

PRETREATMENT UNIT IN FERROMANGANESE PRODUCTION

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ABSTRACT

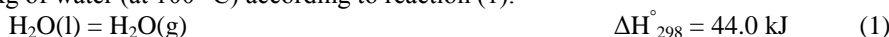
In many metal producing processes, a prereduction unit that will pre-reduce the iron and dry the raw materials will reduce the electrical consumption per ton of alloy. This is not obvious in ferromanganese production as the reduction of higher manganese oxides to MnO is exothermic reactions. This paper will study some examples of using prereduction units versus a standalone electric furnace. Mass and energy balances are calculated to show that the heating is the main advantage of using a prereduction unit. For every 100 degrees of increased temperature, the electrical energy consumption will be reduced by more than 80 kWh/ton. There is no extra savings in having an increased degree of prereduction in the prereduction unit. By heating the raw materials to 600 °C and evaporating the water, the savings in energy consumption can be more than 20%. In addition to reduced energy consumption, there are other advantages with a separate prereduction unit like more stable operation and better control of the carbon balance.

Key words: Ferromanganese production, pretreatment

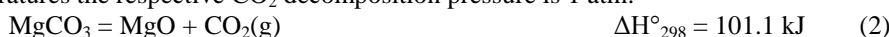
INTRODUCTION

Manganese alloys are mainly used in the steel industry and consist typically of manganese, iron and carbon. Silicomanganese alloys will in addition contain some silicon, typically between 15 and 30%. The production of silicomanganese alloys is not further discussed in this paper, though the trends obtained in energy reduction for ferromanganese alloys will also apply for silicomanganese production.

The raw materials used for ferromanganese production are manganese ores, fluxes and carbon. Also typically re-melt is added. Re-melt is metallic ferromanganese that cannot be sold, either due to the fact that it is mixed with slag or that the size is not sellable. When the charge mix of manganese sources, fluxes and carbon is heated in a reducing atmosphere, a variety of chemical reactions will occur. At low temperatures the water will evaporate, consuming 0.63 kWh/kg of water (at 100 °C) according to reaction (1).

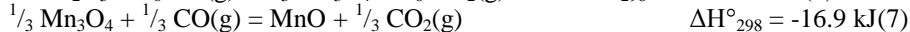
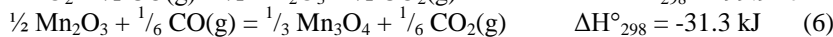
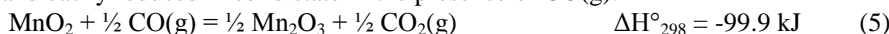


At higher temperatures the carbonates will decompose, also consuming energy. Decomposition of MgCO_3 occurs at about 300°C and for CaCO_3 at about 900°C. Dolomite, $\text{CaMg}(\text{CO}_3)_2$, decomposes at around 600°C. At these temperatures the respective CO_2 decomposition pressure is 1 atm:

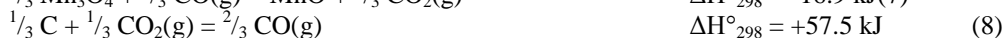
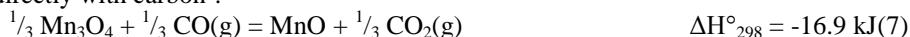


A mixture of 50% MgCO_3 and 50% CaCO_3 will at 600 °C use 40 kWh per 100 kg carbonates at decomposition. If the decomposition takes place above 800 °C, the produced CO_2 may react with C according to the Boudouard reaction, increasing the energy consumption even more.

The higher manganese oxides that predominate in manganese ores (MnO_2 , Mn_2O_3 and Mn_3O_4) are relatively unstable and easily reduced in solid state in the presence of $\text{CO}(\text{g})$:



These are exothermic reactions producing a considerable amount of heat, and thereby preheating the charge materials in the furnace. The CO_2 produced from reaction (5) and (6), will leave the furnace as CO_2 . Some of the CO_2 from reaction (7) will however be produced at temperatures above 800 °C where it will react with carbon, according to the Boudouard reaction, reaction (9). The total reaction between reaction (7)+(8), is reaction (9), where Mn_3O_4 is reduced directly with carbon in an endothermic reaction. In industrial furnaces typically between 40 and 90% of the Mn_3O_4 will react directly with carbon¹.



As the degree of prereduction will affect the total energy consumption, a number of articles on how the various ores are pre-reduced are previously published²⁻⁸.

The reduction of the iron oxides to metallic iron may be reduced in solid state with CO gas.

The reduction of MnO is dependent on a high temperature as well as carbon as reducing agent. The ores will start to melt at temperatures above 1250 °C, and the manganese metal will be produced according to the reaction (10) and the metal will be saturated with carbon.



The energy consumption in producing 1 ton of liquid alloy 76%Mn, 17.5%Fe, 6.5%C at 1400 °C from MnO, FeO and C at 25 °C will always be 1634 kWh/ton of alloy. Hence, the variations in energy consumption within a furnace and between different furnaces are due to variations in the reactions in the prereduction zone, which is reaction (1) to reaction (9).

Due to the exothermic reduction of higher manganese oxides, reactions (5), (6) and (7), there has been less focus on a separate pre-reduction unit in manganese productions compared to other processes like e.g. ferrochromium production. The exception is CDK Kashima Works in Japan, 100 km north east of Tokyo, where a rotary kiln in series with the electric furnace has led to reduced energy consumption as one of the advantages. The exhaust gas from the electric furnace is then used as kiln fuel. Kashima Works produces high carbon ferromanganese from a variety of different Mn-sources. This paper will discuss the electric energy consumption and how it varies with varying pre-reduction in the pre-reduction unit. This will be done through theoretical heat and mass balances based on operational practice. All enthalpy data is taken from the database of HSC Chemistry¹¹.

ELECTRIC ENERGY CONSUMPTION

At Kashima Works, a number of various Mn sources are used - from high oxide Groote Eylandt ore, South African Mn₂O₃ ores to sinter, where the manganese oxide is close to Mn₃O₄. The base case used in the calculations, is a charge from 2011 where the basis is AHGL and BHP sinter, where additional 15% BHP GL is tested. The temperature of the kiln was kept at 600-700°C in the test, as one would not increase exhaust gases volume and change the kiln operations. In addition both limestone and dolomite is used as fluxes. Coke is used for reduction in the electric furnace. Some amount of coal is used as fuel in the pre-reduction unit but the main fuel is the electric furnace exhaust gases. This process has been modeled to calculate the electric energy consumption, and the principles of the model are shown in Figure 7. Some remelt is added directly to the electric furnace in addition to the materials coming from the pre-reduction unit. It is assumed that the temperature of the materials entering the electric furnace is 600 °C and the slag and metal leaves the furnace at 1500 °C and the off-gas - at 400 °C. Tanabe¹⁰ presented the results of using a prereduction unit during pilot scale testing. In their experiments, the materials were cooled dramatically between the pre-reduction unit and the electric furnace. In the calculations presented here, it is only assumed a reduction in temperature down to 600 °C before the materials enter the electric furnace.

The temperatures chosen for the material flows in and out of the electric furnace will of course affect the electric energy consumption. Figure 8 shows the difference in total kWh consumption per ton of alloy if we vary the temperatures of the incoming and outgoing materials. By increasing the temperature of the raw materials with 100 Kelvin, e.g. from 600 to 700 °C, the electrical energy consumption will decrease with about 80 kWh/ton of alloy. This already shows one of the big advantages of a pre-reduction unit. Instead of adding raw material at room temperature, heating the raw material up to 600 °C (without any chemical reactions) would decrease the electric energy consumption between 400 and 500 kWh/ton of metal, that is around 20% of the total energy needed, compared to the process with no prereduction unit used. This also states that the term “pre-reduction” unit does not cover the intent, as one of the main purposes of the prereduction unit would be to heat the raw materials and not necessarily pre-reduce the material. The assumptions of outgoing temperatures from the furnace do not have the same effect on the total power consumption. One hundred degrees difference in off-gas temperature would only change the power consumption with about 25 kWh and hundred degrees difference in the temperature of slag and metal would account for about 60 kWh per ton of alloy as seen in the figure.

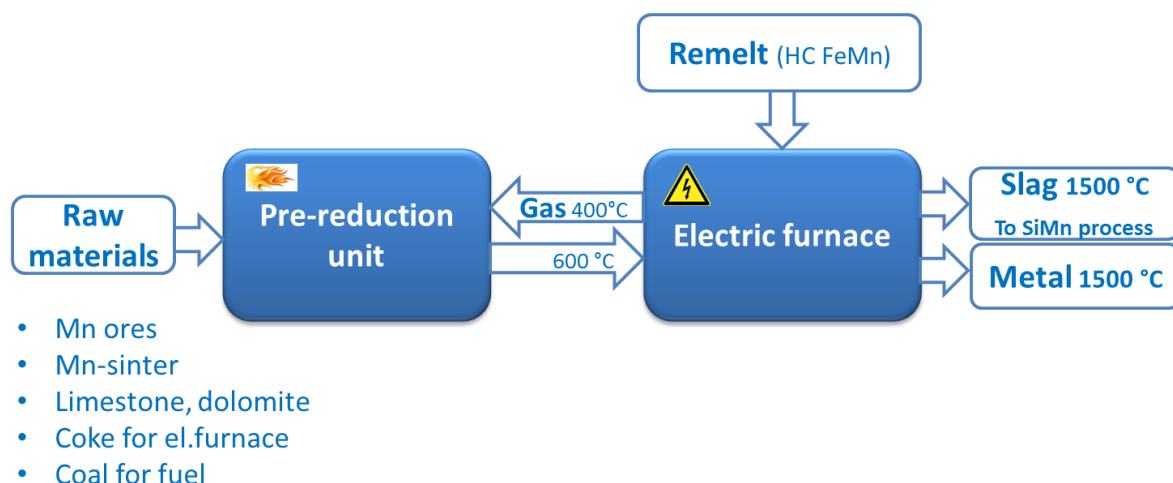


Figure 7. Overview of the material flow and temperatures assumed in the process

Some clarifying remarks must be done of the term “power consumption” given in kWh/ ton of metal. There are at least three different terms of kWh/ton metal:

1. **Theoretical power consumption:** The power consumption is the difference between the energy in the materials coming into the furnace and the energy in the materials leaving the furnace. By set temperatures and composition of the raw materials coming into the electric furnace and set temperatures and composition of the exit materials, the reaction enthalpies and heat capacities will determine the amount of electrical energy that must be added.
2. **Real power consumption:** Some of the electrical energy is lost in the electrical system as well as some energy is lost as heat in the system. Typically one can assume a yield of 80%, which means that 20% of the energy is lost. It also means that the theoretical power consumption will be 80% of the real power consumption. In this paper the numbers given for power consumption is not the theoretical power consumption, but the real power consumption based on a 20% energy loss.
3. **Power consumption for virgin material:** There are two ways to describe the amount of metal produced: first, the total amount of metal coming out of the furnace, and secondly, the metal produced through reduction. The total amount of metal coming out of the furnace is the sum of metal produced and remelt added. The metal produced from oxidic raw materials is often referred to as “virgin” material. As the total power consumption is very much dependent on the amount of remelt added to the furnace, this issue must be taken out of the equation if one would like to compare the quality of the operation based on the power consumption. Hence, for a furnace producing 100 tons of alloy per day where 10 tons are remelt, the power consumption of 2000 kWh/ton alloy would equal to 2222 ($=2000/0.9$) kWh/ton of virgin metal. In this paper both the real kWh/ton is given as well as the kWh/virgin ton of metal.

In this paper the metal loss in the slag is not included, and hence if the metal loss in the slag is e.g. 5%, the total power consumption will increase with a bit more than 5% ($=1-1/0.95$) compared to the numbers in this paper.

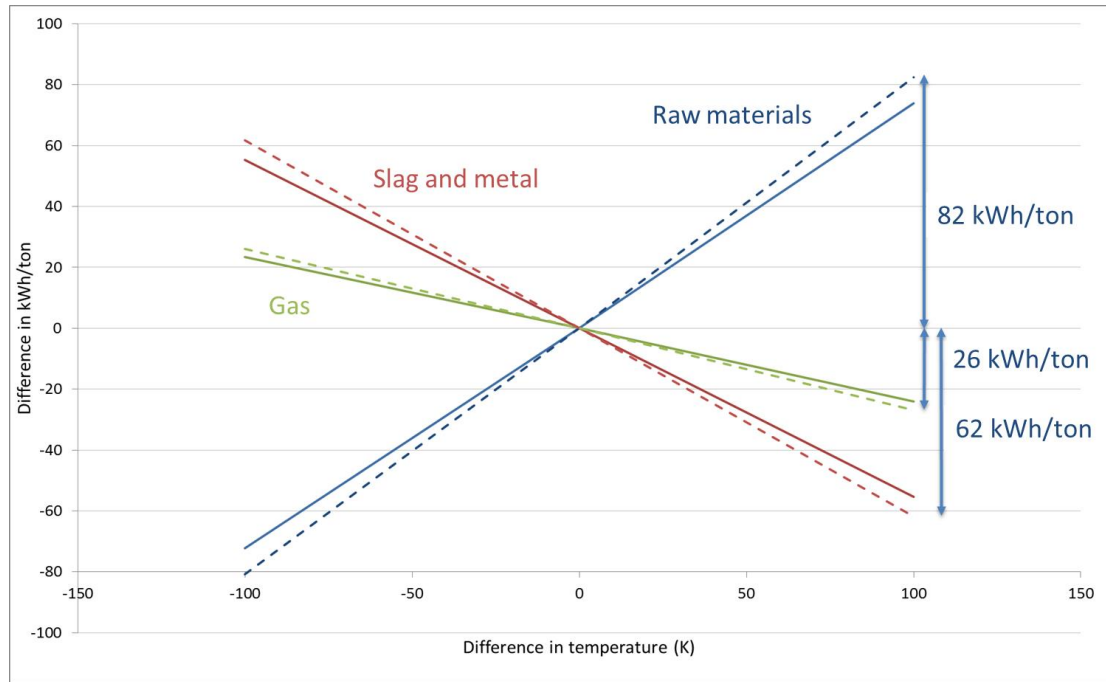


Figure 8. Variation in kWh with variations in temperature of material flows in and out of the furnace. Origo is the point of about 2000 kWh/ton of metal and temperatures of 600 °C on incoming raw materials, 1500 °C on slag and metal and 400 °C in off-gas. Solid lines are kWh/ton of metal and dotted lines are kWh/ ton of virgin metal.

One of the interesting questions of using a pre-reduction unit is how it affects the total electric energy consumption in the electric furnace. It has already been shown above that the electric energy consumption will decrease quite dramatically just by heating raw materials. In addition, the heating will also affect some of the energy producing reactions (e.g. reduction of higher manganese oxides) as well as energy consuming reactions (e.g. water evaporation and carbonate decomposition). To investigate this, the raw material mix of Kashima Works has been used as a case study. The material flows are shown in Table 7. The raw materials entering the prereluction zone are grouped as the different oxides and basic elements. In addition 10 wt% moisture is assumed in the raw materials. Based on three different degrees of prereluction in the prereluction unit, the manganese oxides and iron oxides are reduced to one of the following cases 1) $Mn_2O_3+Fe_3O_4$, 2) Mn_3O_4+FeO , 3) $MnO+Fe$. The slag and metal produced are calculated based on a metal composition of 74%Mn, 19.5%Fe and 6.5% C, which are quite close to the industrial practice. The rest of the materials enter the slag. As no material losses are assumed in the calculation, e.g. to the off-gas, the Mn content in the slag was too high compared to the industrial practice. Hence, the manganese oxides in the raw material were decreased with 5% to obtain the slag close to the industrial slag composition. In the last case where the oxides were reduced to MnO and Fe, there is no carbon consumption according to reaction [9] due to no direct prereluction in the furnace. The total coke consumption was then reduced from 324 kg/ton of virgin alloy to 248 kg/ton of virgin alloy.

The metal composition at Kashima Works is following the specifications of Japanese industrial standard (JIS) No. 1, that is 72-78% Mn, which is somewhat lower compared to European producers producing typically above 78%. As the Mn/Fe ratio in the raw materials is decreasing over time, there is an ongoing discussion whether the Mn content in the metal should be lowered in the specifications as well.

The electric energy consumption for the cases of increased prereluction in the prereluction unit is summarized in Figure 9. For all the cases, energy loss of 20% is assumed, and both energy consumption cases per ton of alloy and per ton of virgin alloy are shown. The first case is where no pre-reduction unit is used and the raw materials are added to the furnace at room temperature. It will give an energy consumption of about 2400 kWh/ton of alloy. By using a pre-reduction unit, where the materials are heated to 600 °C and the water is evaporated, but where no reduction of manganese oxides occurred, the electric energy consumption is reduced to 1800 kWh/ton of alloy. The removal of water in the prereluction unit accounts for more than 200 of the kWh used, which is more than 10 % of the total energy consumption. The rest of the decreased energy consumption is due to the increased temperature of the raw materials. In the pilot scale experiments reported by Tanabe¹⁰, the electric energy consumption was reduced from 2940 kWh in a standalone electric furnace to 2680 kWh at 550°C in the production unit, 2543 kWh at 650 °C and 2340 kWh at 950°C. As their material was cooled dramatically between the prereluction unit and the electric furnace, the reduced energy consumption they observed was probably due to a more stable operation as well as some heating, as will be discussed later.

Table 7. Material flow as raw materials enter the pre-reduction unit, three cases out of the unit, the remelt and electrode into the furnace, slag and metal analyses

GENERAL ASPECTS

Raw materials			Mn2O3	Mn3O4	MnO	Remelt and electrode		Out			Out		
			Case 1	Case 2	Case 3			Metal	Metal	Metal	Slag	Slag	Slag
			Out	Out	Out			%	kg	kmol	%	kg	kmol
Inn	kg	kmol	kmol	kmol	kmol	kg	kmol						
MnO2	1805	20.8											
Mn2O3	6651	42.1	52.5										
Mn3O4	2818	12.3	12.3	47.3									
MnO	231	3.3	3.3	3.3	145.2						45.6	2463	34.7
Fe2O3	1971	12.3											
Fe3O4			8.2										
FeO	257	3.6	3.6	28.3									
CaCO3	219	2.2											
MgCO3	64	0.8											
Mn						688	12.5	74	6760	123			
SiO2	1306	21.7	21.7	21.7	21.7	11	0.2				24.4	1317	21.9
Fe					28.3	203	3.6	19.5	1781	32			
CaO	852	15.2	17.4	17.4	17.4	4	0.1				18.1	978	17.5
MgO	132	3.3	4.1	4.1	4.1	1	0.0				3.0	164	4.1
Al2O3	474	4.6	4.6	4.6	4.6	5	0.0				8.9	478	4.7
C	2306	192.2	192.2	192.2	192.2	145	12.1	6.5	594	49			
C(fuel)	642	53.5											
H2O	1975	109.6	0.0	0.0	0.0								

The decomposition of carbonates is endothermic and will increase the electrical energy consumption, if the decomposition occurs in the electric furnace. As stated previously, the decomposition of carbonates will amount to about 40 kWh per 100 kg of carbonates, or more if the decomposed CO₂ reacts according to the Boudouard reaction. However, the content of carbonates in this charge is quite low (1%) and hence the total electrical energy consumption for this case will only be affected by about 1% if the decomposition occurs in the prereluction unit or in the electric furnace.

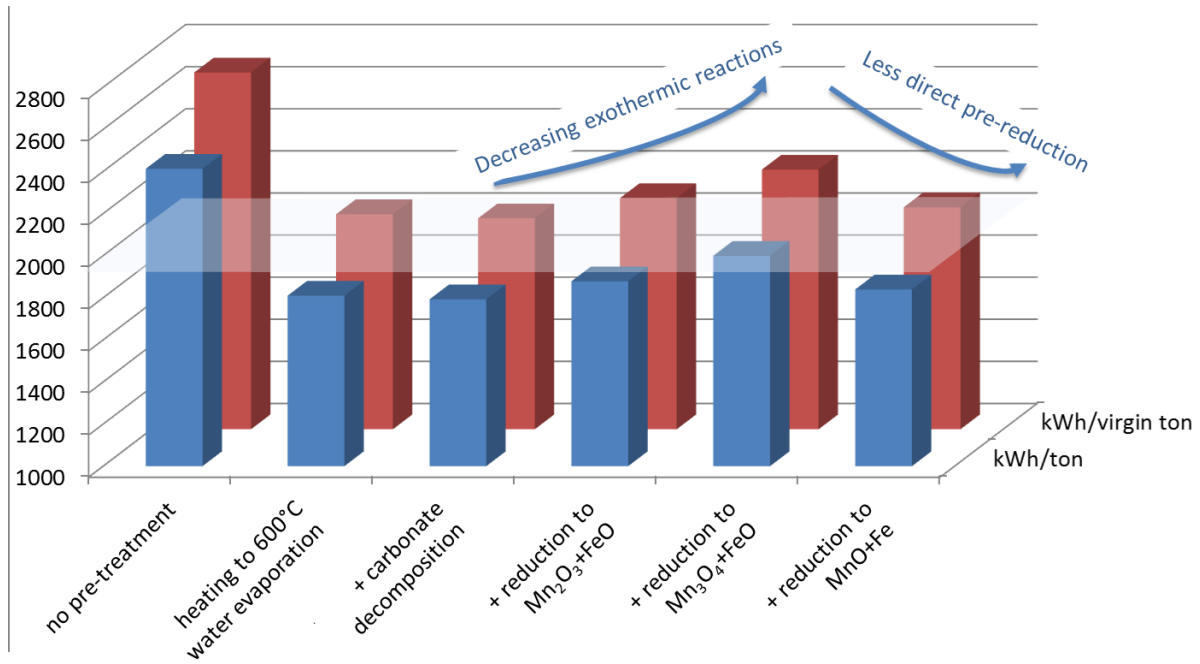


Figure 9. Electric energy consumption per ton of alloy and per ton of virgin alloy with the assumption of 20% energy loss. Text below is the extent of pre-treatment. 2000 kWh/ton of alloy is indicated, as this is close to the energy consumption at Kashima Works

If the prereluction starts to occur in the prereluction zone, the higher manganese oxides will be reduced to lower manganese oxides, MnO₂ to Mn₂O₃ to Mn₃O₄ to MnO as described above in reaction 5, 6 and 7. At the same time the iron oxides will be reduced from Fe₂O₃ to Fe₃O₄ to FeO and Fe. The reduction of manganese oxides is exothermic and hence, an increasing degree of prereluction in the prereluction unit will increase the electrical power consumption in the electric furnace, as seen in Figure 9. The exception is when the reduction in the prereluction unit will reduce the charge all the way down to MnO and metallic iron. In this case there will be no endothermic Boudouard reaction (reac-

tion 8) where CO_2 reacts with carbon, as there will be no CO_2 developed from any reduction of higher manganese oxides. This will lead to reduced electric energy consumption.

One of the assumptions used in these calculations is that the material enters the electric furnace at $600\text{ }^\circ\text{C}$, which means that the material leaves the prereduction unit at a bit higher temperature, e.g. $700\text{ }^\circ\text{C}$. The possible extent of prereduction in the prereduction unit is given by the thermodynamics and the kinetics of the reactions 5, 6 and 7. The thermodynamic conditions for the reduction of higher manganese oxides are shown in Figure 10. If solid carbon is present, determining the oxygen pressure, the oxygen pressure will be less than 10^{-15} in the whole temperature area. This means that the thermodynamic suggests that it will be possible to get down to MnO at $700\text{ }^\circ\text{C}$ in the prereduction zone at a low oxygen pressure. This will be the case all up to an oxygen pressure of 10^{-12} , that is a $p_{\text{CO}_2}/p_{\text{CO}}$ ratio of $4 \cdot 10^4$. Hence, thermodynamically it is possible to reduce the charge down to MnO at $700\text{ }^\circ\text{C}$ even at higher oxygen levels than 10^{-15} . The reduction of higher manganese oxides is however in most industrial cases determined by kinetics, and thus can be controlled by time, sizing and temperature in the prereduction zone¹⁻⁸. The reduction of MnO_2 is however quite fast and happens at relatively low temperatures⁶. It is hence assumed that by some heating, the raw materials will soon come down to Mn_2O_3 and possibly Mn_3O_4 .

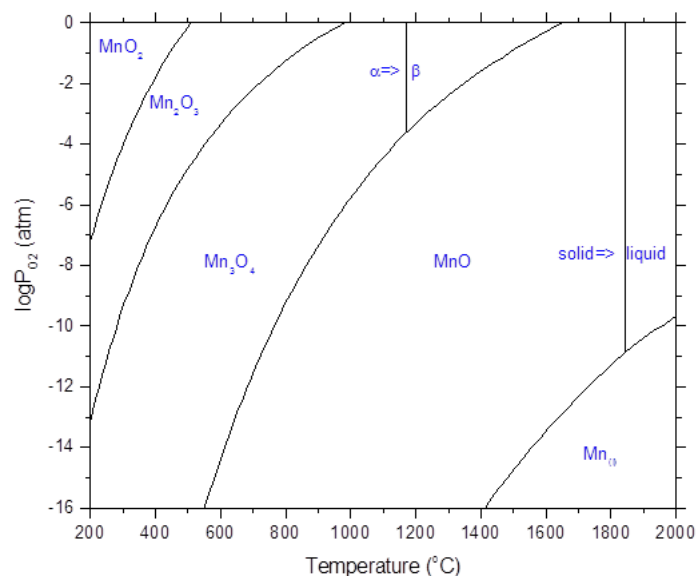


Figure 10. Calculated stability areas in the Mn-O system [Olsen et al. 2006]¹²

As a summary of the electrical power consumption given as kWh/ton of metal presented in Figure 9, it is seen that heating the raw materials in a prereduction unit will decrease the electrical power consumption in the area of 20%. However, if the prereduction starts to occur in the prereduction unit, the electrical energy consumption in the furnace will increase again. The strategy should then be to have as little prereduction as possible, or to reduce the ore all the way down to MnO . The last option will also reduce total carbon consumption in the electric furnace however it may at the same time increase the carbon consumption in the prereduction zone, and thus the total CO_2 emission. It must be emphasized, if the reduction of higher manganese oxides in the prereduction unit is not reduced to MnO , the carbon content in the electrical furnace will be constant. Hence, by using a prereduction unit, the requirement of high-quality carbon agents to use in the electric furnace will not change.

By comparing the operation of an electrical furnace without prereduction unit (2413 kWh/ton) with the operation using a prereduction unit and reducing the charge to Mn_2O_3 (1878 kWh/ton) the reduction is 22% lower energy consumption. This is described as case 1 and 4 in Figure 9. However, how much the energy consumption will decrease will of course be dependent on the type of raw materials. The average Mn-ores used in the above calculation is close to $\text{MnO}_{1.5}$ that is Mn_2O_3 . However, if in an open standalone furnace MnO_2 blend is used, the power consumption is much lower, that is about 1900 kWh/ton, and a prereduction unit would not lower the electric energy consumption. Figure 11 shows how the oxygen level in the ore affects the electrical consumption in a standalone furnace. It is seen that for all charges, except for the MnO_2 blend, there will be considerable electrical savings in using a prereduction unit. As for previous figure, both kWh/ton of metal and kWh/ton of virgin metal are included. A comment regarding the use of MnO_2 ores in a standalone furnace is as follows. The exothermic reduction of MnO_2 may cause hazardous operation in a closed furnace and hence the industry will typically have a safe standard for the amount of oxygen allowed in closed furnaces¹³. The advantages of using relatively high oxygen content in the raw materials were however reported by Tangstad et al.¹³.

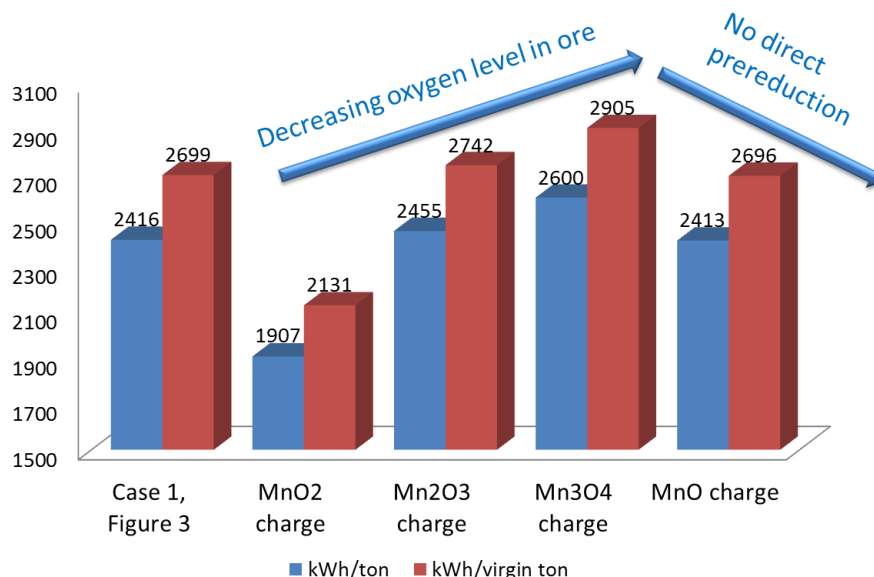


Figure 11. Electrical power consumption in a standalone electric furnace with decreasing oxygen content in the ore

There are a number of advantages using a prereduction unit. The one discussed so far, the lower electrical consumption per ton of metal, has a number of consequences:

- It will reduce the electrical energy cost.
- For a given furnace, with a given capacity, it will increase the throughput that is the tonnage per day.
- In places with difficult electrical situation (Japan, Brazil, South Africa, Korea, ...) the shift of using carbon as fuel instead of electric power will stabilize the situation. The carbon used in the prereduction zone is also quite flexible from natural gas, coal fines and other type of carbon materials. If bio-carbons can be used in the prereduction zone, this would also keep the CO₂ emissions low. CO gas can of course also be used, either from own production or from other industries.

There are also some advantages of using a prereduction unit in addition to the low energy consumption:

- As the water and low temperature volatiles are removed from the charge, more fines can be added to the furnace without the same hazardous consequences.
- In the same line of argumentation, the stability of the furnace will be higher if the raw materials are calcined, that is no water and low temperatures volatiles. This means that one will have a better control of the carbon consumption in the electrical furnace avoiding over-coking or under-coking. The increased stability of the operation will give higher degree of prereduction, thereby lower carbon consumption, better slag-metal separation, better electrode operation and higher operation yield in MWh/day.
- Tanabe¹⁰ reported a reduction of blows in their pilot scale campaign from 5-6 per day in a cold charge operation, to a reduction to 33-50% when the raw materials were heated to 550-650°C, to no blows at 950°C.

There are two main disadvantages using a prereduction unit. First, it will of course give extra cost as one extra process unit is used. The next is the total CO₂ emissions. As electrical energy is replaced with combustion of carbon, the total carbon emission may be increased compared to using a standalone furnace. However, this is all determined by the type of electrical energy used, and the type of carbon fuel used:

- If the electrical energy is nuclear or renewable energy from hydropower or sun, and the fuel used in the prereduction unit is fossil fuel, the total CO₂ emission will be higher using a prereduction unit.
- If the electrical energy is from carbon combustion power plants, the total carbon emission will be lower by using a prereduction unit, as the energy yield is lower when electrical energy is produced (~20-30%) compared to heat (~100%). In addition the total CO₂ emission will be lowered if the off-gas from the prereduction unit is used as fuel for electrical production. This is the case for Kashima Works which sends the furnace off-gases to Kashima Steel Works for the production of electric energy.
- The type of carbon used will of course also affect the total CO₂ emission, where coke has the highest CO₂ emissions, as CO₂ will also be emitted during the coking process. Coal and natural gas will give less CO₂ emission as they are not pre-treated in the same manner. Bio-carbon will of course give the lowest CO₂ emission, as the total cycle of growth and combustion will be close to zero. In a prereduction unit using bio-carbon waste materials from other industries would give very low CO₂ emissions as well as low electric energy consumptions in the electric arc furnace. Monsen¹⁵ showed how charcoal affected the ferromanganese operation.

CONCLUSIONS

In metal producing processes, many producers use a prereduction unit to decrease the electrical energy consumption. Typical processes that use prereduction units are ferrochromium production and ilmenite smelting. However, as ferromanganese production have more exothermic reactions in the low temperature area, the use of prereduction units is quite rare. Kashima Works in Japan, 100 km north east of Tokyo, is however one exception. In this paper, a typical charge from Kashima Works was used to examine theoretical energy consumption in the electrical furnace. It is shown that the major benefit of a prereduction unit is the heating of raw materials up to 600-700 °C including the water evaporation. The use of a prereduction unit will decrease the power consumption with 400-500 kWh/ton of alloy, which is about 20% of the total electrical energy consumption. This large reduction in energy consumption may be obtained with Mn_2O_3 ores and Mn_3O_4 charges. However, if a large amount of MnO_2 is used, the reduction of energy will be much less compared to the conventional standalone electric furnace.

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