

# Thermodynamic modeling of the reaction of simultaneous interaction of nickel (II), cobalt (II) and iron (III) oxides with carbon

*Olga Kolesnikova*<sup>1,\*</sup>, *Farida Nurbaeva*<sup>2</sup>, *Guizhan Sabyrbayeva*<sup>3</sup> and *Lazzat Nurshakhanova*<sup>4</sup>

<sup>1</sup> Master of Engineering and Technology, Junior Researcher, Department of Science, Production and Innovation, M. Auezov South Kazakhstan University, 160012 Shymkent, Kazakhstan

<sup>2</sup> Candidate of Technical Sciences, Associate Professor, Department of Ecology and Geology, Sh. Yessenov Caspian University of Technology and Engineering, 130002, Aktau, Kazakhstan

<sup>3</sup> Candidate of Technical Sciences, Associate Professor, Department of Ecology and Geology, Sh. Yessenov Caspian University of Technology and Engineering, 130002, Aktau, Kazakhstan

<sup>4</sup> Candidate of Technical Sciences, Associate Professor, Department of Petrochemical Engineering, Sh. Yessenov Caspian University of Technology and Engineering, 130002, Aktau, Kazakhstan

**Abstract.** This paper considers thermodynamic modeling of electric smelting of Allarechenskoye deposit dumps for nickel, cobalt, and iron extraction. Thermochemical transformations in the reaction of interaction of carbon with oxides of nickel (II), cobalt (II) and iron (III) are considered. The investigations of thermochemical transformations of the chemical-phase composition of the system were carried out using the software complex "Astra-4" developed by a team of scientists at the Bauman Moscow State Technical University. Thermochemical transformations and chemical-phase composition of the system as a function of T (temperature) and P (pressure) were studied. In particular, the temperature was in the range from 973K to 1773K and the pressure range was taken from 0.001 MPa to 0.1 MPa. According to the results, it was found that iron in the system transitions to condensed iron, with the degree of transition ( $\alpha_{Fe}$ ) from 29.0% at T = 1173K to 99.1% at 1773K, the degree of transitions of nickel to condensed nickel is 100% before T = 1273K, and with an increase in temperature from 1273K to 1773K decreases to 99,98%; the transition of cobalt to the gas phase is distributed similarly to nickel.

## 1 Introduction

Nickel production involves complex processing of oxide-silicate nickel/cobalt ore as well as sulfide copper-nickel ores. Key impurities, such as gold, copper, and platinum group metals, and the demand for them influence the result of nickel processing [1-6].

Nickel is used (as an additive) in the production of corrosion-resistant and special heat-resistant alloys and in the production of stainless steels. These metallurgical products are used in mechanical engineering, instrument engineering, household appliances and electronics.

The U.S. Geological Survey claimed in early 2014 that the world's nickel reserves were about 130 million tons. However, these figures out to be overstated. Over the last decade, proven reserves are estimated at 74 million tons of ore. Australia, Brazil, New Caledonia and Russia are known to have huge explored and proven nickel reserves. Cuba, Indonesia, Canada, China and South Africa also add to that list [1-12].

Oxide-silicate nickel/cobalt deposits form the basis of the global nickel industry. The rest of the deposits, the processing of which extracts nickel, account for 0.1-0.2% of the total metal reserves [1-12].

More than half of the world's proven nickel reserves are in the form of oxide deposits. It is known that only 40% of the metal was extracted from lateritic ores at the beginning of 2014. This is explained by the significant content of nickel and copper in the sulfide ores. Additionally, impurities of rare and noble metals are of special value in ore processing, as this is an additional and substantial profit compared to the target product (nickel).

The sulfide ore deposits are mainly located in the Russian Federation, South Africa, Canada, and China. 50% of Australia's nickel ore deposits produce as well. However, the mineral resource base of Cuba, Indonesia and New Caledonia, and Kazakhstan is mainly represented by oxide and silicate nickel ore deposits [1-5, 7-12].

The world's leading Ni producers use ores that contain at least 1.4% of the metal. However, the use of various methods and technologies related to the processing and enrichment of poor nickel ores is growing in intensity and popularity [1-23].

Today, the Arctic zone of Russia appears to be one of the priority areas of development in economic and environmental terms. Over the years of industrial

\* Corresponding author: [ogkolesnikova@yandex.kz](mailto:ogkolesnikova@yandex.kz)

development of the Arctic zone of Russia, a significant number of industrial enterprises of petrochemical, gas, mining and processing, mining and metallurgical and a number of other industries have been put into operation [13 - 43], which during their operation have been able to accumulate hundreds of millions of tons of various industrial waste, which today contribute to the negative impact on the environment of the Arctic region [55].

At the present stage, when there is a decrease in the content of metals in mined ores, an urgent problem is the involvement of man-made formations in the form of waste, dumps, etc. as secondary mineral raw materials, depending on their chemical composition and content.

One of these dumps is the dumps of the Allarechenskoe deposit (Pechenga District, Murmansk Region), which contains over 12 million tons of waste copper-nickel ore. The deposit was mined using the open-cut method, but it was completed in 1971. The main minerals mined at the deposit in the Murmansk region were nickel, copper, and cobalt. The dumps consist of overburden and mostly ore-free rocks, amphibolites, and granite-gneiss. The granulometric composition is extremely heterogeneous. Moreover, under the influence of precipitation and other natural phenomena, the primary ores in the dump for a long stay were subjected to hypergenic processes, which led to the formation of oxidized ores. As a result, a considerable part of the ore has lost its original quality [13].

Studies show that the most hazardous are sludge and tailings, as well as metallurgical wastes due to their fine-grained structure. However, a number of Russian researchers argue that coarse-grained sulfide-containing wastes from mining enterprises can be no less dangerous. Thus, based on the chemical composition of the Allarechenskoe deposit dumps (the presence of nickel and cobalt oxides) and the theoretical possibility of their use as secondary raw materials for the extraction of useful metals, studies have been conducted. They included the method of thermodynamic modeling of chemical and phase transformations under conditions of simultaneous interaction of nickel (II), cobalt (II) and iron (III) oxides with carbon. The studies were conducted with the help of the complex program "Astra-4"[44]. The purpose of this work is to evaluate the effect of temperature and pressure in the thermodynamic simulation of reductive electric melting [29, 42, 45-50] under conditions of simultaneous interaction of nickel Ni(II), cobalt Co(II) and iron oxides Fe<sub>2</sub>O<sub>3</sub>(III) with carbon and subsequent extraction of Ni, Co and Fe.

## 2 Materials and Methods

The HSC Chemistry software package designed by Outokumpu (Finland) was used to calculate the Gibbs energy. Its essence is the possibility of reduction of metals in the reaction by simultaneous interaction of Ni (II), Co (II), and Fe (III) oxides with C [52]. The software allows the calculation of thermodynamic functions such as entropy ( $\Delta S$ ), enthalpy molar heat capacity ( $\Delta H$ ), Gibbs energy ( $\Delta G$ ) for each substance involved in the reaction, as well as changes of these

thermodynamic functions during the chemical reaction [52-64].

According to the chemical transformation:  $\text{NiO} + \text{CoO} + \text{Fe}_2\text{O}_3 + 5\text{C} = 2\text{Fe} + \text{Ni} + \text{Co} + 5\text{CO}$ , the change in Gibbs energy ( $\Delta G$ ) of the reaction was calculated. The results of the mathematical calculation are shown in Table 1. The obtained Gibbs energy values showed the possibility of the reaction and reverse reduction of metals. Further work on the simulation of the chemical process was continued in the program "Astra-4". Thermodynamic modeling of metal reduction under the conditions of the above main chemical reaction was performed at a pressure of 0.001-0.1 MPa and temperature 973-1773K.

The "Astra-4" algorithm is based on the universal thermodynamic method for determining the equilibrium characteristics of heterogeneous systems. application of this method is based on the fundamental principle of maximum entropy. The "Astra-4" program allows to formulate quite simply the modeling problem statement for the thermodynamic method of research [44, 65 -74].

## 3 Results and Discussion

At a temperature of 873K and a Gibbs energy  $\Delta G$  of -22314 kJ/mol the beginning of the reaction was recorded. A further increase in temperature up to 1773K gives more negative Gibbs energy values, which reach -775345 kJ/mol. These calculations are shown in Table 1.

**Table 1.** Results of the calculation of the Gibbs energy ( $\Delta G$ ) of the reaction  $\text{NiO} + \text{CoO} + \text{Fe}_2\text{O}_3 + 5\text{C} = \text{Ni} + \text{Co} + 2\text{Fe} + 5\text{CO}$ .

T, K	Enthalpy ( $\Delta H$ ), kJ	Entropy ( $\Delta S$ ), kJ/K	Gibbs energy ( $\Delta G$ ), kJ/mol	Constant equilibrium (K) <sup>a</sup>	Logarithm of constant (LogK)
573	738.685	875.730	236.892	2.53E-022	-21.597
673	733.720	867.750	149.725	2.38E-012	-11.622
773	723.210	860.145	63.315	5.26E-005	-4.279
873	721.967	852.556	-22.314	2.16E+001	1.335
973	715.842	845.897	-107.216	5.70E+005	5.756
1073	713.827	843.923	-191.702	2.15E+009	9.333
1173	710.464	840.932	-275.949	1.94E+012	12.289
1273	707.504	838.566	-359.990	5.92E+014	14.773

Subsequent thermodynamic modeling in the software package "Astra-4" shows that the degree of iron ( $\alpha_{\text{Fe}}$ ) conversion in the interaction of NiO, CoO and Fe<sub>2</sub>O<sub>3</sub> with carbon varies with pressure (at P = 0.001 and 0.01 MPa). However, it is independent of temperature and ranges from 99.9% to 100% condensed magnetite  $k \cdot \text{Fe}_3\text{O}_4$ , where k - condensed phase (Tables 2, 3).

**Table 2.** Transition degree results for Ni, Co, Fe, C, O<sub>2</sub> at 973-1773 K and 0.001MPa pressure

Elem.	Temperature, K				
	1773	1573	1373	1173	973
<b>Degree of nickel distribution (<math>\alpha_{Ni}</math>), %</b>					
k•Ni	8.780929	8.834213	8.88642	8.9247534	8.948606
k•NiO	91.2142	91.16565	91.1135	91.075246	91.05139
NiO	9.67E-05	1.09E-06	0	0	0
Ni	0.004775	0.000135	1.28E-06	0	0
Total	100	100	100	100	100
<b>Degree of cobalt distribution (<math>\alpha_{Co}</math>), %</b>					
k•CoO	99.41675	99.99761	100	100	100
Co	0.082338	0.002395	0	0	0
CoO	0.500909	0	0	0	0
Total	100	100	100	100	100
<b>Degree of iron distribution (<math>\alpha_{Fe}</math>), %</b>					
Fe	0.003696	8.08E-05	0	0	0
FeO	0.000581	0	0	0	0
k•Fe <sub>3</sub> O <sub>4</sub>	99.99572	99.99992	100	100	100
Total	100	100	100	100	100
<b>Degree of carbon distribution (<math>\alpha_C</math>), %</b>					
CO	4.336	2.990	1.767	0.867	0.317
CO <sub>2</sub>	95.664	97.010	98.233	99.133	99.633
Total	100	100	100	100	100
<b>Degree of oxygen distribution (<math>\alpha_{O_2}</math>), %</b>					
O	0.000144	2.11E-06	0	0	0
O <sub>2</sub>	0.000182	0.000182	0	0	0
CO	0.194118	0.13379	0.079038	0.0387774	0.014161
CO <sub>2</sub>	8.566385	8.682608	8.787827	8.8647597	8.912373
CoO	0.004508	0	0	0	0
FeO	1.04E-05	0	0	0	0
k•Fe <sub>3</sub> O <sub>4</sub>	2.387805	2.386699	2.385583	2.3846229	2.384195
k•CoO	0.894704	0.894992	0.894564	0.8942036	0.894043
k•NiO	87.95205	87.90173	87.85299	87.817636	87.79523
Total	100	100	100	100	100

**Table 3.** Transition degree results for Ni, Co, Fe, C, O<sub>2</sub> at 973-1773 K and a pressure of 0.01 MPa

Elem.	Temperature, K				
	1773	1573	1373	1173	973
<b>Degree of nickel distribution (<math>\alpha_{Ni}</math>), %</b>					
k•Ni	8.776904	8.834228	8.88642	8.9247534	8.948606
k•NiO	91.22261	91.16576	91.11358	91.07524	91.05139
NiO	9.65E-06	0	0	0	0
Ni	0.000476	1.35E-05	0	0	0
Total	100	100	100	100	100
<b>Degree of cobalt distribution (<math>\alpha_{Co}</math>), %</b>					
k•CoO	99.99124	99.99976	100	100	100
Co	0.008257	0.000239	0	0	0
CoO	0.000502	0	0	0	0
Total	100	100	100	100	100
<b>Degree of iron distribution (<math>\alpha_{Fe}</math>), %</b>					
Fe	0.000369	0	0	0	0
FeO	5.8E-05	0	0	0	0
k•Fe <sub>3</sub> O <sub>4</sub>	99.99957	100	100	100	100
Total	100	100	100	100	100
<b>Degree of carbon distribution (<math>\alpha_C</math>), %</b>					
CO	4.335591	2.989643	1.767016	0.8672793	0.316769
CO <sub>2</sub>	95.66441	97.01035	98.23293	99.13272	99.63323
Total	100	100	100	100	100
<b>Degree of oxygen distribution (<math>\alpha_{O_2}</math>), %</b>					
O	1.44E-05	0	0	0	0
O <sub>2</sub>	0.000997	1.82E-05	0	0	0
CO	0.194124	0.13379	0.079038	0.0337774	0.014161
CO <sub>2</sub>	8.566657	8.682621	8.787851	8.8647597	8.912373
CoO	4.5E-06	0	0	0	0
FeO	1.04E-06	0	0	0	0
k•Fe <sub>3</sub> O <sub>4</sub>	2.38796	2.386729	2.385582	2.3846229	2.384195
k•CoO	0.895399	0.894993	0.894563	0.8942036	0.894043
k•NiO	87.95434	87.90185	87.85297	87.817636	87.79523
Total	100	100	100	100	100

When the pressure rises to P=0.1 MPa, iron goes into the k • Fe phase. The chemical compound k • Fe<sub>3</sub>C at temperature 973K contains 100% Fe. When the temperature is further increased to 1773K the iron content decreases and reaches 0.94% (Table 4).

**Table 4.** Transition degree results for Ni, Co, Fe, C, O<sub>2</sub> at 973-1773 K and a pressure of 0.1 MPa.

Elem.	Temperature, K				
	1773	1573	1373	1173	973
<b>Degree of nickel distribution (<math>\alpha_{Ni}</math>), %</b>					
k•Ni	99.99488	99.99985	100	100	100
Ni	0.005123	0.000146	1.36E-06	0	0
Total	100	100	100	100	100
<b>Degree of cobalt distribution (<math>\alpha_{Co}</math>), %</b>					
k•Co	99.99555	99.99988	100	100	100
Co	0.004454	0.000123	1.25E-06	0	0
Total	100	100	100	100	100
<b>Degree of iron distribution (<math>\alpha_{Fe}</math>), %</b>					
Fe	0.005118	0.00017	2E-06	0	0
k•Fe <sub>3</sub> C	0.942797	2.21578	9.50843	71.02594	100
k•Fe	99.05208	97.78405	90.49152	28.97406	0
Total	100	100	100	100	100
<b>Degree of carbon distribution (<math>\alpha_C</math>), %</b>					
CO	99.74867	99.40921	97.4645	81.05911	44.70088
CO <sub>2</sub>	0.12562	0.295346	1.267711	9.470441	27.64956
k•Fe <sub>3</sub> C	0.125709	0.295442	1.267785	9.470441	13.33314
k•C	0	0	0	0	14.31643
Total	100	100	100	100	100
<b>Degree of oxygen distribution (<math>\alpha_{O_2}</math>), %</b>					
CO	99.74376	99.40931	97.46453	31.05911	44.70033
CO <sub>2</sub>	0.2251241	0.590695	2.535424	13.94039	55.29912
Total	100	100	100	100	100

The degree of carbon transformation ( $\alpha_C$ ) in the temperature interval from 973K to 1373K and pressure equal to 0.1 MPa is presented in the system by the condensed phase with the content of condensed carbon (k • C). a change from 14.31% to 0% at temperatures of 973K and 1173K is observed, respectively, and k • Fe<sub>3</sub>C with a maximum transition rate of up to 13.33% at 973K. The carbon ( $\alpha_C$ ) gas phase is represented as CO compounds ranging from 44.7% to 99.74% at 973K and 1773K, respectively, and CO<sub>2</sub> with a carbon transition rate up to 27.64% at 973K with a decrease to 0.125% at 1773K. (Table 4).

Reducing the pressure to 0.001-0.01 MPa allows us to see how the transition process of carbon ( $\alpha_C$ ) occurs. It should be noted that carbon is represented by mono- and dioxide, which are distributed in the following ratio: CO<sub>2</sub> from 95.66 to 99.683% and CO from 4.336 to 0.317% at P = 0.001 MPa. Increasing the pressure to P = 0.01 MPa gives a CO<sub>2</sub> distribution in the range of 95.664-99.68% and CO: 4.335 -0.316%.

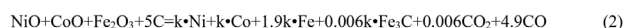
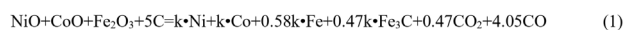
The distribution of the degree of cobalt transition ( $\alpha_{Co}$ ) under the conditions of interaction of nickel (II), cobalt (II) and iron (III) oxides with carbon is shown in Figures 2-4.

According to figure 2.3,  $\alpha_{Co}$  presented by k • CoO in the pressure range of 0.001-0.01 MPa is 99.9%. Increasing the pressure to P=0.1 MPa shows the degree of conversion of cobalt to k • Co and is 100% in the temperature range 973-1373K. A further increase in temperature to 1773K shows that  $\alpha_{Co}$  decreases to

99.99% as the transition to the gas phase begins, similar to the behavior of Ni (Table 4).

Oxygen in the system at a pressure equal to 0.1 MPa goes to CO<sup>2</sup> up to 55.29% at T=973K, and further decreases to 0.25% at T=1773 K. In addition, oxygen is distributed to CO, reaching 44.7 - 99.74% at T=973-773 K (Table 4).

Thermodynamic studies performed in the NiO-CoO-Fe<sub>2</sub>O<sub>3</sub>-C system allowed us to make chemical equations at temperatures of 1173K:



## 4 Conclusions

Thus, studies conducted by thermodynamic modeling of the reaction of simultaneous interaction of nickel (II), cobalt (II), and iron (III) oxides with carbon in the temperature range 973-1773 K and pressure 0.001-0.1 MPa give the following results and conclusions:

- technogenic dumps of overburden and substandard oxidized ores of the Allarechenskoye deposit contain such valuable components as nickel and cobalt at the level of mined ores, and can be used as secondary raw materials for non-ferrous metal extraction and reduction of the anthropogenic load on the environment of the Arctic region.

- joint reduction of iron, nickel and cobalt in the temperature range 1373-1773K is possible. At these temperatures and pressures equal to 0.1 MPa the following chemical transitions are demonstrated: Fe from 90.49% to 99.05%, Ni from 99.994% to 100%, Co from 99.995% to 100.

- the thermodynamic calculations performed for the joint reduction of metals allow obtaining a ferroalloy in the form of ferronickel.

- with the organization of production for the processing of man-made waste from the Allarechensk dump with the production of ferronickel using the above technology, the socio-environmental and economic climate in the Arctic region will be improved, which will further contribute to the development of the Arctic.

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