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Energy distribution in HC FeMn and SiMn energy vs exergy analyses

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Synopsis

The metal producing industry is a high consumer of energy and large amounts of excess heat are produced. Increasing the energy efficiency would be beneficial, both in terms of the environment and also from an economical point of view. In order to do this, it is crucial to know how the energy is distributed throughout process operation. Energy (enthalpy) and exergy analyses were used to discuss the production of high-carbon ferromanganese (HC FeMn) and silicomanganese (SiMn). The two different analysis methods were compared to decide if exergy analyses provide a better understanding of the distribution and recovery potentials for the production process of HC FeMn and SiMn. It was found that the distribution of energy and exergy between the different material streams is highly similar and key potential recovery sites are the same regardless of the analysis method utilized. The additional information provided by the exergy analysis compared to enthalpy is the reduction in energy quality (exergy destruction) due to irreversible processes within the furnace. These values were found to be 13.7% for HC FeMn and 10.8% for SiMn.

Keywords

ferromanganese, silicomanganese, energy, exergy.

Introduction

Among many different manganese alloys, high-carbon ferromanganese (HC FeMn) and silicomanganese (SiMn) are the most common ones. Both alloys are produced by carbothermic reduction in submerged arc furnaces (SAFs) with three Söderberg electrodes submerged in the charge material. The processes are highly energy-consuming in addition to producing large amounts of excess heat. There are two companies producing ferromanganese alloys in Norway: Eramet and Ferroglobe (former Glencore Manganese Norway), the former of which reported an annual electrical energy consumption of 1.92 TWH in 2016 (Eramet-Norway, 2017). Today there is an increasing focus on environmental impact in the industry, both in terms of emission control and reduction of energy consumption. To optimize energy efficiency and increase resource utilization, it is crucial to know how the energy is distributed during operation. Commonly, energy distribution is evaluated in terms of an enthalpy balance based on the first law of thermodynamics, stating that energy is neither created nor destroyed. Thus, energy going in to a system equals the energy going out. The limitation of this method is that it does not consider the quality or dissipation energy. This implies that it may be an inadequate tool for determining key potential recovery sites for optimizing energy efficiency in industrial processes.

Another way of assessing a system is by exergy analysis, which is based on both the first and second law of thermodynamics. The second law states that the quality of energy is always reduced when being converted from one form to another, implying that an exergy balance considers the irretrievable losses of energy due to irreversible processes. Hence, exergy can be defined as the amount of energy that potentially may be utilized and is conserved only in completely reversible processes. Thus, energy can be considered a measure of quantity only, whereas exergy is a measure of both quantity and quality. Several papers have been published where the exergy method is utilized to identify the main sources of exergy loss in a process and to pinpoint key locations for potential recovery. Ayres *et al.* used exergy as a tool for resource and waste accounting for five different metallurgical industries (*e.g.* aluminium and steel) in the USA (Ayres, and Masini, 2006), whereas Hjartanson applied exergy analysis on a ferrosilicon furnace at Iceland when studying waste heat utilization (Hjartarson, 2009). Hjartanson determined the exergy destruction in the furnace to be 47.4%. Further, the exergetic efficiency was found to be about 30%, correlating with the chemical exergy found in the product. No other exergy streams, *e.g.* chemical and thermal exergy in off-gas, were utilized. Conventional energy

analysis showed that 46% (45.5 MW) of the output energy was found in the off-gas, whereas the exergy analysis showed that the available energy was only 18.4 MW. Børset evaluated the silicon production process and found that the exergetic efficiency could be increased from 0.33 to 0.41 if the thermal exergy in the off-gas was recovered(Børset, 2015).

Process chemistry

The production processes for high-carbon ferromanganese and silicomanganese in the submerged arc furnace are very similar; however, different raw materials are used and hence the products are different. Raw materials in HC FeMn are commonly manganese ore and/or sinter, coke, and fluxes. Similar raw materials are also used for SiMn, while quartz is added to obtain a sufficient Si content in the alloy and the end slag from HC FeMn production, with a relatively high content of MnO, is often used as a source of manganese in SiMn production. (Fe)Si-remelt or off-grade qualities may also be added.

The charge mixture enters the furnace at 25°C at the top, from which it starts to descend into the furnace. The temperature increases with the depth of the furnace and evaporation of water will be the first reaction to occur at temperatures higher than 100°C. Some of the water vapour will react according to the water-gas shift reaction $(H_2O(g) + CO(g) \rightarrow H_2(g) + CO_2(g))$, which is spontaneous at temperatures below 700°C. The manganese in the ores is present as different oxides that will reduce according to Equation [1-3] at increasing temperature:

$$MnO_2 + \frac{1}{2}CO(g) \rightarrow \frac{1}{2}Mn_2O_3 + \frac{1}{2}CO_2(g)$$
[1]

$$1/_{2} \operatorname{Mn_2O_3} + 1/_{6} \operatorname{CO}(g) \rightarrow 1/_{3} \operatorname{Mn_3O_4} + 1/_{6} \operatorname{CO_2}(g)$$
 [2]

$$\frac{1}{_{3}}Mn_{3}O_{4} + \frac{1}{_{3}}CO(g) \rightarrow MnO + \frac{1}{_{3}}CO_{2}(g)$$
 [3]

Reduction reactions of Mn oxides are all exothermic and the amount of energy produced is larger for a higher oxygen level.

Carbonates ($CaCO_3 \cdot MgCO_3$, $MgCO_3$, $CaCO_3$) are added to the charge as flux. Decomposition of $MgCO_3$ occurs at approximately 300°C and $CaCO_3$ at 900°C. $CaCO_3 \cdot MgCO_3$ (dolomite) will decompose at approx. 500°C (Olsen, Tangstad, and Lindstad, 2007). The decomposition reactions are endothermic, hence consuming energy.

In addition, iron is always present in manganese ores, reducing together with the manganese oxides:

$$1/{_2}Fe_2O_3 + 3/{_2}CO \rightarrow Fe + 3/{_2}CO_2$$
 [4]

The CO_2 formed from reduction of Mn_3O_4 and iron oxides and carbonate decomposition may react with carbon through the Boudouard reaction:

$$C + CO_2(g) \rightarrow 2 CO(g)$$
^[5]

The reaction is highly endothermic and causes an increase in the total carbon consumption of the process. The amount of CO_2 not reacting is defined as the degree of prereduction. Industrially, the degree of prereduction is typically between 0% and 50% (OlsenOlsen, Tangstad, and Lindstad, 2007).

At temperatures 1200-1400°C, the charge will have formed a liquid slag from which the final reduction of MnO to manganese metal will occur at the slag/coke bed interface. Similarly, SiO_2 will be reduced to silicon. The product metals are saturated with

carbon and the C content is thus 7% and 1.5% for a typical HC FeMn and SiMn alloy, respectively.

$$MnO(l) + C \rightarrow Mn(l) + CO(g)$$
[6]

$$SiO_2(l) + 2C \rightarrow Si(l) + 2CO(g)$$
^[7]

$$C \rightarrow \underline{C}$$
 [8]

The CO gas formed in the metal producing reactions will be partly consumed in the low-temperature zone in the reduction of the higher oxides. A typical HC FeMn alloy contains 78% Mn, 7% C and < 1% Si, whereas SiMn has a silicon content of 17-20%. Due to this, SiMn production requires a higher process temperature than HC FeMn. Typical temperatures are 1500°C for HC FeMn and 1600°C for SiMn.

This paper presents staged energy and exergy balances for HC FeMn and SiMn as a basis to evaluate the energy dissipation and determine the potential sources of recoverable energy. The objective is to assess whether the conventional approach of energy analysis is in adequate for evaluation of resource efficiency and recovery sites.

Staged material and energy balance

A simple material and energy balance considering input and output was executed in HSC Chemistry 9(Outotec, 2017) for HC FeMn and SiMn. The energy consumption of a process can be determined by considering the net effect of exothermic and endothermic reactions in the process, or by evaluating the enthalpy of the materials going in and out of the furnace. The energy used in the process will be transformed into heat or energy in the substance.

Assumptions applied to both processes are as follows:

- > Mixing enthalpies were not included for any of the species
- Losses related to furnace operation (heat loss, cooling water, fines *etc.*) were not included
- Prereduction degree was set to 25% (75% of CO₂ produced from reduction of Mn₃O₄, iron oxides and carbonate decomposition according to Boudouard reaction)
- ► Alkali circulation is not included
- ► Evaporation of Mn and Si was not evaluated.

HC FeMn

The charge mixture is based on two different manganese ores (ores 1 and 2) and sinter in a mass ratio approximately 1:0.4:1. Ore 1 and sinter are considered acidic (high in silica and alumina), whereas ore 2 is high in CaO. Average oxygen level is MnO₁₇ and a total of 17.95 kmol Mn is added. Dolomite is added as flux, corresponding to 10 wt% of the total input charge. Coke is used as reductant and the added amount gives a fixed carbon content of 358 kg. The composition of the raw materials is presented in Table I. A water content of approximately 10% in the raw materials was assumed. The relationship between the MnO liquidus and the slag basicity (CaO+MgO/ SiO_2 +Al₂O₃) has been calculated previously (Olsen, Tangstad, and Lindstad, 2007) and was used to predict the distribution of manganese between alloy and slag. The alloy will be saturated in carbon, correlating to 7%. Raw materials enter the furnace at 25°C, slag and alloy are tapped at 1500°C, and the off-gas is assumed to beat 200°C. The defined input of raw materials and corresponding output are presented in Table I.

Table IChemical composition of raw materials used as input inmaterial balance for HC FeMn

	Ore 1 [wt%]	Ore 2 [wt%]	Sinter [wt%]	Dolomite [wt%]	Coke & electrodes [wt%]
MnO ₂	77.12	32.98	18.93	-	
Mn ₃ O ₄	3.55	36.29	59.56	-	
Fe ₃ O₄	7.79	17.10	8.46	-	3.88
SiO ₂	7.12	7.53	9.36	-	5.58
Al_2O_3	4.06	0.20	3.41	2.27	2.63
CaO	0.20	5.40	0.10	-	1.00
MgO	0.15	0.49	0.20	-	1.00
CaCO ₃	-	-	-	62.63	-
MgCO ₃	-	-	-	35.10	-
С-	-	-	-	85.93	

The specified system has a total energy consumption, and required electric feed, of 2456 kWh per ton of alloy. The energy consumption will vary with operational strategies (*e.g.* slag basicity). Nonetheless, Olsen, Tangstad, and Lindstad (2007). reported an average energy consumption of 2152, 2400, and 3395 kWh/t alloy for three different submerged arc furnaces, which agrees well with the current calculation. The energy streams of the furnace are presented in a Sankey diagram in Figure 1. The streams are given both by specific energy content in kWh and as a percentage of the total.

The Sankey diagram shows that the energy contributions from the electrical power and carbon are comparable. It is seen that 79% of the total energy input is converted into chemical energy and the remaining 21% into thermal energy. The majority of the chemical energy is contained within the alloy, whereas 24.4% is in the off-gas and only 2.4% in the slag. The slag from the HC FeMn process can be used as a raw material in SiMn production, meaning that it may be considered a useful byproduct. This also enables recycling of metal potentially entrained in the slag phase, which always occurs to some extent. The largest recoverable energy source is the chemical energy in the off-gas at 1401 kWh. This energy is already commonly utilized in industry today. *e.g.* for electricity generation or resold to other industries(Eramet-Norway, 2017). Considering this, the loss of energy lies within the thermal energy in all output streams, equal to 21%. The main heat source is cooling of high-temperature alloy and slag, which combined contains 1005 kWh (17.5%) of the total energy output.

SiMn

The staged balance is based on raw materials comprising

Table II

Input and output of staged material balance for production of approx. 1 t of HC FeMn. All raw materials enter the furnace at 25°C. Output temperatures are 1500°C for slag and alloy and 200°C for off-gas

	INF	TUT	OUTPUT		
Species	kmol	kg	Species	kmol	kg
MnO ₂	9.50	826.16	Mn(l)	14.34	787.99
Mn ₃ O ₄	2.82	644.56	Si(I)	0.01	0.40
Fe ₃ O ₄	0.84	193.33	Fe(I)	2.51	139.89
SiO ₂	2.88	172.98	C(I)	5.82	69.87
Al ₂ O ₃	0.74	75.04			
CaO	0.41	22.71	MnO(I)	3.61	256.13
MgO	0.20	8.02	SiO ₂ (I)	2.87	172.12
С	29.85	358.56	$Al_2O_3(I)$	0.74	75.04
CaCO ₃	2.20	220.19	CaO(I)	2.61	146.08
MgCO ₃	1.36	114.67	MgO(I)	1.56	62.84
H₂O	11.00	198.17			
			CO(g)	16.54	463.26
			CO ₂ (g)	11.06	486.59
			H₂O(g)	9.50	171.15
			H ₂ (g)	1.50	3.02
Total	-	2834	Total	-	2834

Nchwaning ore, slag (with entrained metallics) from the HC FeMn process, quartz, coke (Polish and Chinese), dolomite, and silicon sculls. Chemical compositions are presented in Table III. 390 kg remelt material is circulating in the process. Oxygen level in Mn sources, given by x in MnO_x , is 1.2 and the total amount of consumed carbon is 274 kg.

The alloy is carbon saturated, corresponding to 1.5%. It was assumed that the end slag contains 8% MnO and 43% SiO₂. The charge mixture corresponds to the production of approximately 1 t of alloy, excluding the metal entrained in slag (approximately 37 kg in this case). Compositions of input and output streams are presented in Table IV.

The energy analysis of SiMn is presented as a Sankey diagram showing the energy streams in Figure 2, both in specific energy content in kWh and as percentage of the total. The enthalpy balance gives a total energy deficiency and required electrical energy feed of 3419 kWh. The power consumption for standard SiMn is reported to typically be 3500–4500 kWh, which agrees well with the system evaluated here(Olsen, Tangstad, and Lindstad, 2007).

The Sankey diagram shows that approximately 21% of the total input energy has been converted into thermal energy, whereas the remainder is found as chemical energy. As for HC FeMn, the key recovery potential lies within the chemical energy



Figure 1—Sankey diagram showing energy (enthalpy) streams for production of approx. 1 t of HC FeMn alloy. All percentages are given respective to the total energy input/output. Solidification enthalpy is included in thermal streams

Table III

Chemical composition of raw materials used as input in material balance for SiMn (Nchw. = Nchwaning, Dolo. = Dolomite)

	Nchwaning. ore	HC-slag	HC-slag met.	Quartz	Coke		Dolomite	Si-met sculls	Remelt
		-	-		Polish	Chinese			
MnO ₂	34.71								
MnO	37.06	39.85			0.08	0.03	0.13		5.93
Fe ₂ O ₃	14.48			1.00					
Fe			15.00		0.56	0.35	0.05	2.86	3.30
SiO ₂	4.11	22.50		97.00	3.42	5.70		25.60	26.43
Al ₂ O ₃	0.20	11.40		1.00	2.08	3.79		4.92	8.97
Mn			78.00				2.55		28.69
Si							56.72	7.81	
CaO	5.78	13.20		0.50	0.41	0.35	28.97	4.92	12.08
MgO	0.50	5.95		0.50	0.20	0.06	16.89		5.13
H₂O	0.40	1.85			16.80	15.50	8.43	1.53	
K₂O	0.01	1.69			0.18	0.08	0.02		1.04
Р	0.04				0.05	0.02			0.02
CO ₂	1.47						42.97		
FixC			7.00		74.13	71.99		0.66	0.61



Figure 2—Sankey diagram showing energy (enthalpy) streams for production of approx. 1 t of SiMn. All percentages are given respective to the total energy input/ output. Solidification enthalpy is included in thermal streams. (R.M. = remaining materials)

Table IV

Input and output of staged material balance for production of approx. 1 t of SiMn. All raw materials are entering the furnace at 25°C. Output temperatures are 1600°C for slag and alloy and 400°C for off-gas

	Input		Output			
Species	kmol	kg	Species	kmol	kg	
MnO ₂	1.58	137.36	Mn(l)	13.23	726.70	
MnO	9.77	692.70	Si(I)	7.56	212.33	
Mn	3.17	173.88	Fe(l)	1.47	82.20	
Fe ₂ O ₃	0.39	61.48	C(I)	1.30	15.55	
Fe	0.70	39.20				
SiO ₂	12.89	774.43	MnO(I)	1.28	90.97	
Si	2.82	79.06	SiO ₂ (I)	8.14	489.32	
Al_2O_3	1.77	180.37	$Al_2O_3(I)$	1.77	180.37	
CaO	3.82	213.94	CaO(I)	4.33	242.82	
MgO	2.14	86.05	MgO(I)	2.55	102.86	
С	22.81	273.94	K2O(I)	0.25	23.74	
K₂O	0.25	23.74				
CaCO ₃	0.52	51.55	CO(g)	19.74	552.87	
$MgCO_3$	0.42	35.16	CO ₂ (g)	2.71	119.12	
H ₂ O	5.33	96.02	$H_2O(g)$	4.33	78.00	
		H ₂ (g)	1.00	2.02		
Remelt		390.54	Remelt		390.54	
Total	-	3309.32	Total		3309.41	

in the off-gas containing 1631 kWh (20%) of the total energy output. 1299 kWh (16.1%) of the total energy is found as heat in alloy and slag (1533 kWh (19%) including the remelt).

Exergy analysis

The exergy analyses are representative of the HC FeMn and SiMn systems that were described previously for the energy analyses. A general expression for an exergy balance is defined as follows (Kotas, 2013):

$$\sum_{j} \left(1 - \frac{T_0}{T_j} \right) Q + W + \sum_{i} m_i e_i - \sum_{o} m_o e_0 - E_D = 0 \quad [9]$$

where the first term is the potential work utilized from a heat stream, W is added work (electrical energy) and e denotes the exergy with subscript i for input and o for output. E_D is the destrained exergy. The total exergy of a material stream consists of two different contributions similar to the enthalpy: physical (thermal) exergy and chemical exergy:

$$e = e^{ph} + e^{ch} ag{10}$$

$$e^{ph} = (h - h_0) - T_0(s - s_0)$$
[11]

$$e^{-ch} = \Delta G_f^0 + \sum_y n_y e_y^{-ch}$$
[12]

Enthalpies, entropies, and Gibbs formation energies needed in the calculations are found in HSC Chemistry 9 (Outotec,

2017). $e_{y^{ch}}$ is standard chemical exergy respective to a reference environment of $T_0 = 25$ °C, $p_0 = 1$ atm and standard concentration of reference substances in the natural environment. These exergies were given by (Szargut *et al.* 2005) and can also be found in HSC Chemistry 9 (Outotec, 2017).

HC FeMn

The exergy analysis for HC FeMn is presented as a Sankey diagram in Figure 3. Numbers are valid for production of approx. 1 tonne of alloy. The main difference between the exergy balance and the enthalpy balance is that input and output are always equal in terms of enthalpy, whereas the output exergy is always lower than the input. It can be seen from the diagram that 13.7% of the total input exergy is lost during the process due to irreversibilities (energy dissipation), mainly due to chemical reaction. 46.7% of the total exergy was transferred to the product (alloy) as chemical exergy. The remaining exergy has been converted into thermal exergy in various forms and chemical exergy in slag and off-gas, all of which may potentially be recovered. If none of these sources were to be recovered at the plant, the exergy destruction would be 46.2%. As previously mentioned, HC FeMn slag is often used as a raw material in SiMn production, and the chemical energy in the slag may hence be considered recovered. In addition, the chemical exergy in the offgas is often utilized. Considering this, the exergy lost is the sum of the destruction and thermal sources, equal to 26.1%.

SiMn

The resulting exergy balance for SiMn is presented as a Sankey diagram in Figure 4, where exergy streams are given in both kWh and percentage of the total. Numbers are valid for production of approximately 1 t of alloy. If all exergy in the output material

streams was recovered, the amount of destructed exergy would be 10.8%. By considering all exergy streams that seldom are recovered (all thermal exergy and chemical exergy in slag), the exergy destruction is 28.2%. Other observations are that only 43 kWh (0.5%) may be recovered from the low-temperature thermal exergy in the off-gas. In contrast, the high-temperature thermal exergy of slag, alloy, and remelt is 1048 kWh(12.5%).

Discussion

Potential recovery of energy lies within all energy reserves that are not chemical energy in the produced alloy. The chemical potential in the slag is relatively low in both processes; however, the slag in HC FeMn can be used as a raw material in SiMn production and may hence be a useful by-product. The most straightforward potential recovery source is the chemical energy in the off-gas. The gas may be burned to produce thermal heat or electricity; however, burning produces large amounts of CO_2 . Plants located in industry parks may also sell the gas to neighbouring industries(Lindstad, Monsen, and Olsen, 2010). The required technologies are well established and utilized at plants today. The available chemical energy is:

- ► 1401 kWh (24.4% of total output) for HC FeMn
- ► 1631 kWh (20.1% of total output) for SiMn.

These values are valid for an off-gas temperature of 200°C for HC FeMn and 400°C for SiMn, and a prereduction degree of 25%. Both the temperature and the content of CO will vary from day to day in industrial operation and the potential recoverable energy will hence vary correspondingly.

The main source of recoverable thermal energy is cooling of high-temperature alloy and slag. The heat is produced batchwise, which makes recovery challenging. However, a Norwegian plant, Eramet Kvinesdal, has implemented recovery through hot water

Figure 3—Sankey diagram showing exergy streams for production of 1 t of HC FeMn. All percentages are given respective to the total exergy input. Solidification enthalpy is included in thermal (physical) streams

Figure 4—Sankey diagram showing exergy streams for production of 1 t of SiMn. All percentages are given respective to the total exergy input. Solidification enthalpy is included in thermal streams. (R.M. = remaining raw materials)

from the molten slag (Eramet-Norway, 2017). The available energy in alloy and slag during cooling and solidification is:

- 1005 kWh (equal to 17.5% of total energy output) for HC FeMn. The amount that potentially may be recovered is given by the exergy, which is 726 kWh
- 1299 kWh (equal to 16.1% of total energy output) for SiMn, not including thermal energy in remelt. Corresponding exergy is 917 kWh. Values including remelt are 1533 kWh and 1048 kWh for energy and exergy, respectively.

The exergy analyses showed the reduction in energy quality accompanying the transformations occurring in the furnace. These values were found to be 13.7% for HC FeMn and 10.8% for SiMn. A comparison of the calculated enthalpies and exergies can be seen in Figure 5 for HC FeMn(left) and SiMn(right).

When calculating an enthalpy balance, it is assumed that species in the highest oxidation state, *e.g.* carbon in form of CO_2 , has zero enthalpy at $T = 25^{\circ}C$ and p = 1 atm regardless of the species' concentration. In exergy calculations, the species will have a certain amount of chemical exergy if the concentration of the compound deviates from that in the natural environment. Due to this, chemical exergy of material streams may be larger than the corresponding enthalpy. This is, for example, seen for the slag in both systems. Nonetheless, the chemical enthalpy and exergy in off-gas is very similar. Additional information provided by the diagram is the difference in the available energy from heat streams (enthalpy) compared to the amount potentially recoverable (exergy). It can be seen that energy from low-temperature heat sources (such as the off-gas) may less recoverable compared to high-temperature sources.

Conclusions

Staged energy and exergy balances were performed to evaluate the potential for energ recovery during production of HC FeMn and SiMn. Parameters related to furnace operation (energy loss from electrodes, heat losses from furnace shell *etc.*) are a major contributor to the overall energy consumption of the process, potentially resulting in an efficiency of 80% (Tangstad, 1995). However, the focus of this paper was on the energy within the material streams rather than the efficiency of a specific plant, and such considerations were thus not included. With no losses considered, the electric energy consumption was found to be approximately. 2460 kWh/t for HC FeMn and 3420 kWh/t for SiMn, agreeing well with the existing literature. The chemical energy in the off-gas is the largest potential source of recoverable energy, which is both well-known and utilized in the majority of plants today. Other major sources are the thermal energy of the alloy and slag. This energy is more difficult to recover. However, technology has been implemented to produce hot water from molten slag beds at a Norwegian plant.

The distribution of energy and exergy between the different material streams are very similar and key potential recovery sites will be the same regardless of the analysis method utilized. The additional information provided by the exergy analysis compared to the enthalpy is the amount of exergy lost (reduced energy quality) due to irreversible processes (*e.g.* chemical reactions) within the furnace. These values were found to be 13.7% for HC FeMn and 10.8% for SiMn.

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