PLASMA TECHNOLOGY IN FERROALLOY PROCESSING

Rakesh Kumar and D. Sanyal National Metallurgical Laboratory, Jamshedpur - 831 007

Introduction

Plasma technology has come a long way since the early reported applications to metallurgy by Sir W. Siemens (1878) and H.Moissan (1897) about a century back [1]. Major attraction of using plasma in metallurgical smelting operations, especially in the last two to three decades, have emerged from its ability to deliver high grade heat to any environment independently of oxygen potential. Furthermore, compact unit of high throughput, due to high power flux and higher reaction rates attained, and ability to handle cheaper fine feed without causing pollution problems have been the other important attractions. Although, the development of plasma technology in the area of nonferrous extractive metallurgy is still confined to the laboratory and pilot plant research stage, applications in ferrous metallurgy have entered the stage of commercialization. Table 1 lists some of the high capacity iron and steel installations. Ferroalloys production, using carbothermic smelting reduction of oxide ores and by remelting of ferroalloys fines, has been a major driving force in fostering the development of plasma technology. As indicated in Table 1, both ferro manganese and ferro chromium are being produced at present on industrial scale. Research and development in the production of other ferroalloys, e.g. ferro silicon, ferro vanadium, ferro molybdenum, is in an advanced stage.

A brief review is presented in this paper on the application of plasma technology for ferroalloys production. We begin with the definition of plasma in general, followed by a discussion on various plasma furnaces, process fundamentals, i.e. chemistry and transport phenomena in plasma environment, and energy considerations involved in plasma smelting. Finally, the advantages of the technology, vis-a-vis the conventional processing using submerged arc furnace, and relevance of the technology in the Indian context are brought into focus. Specific examples of ferroalloys are cited wherever possible. The reader may refer to the reviews [1-6] for detail.

Plasma - A Basic Definition

Reckoned as the fourth state of matter, a plasma is essentially an ionized gas which may comprise molecules, atoms, ions (in ground or excited state), electrons and protons, and is sufficiently ionized to become electrically conductive. Overall, a plasma maintains electrical neutrality, i.e. the property of quasineutrality. Unlike an ordinary gas, a plasma contains free electric charge which may be produced by a variety of ionization processes, e.g., high energy radiation, such as, UV, X-ray etc., collision processes in intensely heated gas electrodeless RF discharge, microwave, shockwave, laser beam, or simply electrical arc discharge. It is the last process which is of interest in generating plasma for ferroalloys production.

A plasma can be of two types - the equilibrium or thermal or hot plasma and the nonequilibrium or cold plasma. In the former, the temperature of the electrons is close to that of heavy particles and chemical equilibrium prevails. In the later, the electron temperature is far above the sensible

temperature of heavy particles, which is closer to room temperature. Plasma produced by high intensity electric arc, as in the ferroalloys production, falls in the category of thermal plasma. Although, the temperature in the case of thermal plasma ranges from 2000 to 20000 K, most processes of practical interest operate between 4000 to 7000 K.

Plasma Furnaces for Ferroalloys Smelting

Like any conventional electric arc furnace, a large scale commercial thermal plasma device operates on the basis of a high intensity electric arc discharge originating from a cathode and terminating at an anode. Several reviews on plasma furnaces are available in literature [1,2,5-9]. A general description of plasma furnaces used for the smelting of ferroalloys is presented here.

Depending upon the way the arc is struck, the state of the feed material, as well as, its handling strategy, plasma systems are broadly classified into the following three categories : (a) nontransferred arc plasma; (b) transferred arc plasma; and (c) in-flight plasma. Nontransferred arc plasma system, normally designed to heat gases, consists of two electrodes in which one of the electrodes may be tubular type from which the hot gas emerges (Fig.1a). In the transferred arc plasma system, one of the electrodes is in the plasma torch and the material to be heated or melted acts as the second electrode (Fig. 1b). The in-flight plasma system involves the feeding of raw materials (completely or partially) within the plasma to utilize the thermal and reduction potential of plasma flame (Fig.1c). As listed in Table 2, a number of reactor designs have emerged for the smelting of ferroalloys depending upon the configuration of plasma systems in the furnace. Some typical furnaces are schematically shown in Figure 2. MINTEK open bath furnace (Fig. 2a) and University of Minnesota in-flight plasma reactor (Fig. 2b) make use of transferred arc and in-flight plasma system, respectively. The design of Hydro-Qubec-Noranda reactor, as adopted in Davy McKee Hi-plas furnace (Fig.2c), the reactor developed by ASEA, Tetronics plasma furnace and Bethlehem Steel falling film reactor, make use of both transferred arc and in-flight systems. The extended arc flash reactor (EAFR) (university of Toronto) includes' in-flight gas reduction and as well as melt heating by plasma fire ball generated using non-transferred arc system (Fig 2d). SKF design make use of a shaft furnace in which nontransferred plasma system is used essentially for heating the gases. In recent years transferred arc system, using in-flight reduction and bath smelting, have received considerable attention and will be the point of focus in the following sections.

Process Chemistry Consideration

The application of thermodynamic and kinetic principles in the extraction of metals using plasma has been found to be of prime importance. In this respect, chemical driving force and energy balances determined using ΔG and ΔH considerations, the use of acidic or basic slag recipes and study of reaction mechanisms and process stages have all been found to be vital in the refinement and advancement of plasma reactor engineering and design [9,10].

Thermodynamics

Coal or carbon based reductants are potentially the most economical to use with a plasma reactor. Thermodynamic principles described in an earlier chapter apply for the carbothermic reduction of metal oxides in plasma reactor too. Some salient points, relevant to the processing of ferroalloys in plasma environment, are presented here. Thermodynamically, metal oxide reduction with a carbon based reductant can be represented by the following reaction :

$$MO + C = M + CO$$

However for more stable oxides than FeO, as the temperature is increased, metal carbide formation by

...(1)

...(3)

MO + 2C = MC + CO

is thermodynamically more feasible than elemental metal (Fig.3a-b). The metal carbide can also participate in the reduction of metal oxide according to the reaction given below :

$$MO + MC = 2M + CO$$

The reaction (3) generally has the highest free energy of formation and requires a higher temperature than for direct metal formation (1). Table 3 lists the minimum temperature (temperature corresponding to $\Delta G = 0$) for reaction (1), (2) and (3). For carbothermic reduction in a plasma reactor, a further criterion is that the reactor temperature is not only greater than the minimum (i.e. $\Delta G = 0$) temperature but the melting point T_m of metal (or the temperature at which the metal dissolves in the Fe-C melt) and the slag formation temperature. The data presented in Table 4 indicate that, in general, the minimum reactor temperature is dictated by the minimum ($\Delta G = 0$) temperature.

Since carbide formation is favoured at higher temperature for metals which form more stable oxide than Fe₂O₃ and metal formation by reaction (3) requires a very high temperature, plasma smelting is more suitable for the production of bulk carbide or high carbon ferroalloys. Plasma environment, especially in the in-flight mode, may be ideal for the production of low sulfur and low phosphorus ferroalloys. This may be due to the vaporization of sulfur or phosphorus compounds and early slag formation under high temperature. Stable gaseous compound, e.g. SiS (ΔG° at 2000K = -28 kcal/mol) at the slag surface in the absence of any basic flux and likelihood of the formation and volatization of CaS ($\Delta G^{\circ} = -76$ cal/mol at 2000K) and Na₂S ($\Delta G^{\circ} = -76$ cal/mol at 2000K) may facilitate S removal in plasma environment [9,10].

Kinetics and Mechanisms

Plasma environment provides some unique conditions which are favourable for the reduction of mineral oxides with carbon based reductants : the plasma is in nonequilibrium conditions, i.e. electron temperatures are much higher than the heavy species, e.g. ions; gaseous molecules and atoms are present in very reactive states; and a significant degree of ionization of species exists. Such conditions make any proposed mechanism(s) of metal oxide reduction in plasma environment highly speculative. The mechanism(s) for in-flight plasma reduction, which involves intimate plasma particle interaction, are expected to be more complex than bath-smelting using plasma.

In-flight Plasma Reduction: Particles are subjected to steep temperature (1000-10000K) and velocity (100-400 m/s) gradient during flight in plasma flame (Fig 4) [11,12]. Since the residence time of particles in the plasma is small (few milliseconds), complete reduction of particles is not

achieved during in-flight. Due to high temperatures, the particles undergo melting and vaporization to different degrees depending upon size and their physical properties. Chemical reactions, e.g. gasification, reduction and slag formation, involving different combinations of solid, liquid and gaseous phases can also take place simultaneously [13,14].

Carbothermic reduction of metal oxides in plasma environment, like that in conventional process, occurs via the kinetically more favourable gaseous reductant CO, i.e.

...(4)

...(5)

$$MO + CO = M + CO_2$$

and the Boudouard (C gasification) reaction is the rate controlling reaction, i.e.,

$$CO_{2} + C = 2CO$$

Addition of reaction (4) and (5) results in overall thermodynamic reaction (1). Under certain circumstances a small amount of oxygen may need to be injected in order to initiate and sustain gasification reaction and carbothermic reduction process under plasma.

There have been some attempts to analyze the reaction product of in-flight reduction, by X-ray, SEM-EDS and electron microprobes, and conjecture the reduction mechanism(s) for the reduction of chromite and taconite [14]. Two reduction mechanisms have been postulated based on the production of microchannels of plasma in larger mineral particles, e.g. greater than 100 µm for chromite (Fig.5a), and the formation, slag-metal microcells in the finer mineral particles, e.g., less than 50 µm for taconite (Fig.5b). Microchannels of plasma in large particles, produced due to outgassing through microcracks or channels developed on being subjected to the turbulent plasma medium and very high heating rates, are proposed to act as highly reactive areas along which reduction of mineral is initiated giving rise to fine spherules of liquid metallics. Given sufficient time within the plasma and continued efficient plasma particle interaction, these liquid metallic spherules coalesce in order to reduce their surface energy, eventually forming large spherules and globules of reduced metallics saturated with carbon. The smaller mineral particles tend to melt completely on introduction into the plasma medium, thereby partly or completely inhibiting the formation of microchannels of plasma within the particles. In this condition very fine slag-metal microcells are established within the plasma. Such microcells provide favourable reduction kinetics and as such effect the necessary reduction reactions producing metallic products embedded in a slag phase. The slag phase will be controlled by the composition of the ore and any flux additions, e.g. SiO₂ or CaO, made to the feed material. Since the metallic phase will usually have higher melting point than the slag phases, the metallic phase will solidify first and will be surrounded by a slag phase. It is also likely that reduction of mineral, e.g. Fe₃O₄, will proceed through one or several intermediate stages during reduction, e.g., Fe₃O₄-FeO-Fe. Therefore, it may be expected to find the metallic phase situated predominantly in a partially reduced mineral or slag phase.

The limiting particle size which determines which of these reduction mechanisms predominates is likely to depend on the thermal and physical properties of the mineral, and it is also feasible that both of these mechanisms may operate within a mineral system. Open Bath Smelting : Kinetics considerations, which are relevant to the conventional processing in submerged arc bath, are also applicable during plasma processing. The transfer of heat from plasma arc to the bath and bath circulation has been modelled by Szekely [11]. A large proportion of plasma energy is dissipated in the high temperature pumped gas diverging from the anode attachment region; and in a contained furnace environment, a complex interaction of the convective component, the radiative component, the evolved process gases and the liquid slag bath would be expected. Figure 6 illustrates the general arrangement of an open bath plasma furnace and behavior of the various feed particles in the liquid slag. Equations 1-3 marked in figure correspond to the carbothermic reduction of a metal oxide, in the presence of a silicate slag, as exemplified below :

$$C + (MO) \rightarrow [M,C,Si] + CO\uparrow Eq. 1$$

$$(MO) + [M,C,Si] \rightarrow [M,C,Si] + CO\uparrow + SiO\uparrow Eq. 2$$

$$C + (SiO) \rightarrow SiO\uparrow + CO\uparrow Eq. 3$$

Mass transport and kinetics of reaction will very strongly depend on the local temperature, and given the high temperature, high power-density nature of plasma ares, large temperature gradients are the essential feature of bath smelting configuration. Use of only the surface of the slag in the furnace as the reaction zone imposes a constraint on the reaction rates. The distribution of the feed particles throughout the slag volume would be expected to greatly increase the reaction kinetics and, consequently, capability of the furnace to accept higher feed rates. Such distribution could be achieved in a number of ways, e.g. by stirring of the bath with inert gas, pneumatic injection of the feed material into the bath via a vertical lance, or bottom or side tuyeres, electromagnetic stirring of the bath etc [6,15].

Slag Chemistry

During smelting in conventional submerged arc furnace, the dual functions of slag as electrical load and as chemical reaction site are not complimentary in specific cases, e.g. smelting of ferro manganese. An acid slag $(SiO_2+Al_2O_3)$ is more resistive and is required for effective energy input and higher production rates. On the contrary, a basic slag (CaO+MgO) increases the activity of iron and manganese oxide in the slag phase and, consequently, promotes reduction and results in higher yields. This problem is overcome in a plasma furnace because energy input to a plasma furnace is not dependent on the slag resistivity and, therefore, slag chemistry can be optimized for reduction reactions. Typically, for the smelting of a manganese ore from South Africa in a transferred arc DC plasma furnace at MINTEK, the basicity was varied between 1.3 and 1.5 and the corresponding values of the manganese distribution to the metal/slag/vapour phases were found to be 63/20/17 and 79/10/11, respectively. The MnO content of the slag at 1.5 basicity decreased to about 10 percent compared to about 30 percent for conventional submerged arc operations [1].

Energy Related Issues

Power Input and Jurnace Type .

Power delivered to an arc furnace is the product of current and voltage. Figure 7 shows the

current, voltage and power relationships for some typical commercial plasma arc systems, used in ferroalloy furnaces, from high current low voltage end to low current high voltage end. The operating voltage of nontransferred plasma torches (AC or DC), e.g. as used in the gas heater for SKF shaft furnace, are higher than for transferred arc torches. DC transferred arc technology is widely preferred for metallurgical processing, e.g. ferroalloy smelting. DC transferred arc furnace using non cooled hollow graphite electrode, e.g. MINTEK furnace, operates at higher current and power level as compared to water cooled tungsten cathode, e.g. Tetronics Design, furnace.

Energy Requirement and Distribution

The gross energy required for plasma smelting includes the following components :

- 1) The net energy required for reduction reaction, i.e. the energy required to heat the reactants to the reactor temperature plus energy for any change of state (e.g. solid to liquid changes, coal devolatization etc) and heat of reaction
- 2) Energy required to heat any nonreactant gas used for arc stabilization, gases for conveying the solids, slag additives, cooling water for arc heater electrodes and losses to the crucible.

The energy requirement (excluding losses), which is of the order of 0.5 MWh/t for the melting of scrap, may vary between 3 to 8 MWh/t for the ferroalloys [5]. Figure 8 shows the power distribution for the processing of high carbon ferro manganese fines in a Hi-plas Davy McKee furnace. About 80% arc power may be utilized by the process energy (sum of sensible heat and energy involved in the chemical reaction) [16]. Percentage contribution of reaction energy increases with increase in the oxygen content of fines. During production of ferroalloys, the heat of reaction is a substantial fraction of the net energy and may equal to the energy required to heat the feed upto the reactor temperature. The capacity of a plasma heater to provide a high heat flux at high reaction temperature is an important factor that can minimize the capital and operating costs of a plasma reactor compared to other reactor (Fig.9).

Energy Efficiency

It has been found that at the same throughput rate and power flux, the efficiency of open bath plasma furnace is lower than that of a submerged-arc furnace. Factors responsible for this are:

- 1) more energy is lost, by radiation from open arc and molten bath, to the wall and roof of the furnace,
- 2) some vaporized material is lost to the off-gas stream from the arc attachment zone, and
- 3) little of the sensible energy of the gases evolved is utilized in preheating.

The energy efficiency is directly related with the throughput rate (Fig.10) and can be increased by increasing the throughput rate. Plasma furnaces in which both in-flight mode and open bath configurations are coupled also shows higher efficiency. It has been further observed that integration of prereduction/preheating step with a plasma smelting process can also result in considerable improvement in energy efficiency. For the smelting of South African manganese ores for the production of high C ferro manganese in a Noranda type plasma reactor (Fig 3c), researchers at Davy McKee has reported that ore pretreatment results in reduction of approximately 45% in electrical energy and 35% coke over the process in which no ore pretreatment is carried out; energy required with and without treatment were 2100 kWh/t and 3850 kWh/t, respectively [17]. The integrated ore reduction pretreatment and plasma smelting process consumes only 2100 kWh/t energy in comparison with conventional submerged arc-furnace which consumes 3100-3200 kWh/t for a similar ferroalloy and slag composition and no recovery of the energy from furnace gas [17].

Advantages Over Conventional Process

The production of ferroalloys by plasma technique offer several advantages over smelting in conventional submerged arc furnace :

- a) no need for costly sizing, screening and agglomeration of fines.
- b) no problems due to electrical conductivity of the carbonaceous reducing agent and the slag, since the plasma furnace operation is not affected by an uncontrollable rate of descent of the feed in the hot zone, as in an electric furnace.
- c) much lower cost of installation of power supply for plasma furnace (typical figure are \$ 165 and 360 per installed kilowatt for plasma and conventional furnace, respectively).
- d) no need for very large vessels to accommodate the large volume of unreacted burden.
- e) no need for lumpy metallurgical coke; any carbonaceous reducing agent can be used, provided it will not contribute to undesirable impurities in the product.
- f) trivial cost of replacement (including down-time) compared to cost of consumable cathode used in arc furnaces, despite the fact that the cathode used in transferred-arc furnaces have a limited life (200-400 h)
- g) no flicker on the power grid
- h) little noise and gaseous pollution and less environmental problems

Relevance in the Indian Context

The ferroalloy industry in India is characterized by low capacity utilization. Any further addition in capacity based on a new process flow sheet, using existing or new raw materials, is unlikely in near future. The electrical supply characteristics and geometric arrangement of the transferred arc plasma furnace with graphite electrode are similar to conventional submerged arc furnaces and change to DC plasma furnace operations is relatively straightforward. For example, the modification or retrofit of an existing MSA submerged arc furnace, used for ferro chromium production, has been successfully attempted by MINTEK in South Africa. For a retrofit process the economic advantage will be primarily in lower costs of raw materials, improved furnace, operations and in higher yield. In comparison to submerged arc furnaces, it will be possible to use cheaper ores or oxide concentrates and cheaper reductant. There will be furnace savings from decreased electrode consumption and smoother electrical operations and improved furnace control.

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Product	Company	Process	Capacity MW	
1			104	tpa
Iron Direct Reduction	SKF Sweden	SKF Torhes Gas Reforming	6	7
Iron Direct Reduction	USCO South Africa	Huels Torches Gas Reforming	3x8	3 2
Pig Iron	SKF Sweden	Coke-Filled Shaft SKF Torches	. 7x6	2 5
Pig Iron	ASEA South Africa	Transferred Arc Graphite Cathode	40	6 0
Steel	Mannesmann, GHH, Germany	Transferred Arc Graphite Cathode	6	-
Steel	ASEA/Krupp, Germany	Transferred Arc Graphite Cathode	18	
Steel	V E Freital, Germany	Transferred Arc Multiple Torches	3x3	•
Steel	V E Freital, Germany	Transferred Arc Multiple Torches	4x3	-
Steel	Voest Alpine, Linz, Austria	Transferred Arc Multiple Torches	4x7	-
Ferro- Chrome	Middleburg Steel and Alloy South Africa	Transf. Arc Hollow Graphite Cathode	20	5
Ferro- Chrome	SKF Sweden	Coke-filled Shaft SKF Torches	4x6	6
Ferro- Manganese	Samancor, South Africa	Transferred Arc	10.8	3
TiO _{2'} Slag	Richard's Bay Mining , South Africa	Graphite Electrodes A C Transferred Arc	6x10	5 0

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Table 1 : Commercially Operating Plasma Installations for Iron and Steel Production

Reactor Type	Ferro Alloys For Which Used	Originator/user		
DC Transferred arc furnace	FeCr, FeMn, FeMnSi	MINTEK South Africa		
DC Transferred arc furnace	FeCr	Middleburg Steel and Alloys, (MINTEK, ASEA Desgn)		
DC Transferred arc furnace	FeCr	ASEA, Vasteras, Sweden		
DC Transferred arc furnace	FeMn fines	*Samancor, Voest Alpine Design		
DC Transferred arc furnace	FeMn fines, FeCr	Davy Mckee, UK (Hydro-Qubec Noranda Design, Canada)		
DC Transferred arc furnace	FeCr	Tetronics R & D Ltd., UK		
Falling film reactor	FeCr, FeV	Bethlehem Steel Research Lab., USA		
Extended arc flash reactor (EAFR)	FeCr dust	University of Torento, Canada		
In-flight plasma reactor	FeCr, FeTi	University of Minnesota, USA		
Coke filled shaft furnace	FeCr, FeMn, FeSi	SKF Sweden		
**	FeCr	Swede Chrome AB Malmo, Sweden		
Packed bed reactor	FeMn	*SKW Trostberg, Sweden (Voest Alpine Design)		
Plasma augmented blast furnace	FeMn	SFPO, France		
RF plasma reactor	FeMo	McGill University, Canada		

Table 2 : List of Different Types of Plasma Reactor, Ferro Alloys for which Used and Originator or User

(Ferroalloy production by carbothermic reduction in oxide/carbide/metal system)					
Reaction Final Product	(1) Carbide	(2) Metal	(3) Metal		
System	Temperature °C				
Fe ₂ O ₃ /Fe ₃ C/Fe	644	640	571		
SiO ₂ /SiC/Si	1464	1551	1733		
Cr2O3/Cr23C6/Cr	1276	1255	1734		
MnO/M ₇ C ₃ /Mn	1273	1401	1762		
VO/VC/V	1048	1515	1947		
Al ₂ O ₃ /Al ₄ C ₃ /Al	1923	2036	2271		
CaQ/Ca ₂ C/C	1848	2152	2274		

Table 3 : Minimum temperature for $\Delta G=0$

Table 4 : Minimum Plasma Reactor Operating Temperatures, T_{Reactor}

Temperature °C								
System	T _{ΔG} =0	T _{M,metal}	Oxide	T _{M,oxide}	T _{M,Fe/x}	x	%. X	T _{Reactor}
Fe	644	1536	Fe ₃ O ₄	1597	1147	C	4.3	1536
Si	1733	1413	SiO ₂	1723	1200	Si	20	1733
Cr	1734	1898	Cr ₂ O ₃	2266	1510	Cr	20	1898
Mn	1762	1244	MnO ₂	1564	1232	Mn	87	1762
v	1947	1912	V ₂ O ₃	1970	1470	V	30	1947
Al	2271	659	Al ₂ O ₃	2050	1232	Al	42	2271
Ca	2274	850	CaO	2614	-	-	-	2274





Fig.1 Different kinds of plasma systems (a) nontransferred arc plasma [1,p77]

- (b) transferred arc plasma [1,p77]
- (c) in-flight plasma [1,p57]



- kinds of plasma systems
- (a) MINTEK open bath furnace [1,p145]
- (b) Uniersity of Minnesota in-flight plasma reactor [1,p85]

Contd. ...





Fig.2 Contd.

- (c) Davy Mckee Hi-plas furnace [16](d) University of Toronto extended arc flash reactor [3]

(d)

(c)



Fig.3 ΔG° -T data for carbothermic reduction of several metal oxides [10]



Fig.4 Velocity and temperature isocontours for a typical nitrogen/ hyodrgen plasma jet; r-radial direction and z-axial direction [12]







- (a) production of microchannels of plasma in large mineral particles; and
- (b) formation of slag-metal microcells in finer mineral particles which may tend to melt completely on introduction into the plasma medium



Fig.6 General arrangement of an open bath plasma furnace and behaviour of various feed particles in liquid slag [6]



Fig.7 Current voltage and power relationship for some commercial plasma arc systems [1,p81]



Fig.8 Power distribution for the processing of high carbon ferro manganese fines in a Davy McKee Hi-plas furnace [16]



Fig.10 Efficiency relationship for a hypothetical 30 MW DC transferred arc furnace with a rate of loss of energy of 3 MW and operating on a process having an ideal energy requirement of 1 MWh/t [5]