual microscopic constituents of the objects (microanalysis) could be determined.

The samples were first embedded in cylindrical epoxy metallographic mounts and then subjected to conventional metallographic specimen preparation procedures. These involved grinding to 600 grit silicon carbide followed by polishing using 6μm and 1μm diamond paste. The polished sections were then examined using both a Zeiss reflected light optical microscope and a JEOL SEM which was equipped with an Oxford Isis EDX analysis system with a Ge detector and an ultrathin detector window. Examination in this as-polished (unetched) condition provided information on the presence or absence of gas and shrinkage porosity, the non-metallic inclusion abundance and distribution and the state of corrosion damage. Also at this stage semiquantitative chemical analysis was carried out using SEM-EDX in order to determine the bulk contents of silicon, phosphorus, sulphur and manganese as well as the compositions of the non-metallic inclusions. Carbon contents were estimated from the microstructures as described below.

Standard reference materials (SS 551, SS 554, 183/3, all British Chemical Standards, issued by the Bureau of Analysed Samples, Ltd, and NBS 661, from the US National Bureau of Standards) were also analysed for silicon, phosphorus, sulphur and manganese by performing SEM-EDX analyses under conditions identical to those used for the samples. This confirmed that the lower limits of detection of these elements were approximately 0.1–0.15%. The SEM-EDX results can be considered to have a precision and accuracy of ± 10–20% relative to the values obtained.

The polished samples were then etched with 2% nital (2% nitric acid in alcohol) and re-examined using both optical and scanning electron microscopy to characterize the microstructures, including all of the microstructural constituent phases.

Since SEM-EDX is not suitable for carbon determination, the carbon contents of the white cast irons were estimated from photomicrographs of representative areas of the etched microstructures using procedures described in Wayman and Wang (2003). The carbon contents thus obtained are estimates, accurate to no better than ±0.1% carbon. Furthermore, this technique cannot be applied to grey or mottled irons which always contain some graphite, thus for objects with these microstructures it was not possible to obtain estimates of carbon content.

The metallurgy of the objects

The results of the compositional analyses are given in Table 2 and the microstructural results in Table 3. Of the 19 objects examined, 13 proved to be white cast irons, five were grey cast irons and one was a mottled cast iron.

Compositions

The elements of major importance in determining the microstructures of cast irons are carbon, silicon, phosphorus, sulphur and manganese. Detection limits for these elements (other than carbon) were in the order of 0.1%, so concentrations less than this amount would not have been detected by SEM-EDX; such cases are reported in the Tables as 'nd'.

The results of the compositional analyses are shown in Table 2, where the objects are listed in three categories: white, mottled and grey cast irons. It can be seen that the silicon contents of the objects ranged from below the detection limit to above 3%, however it is apparent that the grey and mottled irons had in general higher silicon contents (1.2–3.0%, average 2.0%) while the white irons were lower (highest: 1.12%, average: 0.5%). On the other hand, the phosphorus contents ranged widely between the detection limit and 1.16%, with little differentiation between grey and white irons. The silicon was found to occur preferentially in the ferrite and pearlite regions of the microstructure rather than in the cementite (iron carbide), while the phosphorus was found to be present in the form of steadite, as discussed below.

The sulphur contents also differed among the different categories of cast iron, with the grey and mottled irons having no more than 0.20%S while the white irons ranged as high as 1.36% with an average of 0.63%.

Table 2: Compositional results

Object	Reg no.	Bulk composition (wt%)					Inclusion composition (wt%)					Form
		C (est.)	Si	P	S	other	Fe	Mn	Ti	S	other	
White irons												
head of Bodhisattva	1922.11-17.1	5.2	nd	0.71	nd	-	19.4	45.6	nd	34.6	-	angular
altar vase	1982.2-13.1	2.7	0.43	0.42	0.88	-	63.5	1.2	0.7	34.3	-	globular
bell	1988.5-19.2	1.8	1.10	0.37	0.86	-	61.0	1.2	0.8	36.4	tr Cu	globular
jade maiden	1989.10-10.1	2.3	0.42	0.89	0.88	-	62.1	1.7	0.2	35.5	-	globular
seated Luohan	1990.4-13.1	3.1	0.53	0.41	0.49	-	63.2	1.4	1.2	33.8	-	globular
standing figure	1990.5-26.1	2.4	0.20	1.16	0.52	-	63.2	2.4	0.4	34.0	-	globular
incense burner	1990.11-22.1	2.0	1.12	0.21	1.36	-	63.2	0.5	0.5	34.9	0.5 Cu	globular
ox standing on frame	1993.8-4.1	2.2	0.51	0.77	0.76	-	60.9	2.1	0.6	36.5	tr V	globular
warrior	1993.8-9.1	2.8	0.51	0.44	1.00	-	61.3	1.3	0.9	36.4	-	globular
Buddha	1993.11-11.1	2.3	0.38	0.77	0.65	-	62.4	1.1	0.66	35.5	-	globular
standing horse	1994.1-29.4	4.5	0.81	0.71	0.23	0.33 Mn	22.6	42.8	nd	34.2	-	angular
standing ox	1994.1-29.5	3.9	na	0.46	0.20	-	na	na	na	na	-	few
figure of Bodhisattva	1994.1-29.6	2.6	0.31	0.83	0.39	-	61.9	1.4	0.6	35.8	-	globular
Mottled iron												
head of Bodhisattva	1943.2-15.1	-	1.83	0.17	0.2	0.48 Mn	9.2	56.7	0.5	33.2	-	angular
Grey irons												
pot-bellied figure	1988.5-19.3	-	1.24	1.02	nd	0.5 Mn	9.2	56.7	0.5	33.2	-	angular
judge	1993.7-30.1	-	3.01	0.26	0.11	0.29 Mn, 0.16 Ti	10-65	nd-53	nd-67	9 to 30	-	both
cat	1993.7-14.2	-	1.98	0.24	0.09	0.71 Mn, 0.12 Ti	20-59	nd-48	nd-78	nd-26	-	both
horse	1993.7-14.1	-	1.70	nd	0.14	0.28 Mn	24-40	33-44	0.4-0.8	25-32	-	both
plaque	1992.6-13.1	-	1.98	nd	nd	0.62 Mn, 0.35 Cu	3.2	61.3	0.22	34.6	-	angular

Note: nd = not detected; na = not analysed; tr = trace

Table 3: Microstructural results

Object	Reg no.	Microstructure	Steadite present	Graphite flake types
White irons				
head of Bodhisattva	1922.11-17.1	divorced eutectic with 72% cementite	yes	
altar vase	1982.2-13.1	divorced eutectic with 72% pearlite	yes	
bell	1988.5-19.2	divorced eutectic with 88% pearlite	yes	
jade maiden	1989.10-10.1	divorced eutectic with 80% pearlite	yes	
seated Luohan	1990.4-13.1	divorced eutectic with 65% pearlite	yes	
standing figure	1990.5-26.1	divorced eutectic with 78% pearlite	yes	
incense burner	1990.11-22.1	divorced eutectic with 85% pearlite	little	
ox standing on frame	1993.8-4.1	divorced eutectic with 82% pearlite	yes	
warrior	1993.8-9.1	divorced eutectic with 70% pearlite	yes	
Buddha	1993.11-11.1	divorced eutectic with 80% pearlite	yes	
standing horse	1994.1-29.4	divorced eutectic with 60% pearlite	yes	
standing ox	1994.1-29.5	ledeburite with 10% pearlite	yes	
figure of Bodhisattva	1994.1-29.6	divorced eutectic with 75% pearlite	yes	
Mottled iron				
head of Bodhisattva	1943.2-15.1	pearlite, cementite, graphite, ledeburite	yes	D, A
Grey irons				
pot-bellied figure	1988.5-19.3	ferrite (mainly), pearlite, graphite	yes	B, D
judge	1993.7-30.1	ferrite, graphite	yes	D, A
cat	1993.7-14.2	ferrite, graphite	yes	D, A
horse	1993.7-14.1	pearlite (mainly), ferrite, graphite	very little	A, B
plaque	1992.6-13.1	ferrite (mainly), pearlite, graphite	very little	D, A

Note: Graphite flake types (A, D *etc*) are according to the American Foundrymen's Society-ASTM designations (ASTM 2003).

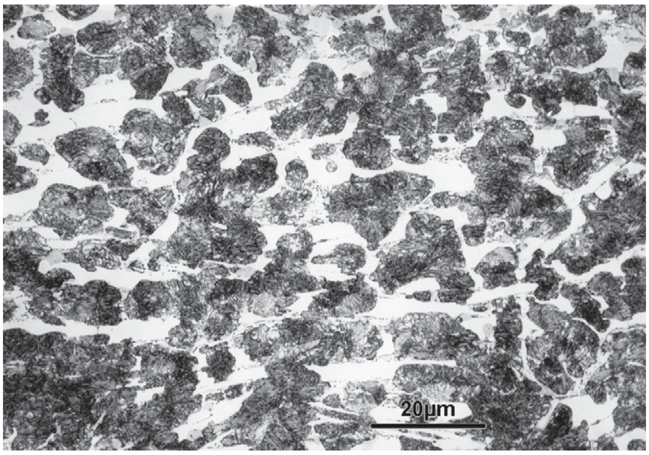

Figure 3: Divorced eutectic microstructure showing pearlite dendrites in a cementite matrix (Buddha 1993.11-11.1). Nital etch.

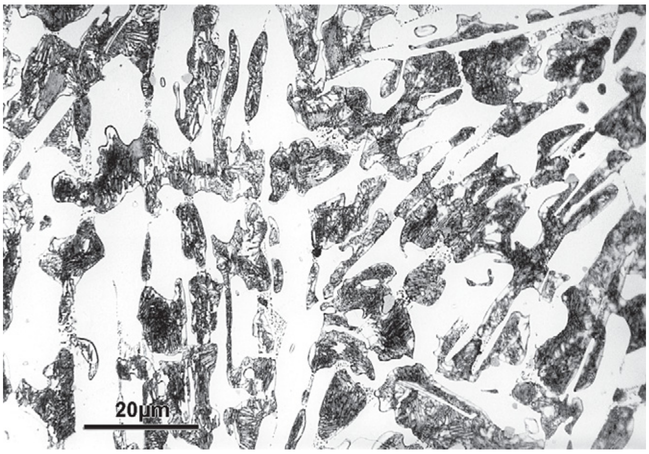

Figure 4: Divorced eutectic microstructure showing cementite plates in a pearlite matrix (standing horse 1994.1-29.4). Nital etch.

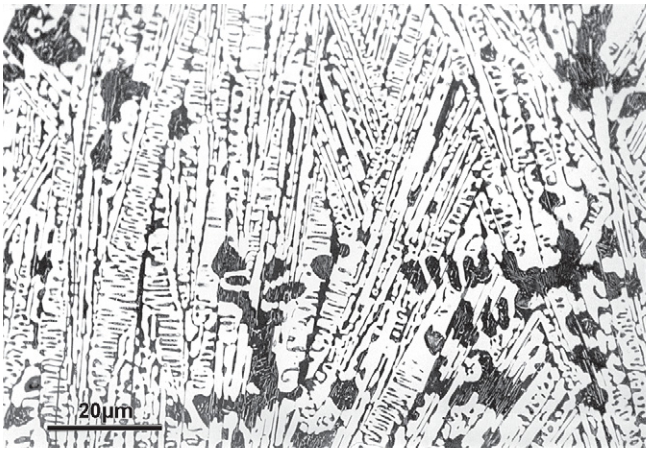

Figure 5: Ledeburitic microstructure showing pearlite dendrites in a ledeburite matrix (standing ox 1994.1-29.5). Nital etch.

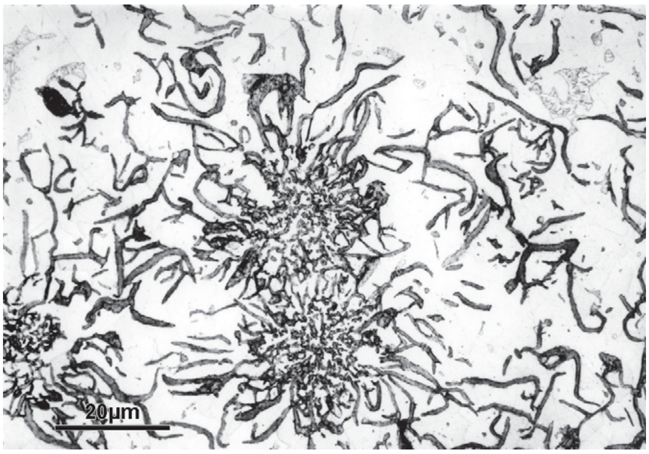

Figure 6: Ferritic grey iron microstructure showing graphite flakes in a ferrite matrix (judge 1993.7-30.1). Nital etch.

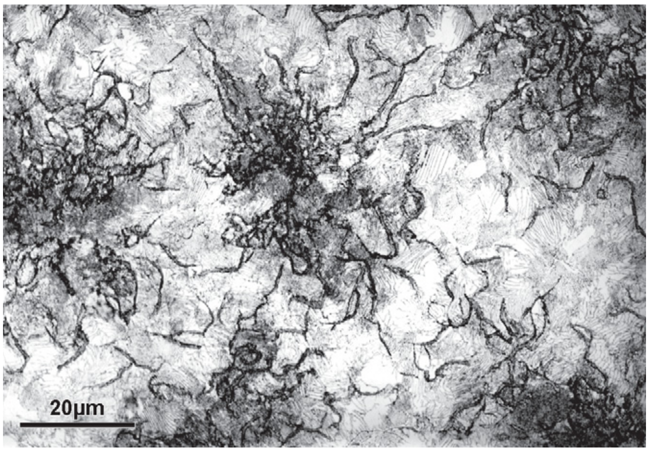

Figure 7: Pearlitic grey iron microstructure showing graphite flakes in a mainly pearlite matrix (horse 1993.7-14.1). Nital etch.

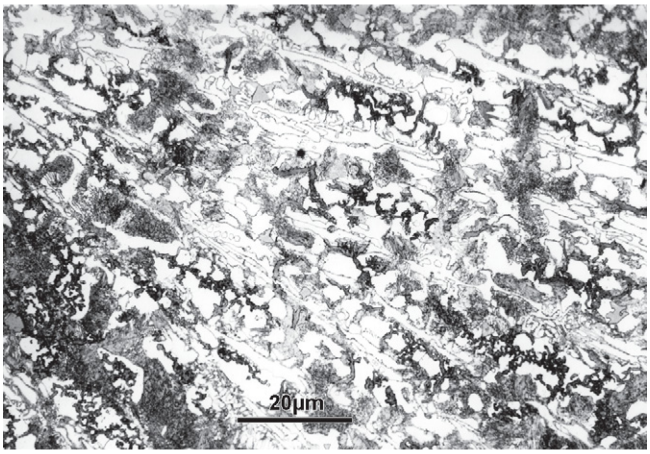

Figure 8: Mottled iron microstructure showing pearlite, cementite, ledeburite and graphite (Bodhisattva head 1943.2-15.1). Nital etch.

Microstructurally, sulphur occurred in these cast irons in the form of sulphide inclusion particles; in the matrix between the inclusions the sulphur was found to be below detection limits. Manganese was present in the matrix of the irons at levels higher than the detection limit in seven objects: all five of the grey irons, the mottled iron and one of the 13 white irons. The other 12 white iron objects all had bulk manganese contents too low to be detected. Manganese was also observed to be present in the sulphide inclusions in some of the castings.

Estimates of carbon contents could only be obtained for the white irons, as noted above, and were found to be widely spread over the full cast iron range from 1.8% to 5.2%C. The significance of these compositions and their relation to the microstructures are considered further in the Discussion section below.

Microstructures

The microstructures of the objects are tabulated in Table 3, again grouped as white, mottled and grey cast irons. It can be seen that among the white irons there were several sub-categories of microstructure. Twelve of the 13 white iron objects had divorced eutectic microstructures consisting of a proeutectic phase in a cementite matrix, without any of the eutectic constituent ledeburite (divorced eutectic microstructures will be considered in more detail in the Discussion section below). Of these 12 divorced eutectic microstructures, 10 were hypoeutectic with pearlite (originally proeutectic austenite) dendrites in a cementite matrix as shown in Figure 3, while the other two displayed hypereutectic microstructures with proeutectic cementite plates in a pearlite matrix as in Figure 4. The remaining white iron casting exhibited a very different microstructure, consisting of pearlite dendrites in a ledeburite matrix (Fig 5).

The five grey iron objects included two ferritic grey irons, which had graphite flakes in a ferrite matrix (Fig 6).The other three were pearlitic grey irons with mixed pearlite-ferrite matrices (Fig 7). The types of flake graphite present in these grey irons are also reported in Table 3 according to the AFS-ASTM designations (ASTM 2003).

The microstructure of the mottled iron consisted of pearlite, ledeburite and graphite (Fig 8).

Two additional constituents were also observed in the microstructures of the objects. One of these was steadite, the binary or ternary eutectic constituent (*ie* the ferrite-iron phosphide eutectic mixture or the ferrite-iron phosphide-iron carbide eutectic mixture respectively) (Fig 9). The steadite was observed to a greater or lesser extent in the microstructures of all of the objects, although in several of the grey irons very little was found. The abundance of steadite correlated well with the analysed overall phosphorus contents of the objects.

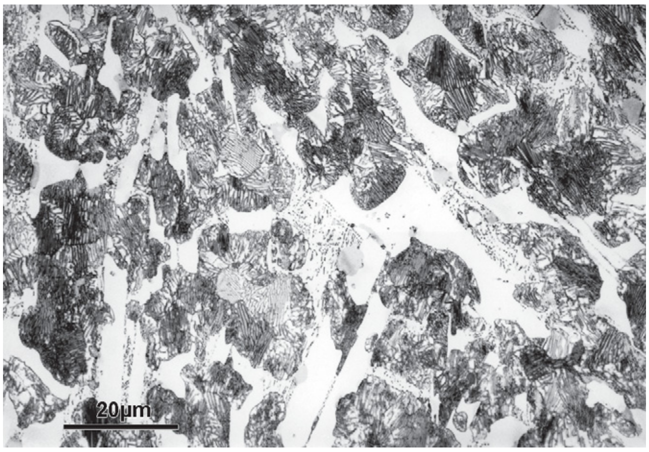

Fig. 9: Steadite and iron sulphide inclusions in divorced eutectic microstructure (Bodhisattva 1994.1-29.6). Nital etch.

The other important microstructural constituent found in these cast irons was sulphide inclusion particles (Fig 9), which were found in all of the objects, but were observed to be of two types (Table 2). Some objects contained iron sulphide inclusions (FeS) while others had manganese sulphide or mixed manganese-iron sulphide inclusions *ie* MnS, or (MnFe)S with manganese predominating. Measured manganese contents of sulphide inclusions ranged to above 60%Mn, however it must be noted that some of these inclusions were smaller than the minimum analysis size of the SEM-EDX system so that the apparent composition determined by the analysis actually included a contribution of extra Fe from the surrounding matrix, *ie* the true manganese content is higher than that measured. Those sulphide particles in which manganese was predominant were observed to be dove grey in colour with angular shapes, whereas the iron sulphides were a yellowish colour with globular rounded shapes. The manganese-rich sulphides were found in all seven of the objects that had detectable bulk manganese contents, as well as in one of the other white iron objects. In contrast, the remaining 11 white iron objects contained iron sulphide inclusions (containing less than 2%Mn and less than 1%Ti) (Table 2).

The sulphide particles and the steadite were typically found in the same parts of the microstructure, specifically in regions that are recognizable as being the last to solidify. In several of the grey irons, small titanium-rich intermetallic particles were also observed. Micro-porosity, in the form of both shrinkage and gas porosity, was frequently present.

Discussion

The classification of cast irons as grey, white or mottled derives from the form in which the carbon exists in the iron, as manifest in the appearance of the fracture surface. White cast irons have virtually all of their carbon in the form of cementite, both as free cementite and as the cementite constituent of pearlite. In contrast, in grey cast iron some or all of the carbon occurs as graphite, typically with a flake-like morphology. Here the background matrix in which the graphite is embedded can be either ferrite or pearlite or a pearlite-ferrite mixture, leading to the sub-categorization of grey cast irons as ferritic or pearlitic (those with a mixed ferrite-pearlite matrix are assigned to the pearlitic category). Mottled cast irons are intermediate between grey and white, having both graphite and cementite as major microstructural constituents.

Of the cast irons examined in the study reported here, 13 were found to be white cast irons, one was a mottled cast iron and five were grey cast irons. Whether a cast iron solidifies as grey or white or mottled iron is determined by both the chemical composition and the rate of cooling during the casting process, with faster cooling rates being more likely to give a white cast iron when the compositional factors are similar.

The main compositional factors which determine whether a cast iron is grey or white are its carbon, silicon and sulphur contents with high carbon, high silicon and low sulphur stimulating the formation of graphite at the expense of iron carbide (cementite) and hence promoting grey cast iron. Sulphur contents above 0.5% are likely to be sufficient to ensure a white iron regardless of the silicon content and cooling rate (Rostoker and Bronson 1990). Manganese by itself also tends to stimulate the carbide but its role is more complex because, when sulphur is present, manganese reacts with it to form manganese sulphide MnS (or mixed Mn-Fe sulphide) particles, thereby removing sulphur from its carbide-stimulating role. The role of phosphorus is also complex. It has a mild direct effect of stimulating graphite formation but in addition its presence increases the solidification temperature range of the cast iron, thus during solidification the cast iron spends more time in the mixed solid-liquid state, giving more time for graphite to form. In the study reported here, it was possible to determine the silicon, phosphorus and sulphur contents in almost all cases, however the carbon content could be determined only in those microstructures which did not contain graphite, *ie* only in the white irons, and even then only as estimates.

The compositions of the irons analysed here (Table 2) were in general able to account successfully for whether the castings solidified as white irons or grey irons, without taking cooling rate into consideration. As shown in Table 2, all of the low silicon irons (most of which also had high sulphur contents) solidified as white irons, whilst the castings with high silicon contents (all of which also had low levels of sulphur) solidified as grey irons, as would be expected. It is noteworthy that the grey irons all contained measurable amounts of manganese, so that even the small amounts of sulphur they did contain would have been rendered ineffective at stimulating cementite formation.

Details of the microstructures of each category of cast iron as observed in the objects studied are now considered and attempts made to interpret them in terms of the technologies involved.

White Cast Irons

White cast irons fall into two different microstructural categories, namely the divorced eutectic type and the ledeburitic type. The metallurgy of these materials including the effect of compositional variables is discussed in detail in a recent related publication (Wayman and Wang 2003).

Divorced eutectic microstructures are expected to form in cast irons whose carbon contents are relatively far from the eutectic composition, especially when the phosphorus content is high and/or the cooling rate is relatively low. In the present work, 10 of the 12 divorced eutectic objects were found to have carbon contents between 1.8 and 3.1%C, *ie* well below the eutectic composition, which varied between 3.9 and 4.1%C (a rule of thumb is that each percent of total silicon plus phosphorus lowers the eutectic carbon content by 0.3%C). As well, they contained relatively high phosphorus levels (up to 1.16%). As a consequence, these 10 objects have divorced eutectic microstructures consisting of pearlite dendrites (transformed from proeutectic austenite dendrites during subsequent cooling) in a matrix of eutectic cementite as illustrated in Figure 3.

The remaining two of the 12 divorced eutectic alloys had much higher carbon contents (4.5 and 5.2%), and as a result they exhibited microstructures consisting of cementite laths in a matrix of pearlite (transformed from eutectic austenite during subsequent cooling), as illustrated in Figure 4. The eutectic carbon contents of these two castings are 3.9 and 4.1%C respectively, after accounting for the effects of their silicon and phosphorus contents, so both castings are sufficiently far above the eutectic composition (and have high enough phosphorus contents, both around 0.7%) to explain why they formed divorced eutectic microstructures.

Only one of the 13 white cast iron objects had a ledeburitic type microstructure. This object had a carbon content of 3.9% while its eutectic composition was estimated to be 4.1%. When the composition is this close to the eutectic composition, the ledeburitic microstructure is to be expected, and the phosphorus content, at 0.46%P did not compensate for this. The result was the typical hypoeutectic ledeburite microstructure shown in Figure 5 where dendrites of pearlite (transformed from proeutectic austenite during cooling) are seen embedded in a eutectic pearlite-cementite matrix.

Networks of casting flash were observed on the surfaces of all of the white iron objects analysed. Similar networks were also observed on several other cast iron objects in the collection that could not be analysed because of sampling restrictions. This flash arises because of the piece-mould technique used for casting the objects, when liquid metal penetrates between the mould segments during pouring. This casting flash is often removed from castings by abrasion (filing or grinding) after casting but its removal from a white cast iron is extremely difficult because of its high hardness. The existence of these networks shows unambiguously that in fact the piece-mould technique was used, rather than, for example, the lost-wax method of casting. This will be discussed further below.

Grey Cast Irons

Five of the castings were found to be grey cast irons, meaning that some or all of their carbon was present in the form of graphite flakes. As in the case of white irons, cooling rates play a role in the microstructural development. Thus, two of the objects were observed to be ferritic grey irons (Fig 6), a type of microstructure indicative of slow cooling of the casting in the vicinity of the eutectoid temperature, around 725°C, so that when the austenite decomposes there is enough time for its carbon to migrate into the pre-existing graphite rather than forming pearlite. The other two exhibited mixed ferrite-pearlite matrix microstructures with ferrite predominating over pearlite, indicative of moderately slow cooling after solidification.

The grey irons had relatively high silicon contents, as well as high manganese levels, in comparison with the white irons. These factors are undoubtedly sufficient to explain their being grey, even without the relatively slow cooling which is also favourable to grey iron. The manganese has combined with the sulphur to form the manganese sulphide inclusion particles, preventing the sulphur from inhibiting graphite formation. The levels of silicon and manganese are determined by conditions inside the iron-smelting furnace, and consideration will be given to this in a following section.

Of the five grey irons, one exhibited the vestiges of a casting flash network that appeared to have been almost completely removed by abrasion, while on one of the others the flash network was clearly visible. The other three grey irons showed no evidence of casting flash networks, thus if these, like the others, had been cast by the piece mould method, the flash had been removed after casting. Removal of this flash from grey cast iron objects would not have been difficult, in contrast to the situation for white cast irons. In fact, it has sometimes been assumed that observation of a network of flash on a cast iron object is an indication that it is a white cast iron, however exceptions to this, including the monumental Cangzhou lion (Wagner 2000), have been noted by other workers and it is becoming clear that identifying the type of cast iron on the basis of the existence of flash can be misleading.

Mottled Cast Iron

One of the objects, a Bodhisattva head, had a microstructure that contained both graphite and ledeburite, as well as pearlite, and hence this is categorized as a mottled iron. This casting contained 1.8%Si and low sulphur, a combination that suggests that it might have been expected to solidify as a grey iron. The fact that it was mottled could be the result of a relatively low carbon content, (it was not possible to estimate the carbon content metallographically because graphite was present) or a relatively fast cooling rate as it solidified. Such a situation is exemplified by the lotus component of the Cangzhou lion (Wagner 2000), which is a white iron with some mottling while the main body of the 50-ton casting is grey iron. Consistent with this, more ledeburite was observed around the outside of the cross-section of the Bodhisattva head, with more pearlite and graphite in the centre, indicative of differential cooling rates during solidification.

A network of casting flash was visible on the surface of this object, but was not as well developed as on the white iron objects. This could be expected in mottled iron, whose abrasion resistance would be intermediate between grey iron and white iron, and possibly variable with location within the casting.

Casting Conditions

Characteristics that can be useful in interpreting casting conditions, such as the cooling history of the casting, include its surface topography and the details of its microstructure. In the present case the networks of casting flash on the surfaces of most of the castings showed that the piece-mould technique had been used, rather than lost-wax casting. In fact, piece-moulding is the expected casting method for these objects. The marvellous bronze castings of the Chinese Bronze Age, beginning before the mid-second millennium BC, were made by the piece-mould technique, and although alternative techniques, notably the lost-wax method, supplanted piece-moulding for bronze objects, this does not appear to have been the case for cast iron (Kerr 1990, 68, Cowell *et al* 2003).

The cooling history of a cast object is determined by its size and shape, by whether it is solid cast or hollow cast, by the materials from which it is made, by the shape and thickness of mould and core (if any), by the temperature of the liquid metal and of the mould just prior to casting, by the environment in which the mould cools, *etc*. Furthermore, different parts of a casting can cool at different rates, while the selection of the metallographic sampling site is often restricted. The result of this complexity is that even if the cooling history of a particular sample could be precisely determined by metallurgical analysis, it would still not be easy to resolve the details of the casting conditions of a particular object.

In the case of cast iron, the microstructural information that must be interpreted includes: whether the casting is white or grey or mottled; if white, whether it is a divorced eutectic or ledeburitic; if grey, the nature of the matrix (fully ferritic, fully pearlitic or intermediate) and the size and distribution of the graphite flakes. Ideally these parameters should be assessed both at the surface of the object and at the centre of the section, noting any gradients or local heterogeneities, but it is often not feasible or permissible to cut the necessary sections from artefacts. Of course all of these microstructural parameters depend not only on cooling rate but on other factors as well; these include, notably, chemical composition, which therefore must be fully considered (including its heterogeneity) when interpreting the microstructures.

Obviously obtaining information about casting conditions by metallurgical analysis is not a straightforward matter. However, in most of the cast iron objects considered here, the chemical compositions of the objects were sufficient to explain the gross aspects of their microstructures without taking cooling rate into account. This could imply that the cooling rates of all the cast objects had been similar, which could suggest, for example, that all were cast under similar conditions into moulds of the same type, presumably made of clay. The size of the casting would be expected to have an influence; however, although the castings analysed displayed a wide range of sizes, none was particularly thin in section. Almost all of the objects were hollow castings, with section thickness of the order of 50 to 100mm.

Post-casting processing

Examination of the microstructures revealed that all of the objects were in the as-cast condition, with no indications that, following the casting operations, they had been subjected to any thermal processing (*eg* heat treating). At least as early as the third century BC, Chinese iron-workers learned to heat-treat white cast iron in order to decompose the cementite and then precipitate the carbon as agglomerations of graphite (see, *eg*, Wagner 1989), making what would become known as 'malleable cast iron'. Alternatively the heat treatment could remove all or some of the carbon from the cast iron without the precipitation of graphite, transforming it into wrought iron or, more usefully, steel. In the case of these castings, there is no obvious reason why heat treatment would have been beneficial; therefore it was not surprising that no evidence for it was found.

Similarly, no evidence was found for the mechanical working of any of these objects. For the white cast iron objects this is to be expected because the hardness and brittleness of white cast iron would make such working impossible under normal conditions. Removal of casting flash from the surfaces of the grey iron objects would have been performed by mechanical means but only in localized areas. It can only be said that at the locations where the samples were taken, no mechanical working had occurred.

Some early Chinese iron castings are known to have been inlaid with precious metals, and traces of paint on other such objects have also been found. For example, four Ming dynasty (late 15th century AD) iron statues studied by Moffatt *et al* (2001) were found to have been decorated with multiple lacquer and polychrome pigmented layers as well as having been gilded in some areas using gold leaf. There were indications that two of the objects studied in the present work, the seated judge and the crowned Buddha, had originally been at least partially gilded, and there was also evidence of red pigmentation on the Buddha. Of course, it is possible that others had originally been given surface decoration, evidence for which has been lost since the original production of the object.

Smelting conditions

In principle, information about the smelting conditions should be reflected in the microstructure and composition of the iron castings. In this light it is of interest firstly to address the question of whether this iron was smelted using mineral fuel (coal or coke) as opposed to charcoal to provide the high temperatures and reducing atmospheres needed for blast furnace operation. It is generally accepted that coal or coke was being used for iron smelting in Song times, and that there are indications of much earlier use. The most widely quoted analytical evidence for this is the presence of sulphur in the smelted iron. Consideration of this issue by Wayman and Wang (2003) led to the conclusion that although a high sulphur content cannot be taken as absolute proof that the iron was smelted with mineral fuel, data compiled from the work of many previous researchers failed to reveal any charcoal-smelted iron with sulphur contents much above 0.1% and only one above 0.2%. Thus the empirical evidence is that high sulphur content can be used to indicate coke smelting, albeit with some caution. On the other hand a low sulphur content is not strong evidence for the use of charcoal.

Higher furnace temperatures are suggested by the high silicon and manganese contents of the grey and mottled cast iron objects. These all had silicon contents above 1.2% (accounting for the presence of graphite) and elevated manganese contents compared with almost all of the white irons. In contrast, only a handful of the early (prior to the fourth century AD) Chinese irons in the compilations of Tylecote (1976) and Rostoker and Bronson (1990) had silicon contents above 0.3%, suggesting low furnace temperatures, as might be expected in these earlier times. Silicon contents in the compiled European cast iron analyses range from less than 0.3% to above 3% silicon, suggestive of a wide range of furnace temperatures.

Here again, the choice of smelting furnace fuel could play a role. Since coal- or coke-fired furnaces can be expected to operate at higher temperatures than charcoal-fired furnaces (see, *eg*, Rehder 1987), the argument could be made that high-silicon cast irons are more likely to have been coal- or coke-smelted. Data compiled by Rostoker and Bronson (1990, Table 10.3) show silicon levels for most of the reported coke-smelted irons to be above 1.2%, while most of the cold-blast charcoal smelted cast irons had lower silicon contents. However many variables other than the fuel are involved in determining furnace temperature, not the least of which are furnace size and air blowing rate, and Rostoker and Bronson (1990, 109) point out that "Chinese ironmakers succeeded in producing low-silicon pig iron with coke and charcoal pig containing almost no silicon."

A complicating factor is that higher blast furnace temperature can also lower the sulphur content of the iron by driving sulphur into the slag phase, especially if limestone is added as a flux. Thus the sulphur content remaining in the iron could be lowered enough that it would be a less clear indicator for coal-smelted vs. coke-smelted iron. Hence both silicon/manganese and sulphur must be looked at together in evaluating the possible furnace fuels. Furthermore these variables have a strong effect on the type of cast iron formed, since a furnace temperature high enough to give high silicon iron will thereby stimulate the formation of grey iron, provided that the sulphur content does not remain too high.

General

Although there have been numerous analyses of early Chinese cast irons from before the fourth century AD (see, *eg* Wagner 1993), very few analyses of more recent Chinese cast irons, with dates similar to those analysed here, have been reported. Pinel *et al* (1938) report four 6th century AD grey or mottled iron castings with compositions very comparable with those of the grey irons analysed here (1.98–2.42%Si, 0.17–0.31%P, 0.06–0.13%S, 0.13–0.78%Mn, 3.22–3.35%C). Rostoker *et al* (1984) analysed three 16th century white cast iron bells and found 0.29–0.57%S and 3.2–4.7%C, once again very comparable with the white irons analysed herein. Wayman and Wang (2003) analysed 37 Song dynasty coins and found all of them to be white cast irons, about equal numbers having divorced eutectic and ledeburitic microstructures. The divorced eutectic coins had compositions which agreed in general with the hypoeutectic divorced eutectic objects reported here (0.26–0.78%Si, 0.6–1.65%P, 0.39–2.05%S, 2.6–4%C) while the ledeburitic coins had compositions similar to the single ledeburitic white iron casting.

Consideration of the dates of the objects (Table 4) reveals that the earlier objects, including the one pre-Song object, tend to have lower sulphur contents. This

Table 4: Results listed chronologically

Object	Date AD	Bulk composition (wt%)					Type	Microstructure
		C (est.)	Si	P	S	other		
plaque	718	-	1.98	nd	nd	0.62 Mn, 0.35 Cu	grey	ferrite, some pearlite, graphite
standing ox	9th-10th C	3.9	na	0.46	0.20	-	white	ledeburite with 10% pearlite
head of Bodhisattva	9th-10th C	-	1.83	0.17	0.20	0.48 Mn	mottled	pearlite, cementite, graphite, ledeburite
warrior	1098	2.8	0.51	0.44	1.00	-	white	divorced eutectic with 70% pearlite
head of Bodhisattva	12th-13th C	5.2	nd	0.71	nd	-	white	divorced eutectic with 72% pearlite
standing horse	12th-13th C	4.5	0.81	0.71	0.23	0.33 Mn	white	divorced eutectic with 60% cementite
horse	13th-14th C	-	1.70	nd	0.14	0.28 Mn	grey	pearlite, some ferrite, graphite
ox standing on frame	13th-14th C	2.2	0.51	0.77	0.76	-	white	divorced eutectic with 82% pearlite
seated Luohan	1494	3.1	0.53	0.41	0.49	-	white	divorced eutectic with 65% pearlite
cat	15th-16th C	-	1.98	0.24	0.09	0.71 Mn, 0.12 Ti	grey	ferrite, graphite
standing figure	16th C	2.4	0.20	1.16	0.52	-	white	divorced eutectic with 78% pearlite
Buddha	16th C	2.3	0.38	0.77	0.65	-	white	divorced eutectic with 80% pearlite
altar vase	1579	2.7	0.43	0.42	0.88	-	white	divorced eutectic with 72% pearlite
jade maiden	16th-17th C	2.3	0.42	0.89	0.88	-	white	divorced eutectic with 80% pearlite
figure of Bodhisattva	16th-17th C	2.6	0.31	0.83	0.39	-	white	divorced eutectic with 75% pearlite
judge	17th C	-	3.01	0.26	0.11	0.29 Mn, 0.16 Ti	grey	ferrite, graphite
pot-bellied figure	17th-18th C	-	1.24	1.02	nd	0.5 Mn	grey	ferrite, some pearlite, graphite
incense burner	18th-19th C	2.0	1.12	0.21	1.36	-	white	divorced eutectic with 85% pearlite
bell	1848	1.8	1.10	0.37	0.86	-	white	divorced eutectic with 88% pearlite

Note: nd = not detected; na = not analysed.

by no means excludes the possibility of their being coke-smelted, as discussed above. In fact, charcoal-smelted iron continues to be made in China to the present day (P T Craddock, pers comm). On the other hand, ten of the 12 white iron castings had sulphur contents above 0.39%, and these are expected to be coke-smelted irons. It is perhaps noteworthy that these are the ten objects with hypoeutectic divorced eutectic microstructures, *ie* carbon contents well below the eutectic composition. It is known that high phosphorus and silicon restrict the ability of liquid iron to absorb carbon, and the effect of sulphur in this respect is likely to be comparable. A similar effect was noted in the recently-analysed Song dynasty cast iron coins (Wayman and Wang 2003).

It is obviously possible that the iron used to make some of these objects was coke-smelted while others were made of charcoal-smelted iron, as is believed to be the case with the Song coins. As iron smelting using coal or coke became more common in China, it is to be expected that both charcoal- and coal/coke-fired furnaces might have been in use in the same region at the same time. Furthermore it is not impossible that some smelting furnaces were burning mixtures of charcoal with coal or coke, nor is it unlikely that the castings were poured from a remelting furnace where pig iron and/or scrap, possibly from different sources, were mixed together. Consequently, the wide range of silicon and sulphur contents observed is not surprising.

A final question that must be addressed is whether white iron or grey iron was deliberately chosen for a particular object with its service conditions in mind. For example all of the Chinese cast iron coins analysed in the complementary study (Wayman and Wang 2003) were made of white cast iron, whose wear resistance is far superior to that of grey cast iron, a desirable property for coinage. Hence smelting conditions that led to white cast iron may well have been intentionally selected for coinage. For the particular statuary and other ritual objects analysed here, both white iron and grey iron would have had acceptable mechanical properties, since the demands in terms of mechanical loading would not have been great. There are, however, two ways in which grey iron is superior: it is better able to replicate the fine-scale surface features of the inside of the mould, and the removal of the casting flash would be possible.

Only a handful of Chinese cast irons dating to before the fourth century AD are identifiable as grey irons (see, *eg*, Wagner 1993, translation 7.5 and Figs 7.24 to 7.27 for examples). Since these all have low silicon contents, presumably from low furnace temperatures, the only way they could have been made as grey irons was by enforcing extremely slow cooling conditions, much as was done for 19th century wok pans (Wagner 1993, 348). This in itself is an indication of intent, since deliberate efforts would have been necessary to achieve this very slow cooling. Furthermore Wagner notes (1993, 346) that the few very early grey iron castings as well as some of the early white and mottled irons did in fact have the microstructures appropriate for their service functions, raising the possibility of intentional control over smelting and/or casting conditions even before the fourth century AD. Rostoker and Bronson (1990) point out that the sixth century AD Chinese grey cast irons analysed by Pinel *et al* (1938) were all objects which whose service performance would have benefited from their being grey irons.

It therefore appears plausible that deliberate efforts were being made to control the microstructure and hence the properties of some Chinese cast irons. Analysis of more objects will be required to provide further evidence in this regard.

Conclusions

A group of 19 Chinese cast iron objects, mainly statuary and ritual objects dating from between the eighth and 19th centuries AD, has been subjected to microstructural and compositional analysis. The objects analysed include a wide range of types of cast iron, including white cast iron with both divorced eutectic and ledeburitic microstructures, both ferritic and pearlitic grey cast iron, and mottled cast iron. The type of cast iron formed was found to be explicable by the chemical composition of the iron, especially its silicon, phosphorus and sulphur contents. Thus the cast objects are unlikely to have had widely different thermal histories, such as might have been the result of different modes of casting or casting mould materials. The different chemical compositions can be partially explained by different smelting conditions, such as the use of coke-smelted iron and charcoal-smelted iron, and the use of iron ore from different ore bodies. The sulphur contents, while not providing definitive evidence, suggest that at least some of the cast irons were smelted using coke rather than, or in addition to, charcoal.

All of the castings studied were found to be in the as-cast condition with no signs of heat treatment. Casting flash may have been removed from some of the grey iron castings, presumably by filing or grinding, but there were no other signs of mechanical working. Where it was possible to tell, specifically for the white and mottled irons, the objects had been cast using the piece mould process.

This work shows how, by the judicious consideration of a wide range of evidence, it is possible to begin to chart the progress of iron smelting throughout the past two millennia.

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