# REDUCIBILITY AND ELECTRIC RESISTANCE OF CARBONACEOUS MATERIALS WITHIN FERROALLOY SMELTING PROCESSES

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## ABSTRACT

*Reactivity and electric resistance are the main Q-factors of reducing agents used for ferroalloy production in SAF furnace. These factors depend on the coal nature and its processing conditions. Reactivity and electric resistance of a reductant have an impact over furnace power and production capacity, furnace charge and energy consumption as well as on the yield of the target component.*

*The article describes comparative electric resistance between coke and special coke produced through high-speed pyrolysis process from the coal of Shubarkol coal deposit (Republic of Kazakhstan). Laboratory tests have been performed under temperatures up to 1600 °C. An impact of a reducing agent on electric resistance of a furnace charge mixes used for FeSi and HC FeCr smelting has been defined.*

*The article also contains reactivity data of two types of carbonaceous reducing agents. Kinetic research has been carried out with regard to HC FeCr and Si (FeSi) smelting conditions. Results showed kinetic behavior of Si and chromite reduction by different types of carbonaceous materials.*

***KEYWORDS:*** *Silicon, chromium, reduction, kinetics, electrical resistance, reducing agent, coke, special coke, temperature, SAF furnace.*

## 1. INTRODUCTION

### 1.1. Reactivity of reductants

The type of carbonaceous reductant affects the rate of reduction and thus the overall process. Slag rich and slagfree processes of ferroalloy smelting have different reduction mechanisms. HC FeCr smelting (slag rich process), is performed by solid phase chromite reduction by carbon. Reduction mechanism is more complicated during Si alloy smelting with slagfree technology. In this case the following is observed: solid-phase reactions, reduction with gaseous-phase reactions, interaction of products resulting in chemical reactions etc.

Chromite reduction. According to reports [1-2] $Cr_2O_3$ reduction by carbon takes place at diffusion control mode. Initially reduction of $Cr_2O_3$ occurs locally at the carbon-oxide boundary layer. As a result of this interaction, the layer with $Cr_2O_3$ as well as $Cr_3C_2$ and $Cr_7C_3$ is formed on the oxide surface. Further, the rate of $Cr_2O_3$ reduction is limited by carbon diffusion through carbides and chromium [1]. The chromite reduction is accompanied by metal-slag layer formation at the chromite surface which forms the diffusion barrier for evolution of reduction over the overall depth of chromite [2]. The type of carbonaceous material effect on the Cr reduction rate has been considered in a number of papers and it is still a controversial issue. The authors [3] studied the kinetic of briquettes (chromite + coal) reductions, using coke and the Angarsk`s char as a reductant.

The investigation concluded that despite its high reactivity by CRI the char had no rate benefits compared to coke. According to the authors, reductant wetting by slag is a key factor in the reduction which dominates the reduction rate. V.S. Volkov evaluated reactivity of various materials sized to 0.2-0.4 mm [1]. As reducing agent graphite, coke, char, charcoal and coal were used. The investigation resulted in that type of reductant does not affect the reduction rate. According to [4] chrome oxide reduction rate by coke 5-10 mm large at 1350-1500 °C rises as the CRI increases.

Silicon reduction. At the moment there is no common theoretical model describing processes in silicon smelting furnace and the matter is still under discussion. However, it is common that one of the process main stages is the SiO generation, and the SiO gas and carbon interaction to SiC formation on reductant surface. According to the theories silicon reduction is performed through some reactions sequence: Initial solid state materials interact with gaseous products formation (CO, SiO). CO and some of SiO escape from the furnace, but the main part of SiO reacts with coke (char) and a SiC layer is formed on it. In the beginning, the rate control of $SiO + 2C = SiC + CO$ (1) reaction is dominated by chemical reaction rate. The more coke surface is covered by the SiC layer, the more a diffusion effects on process rate, and eventually the reaction passes into the mode which rate control is dominated by diffusion. The carbide blocks oxidizing gas penetration into coke i.e. diffusion barrier is generated and its thickness depends on the surface of coke which has access for gas penetration. Moreover SiC reacts with SiO to Si and if silica is present, carbide would react with silica and Si, SiO and CO would be generated. Under such conditions, the reductant properties such as specific area, porosity and pore-size distribution affect the reduction process. Low coke reactivity towards to SiO affects the Si losses. Since its effect on Si yield, trapping SiO is important matter which depends on reductant performance. It may be noted that chemical activity of reductant also effects the process [5].

### 1.2. Electrical resistance of reducing agents and charges

One of the most important properties of a reducing agent used in ferroalloy smelting is electrical resistance. Granular solids bed resistance is summarized by the electrical material resistance and surface resistance of pieces. For successful reduction process evolution it is necessary to provide particular conditions with thermal energy generation at electrodes tips and minimum energy generation at top charge layer. This operating mode is provided by deep electrodes penetration into burden. In practice, it is achieved either by secondary voltage decrease or a high resistance materials usage. As power and efficiency of a furnace decrease with low voltage, the first technique is ineffective. High burden resistivity allows using more high voltage without disturbances of process. To increase burden resistivity materials with low conductivity are used such as a woodchip, wood scrap and other wood products, however, in many countries they are in shortage. Char and coal usage as a reductant is the most popular method of increasing burden resistivity. Therefore it is important to investigate resistance of charge with a new reductant before it would be selected for practice.

## 2. EXPERIMENTS AND RESULTS

### 2.1. Reactivity tests

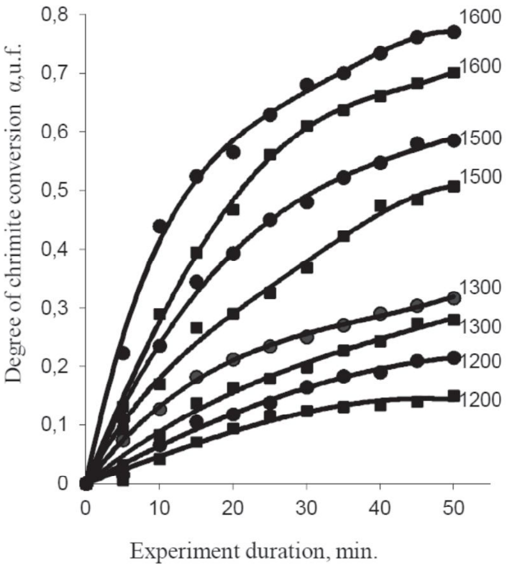
Special coke reactivity tests have been carried out with the aim to predict a new reductant performance in ferroalloys smelting processes [6-8]. Special coke is produced from noncoking coal charred under flash heat conditions and oxidizing environment [9-10]. Proximate analysis of reductants used is shown in table 1.

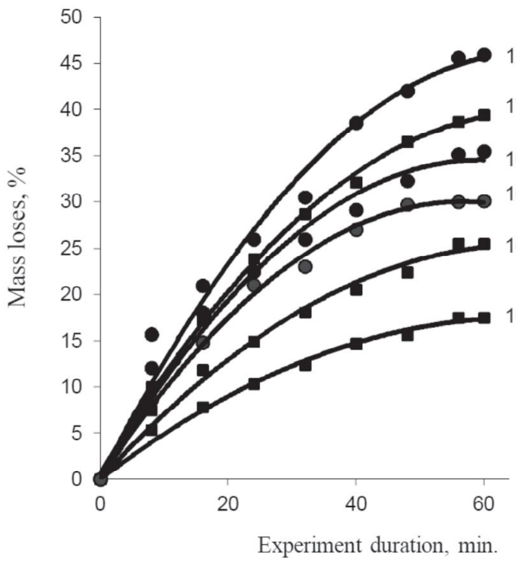
Chromite reduction with coke and special coke has been investigated by thermal gravimetric method at 1200°, 1300°, 1500° and 1600°C. Chromite ore (50% $Cr_2O_3$) and reductants were sized to 0.315-1.000 and 3-6 mm respectively and mixed in stoichiometric ratio. Fe and Cr conversion (α) was calculated by mass loss on the assumption that it was due to CO evolution.

**Table 1:** Proximate analysis of reductants

| **REDUCTANT** | **Proximate analysis, %** | | | | |
|---|---|---|---|---|---|
| | **moisture** | **ash** | **volatile** | **sulfur** | **phosphorus** |
| Russian coke | 1.8 | 13.5 | 1.5 | 0.50 | 0.045 |
| China coke | 3.5 | 14 | 2.7 | 0.27 | 0.015 |
| Char | 4.1 | 10 | 20 | 0.51 | 0.040 |
| Special coke | 3.2 | 7.1 | 7.5 | 0.27 | 0.020 |

Figure 1 shows that in comparison with coke, chromites conversion and chromite reduction rate by special coke are higher at all temperatures. Temperature increases rate and degree of reduction. It can be seen that curves are exponential i.e. the more conversion, the less reduction rate.





Numbers under the curves are process temperatures °C;
■– mixture with coke; ●–special coke mixture

**Figure 1:** Reduction of Chromite ore

**Figure 2:** Mass loses of FeSi75 mixtures

For experimental data interpretation a number of kinetic models, which describes diffusion controlled reactions, has been involved. If experimental data are fitted in Gistling &Brownstain equation plot points are nearest to straight line. According to this model rate constant is expressed by equation [11]:

$$1-(2/3\cdot\alpha)-(1-\alpha)^{2/3}=k\tau,$$

where: α – conversion degree,

k – reaction rate constant

τ – time, min.

Through this model reaction rate constants (k) has been calculated (table 2). Reaction rate constant increases by 1-2% per 1° and this fact confirms that diffusion plays main role in the process. Diffusion barrier is a complex mix, which comprises chromite reduction reaction products, slag and unreacted chromite ore. The mix is a solid spherical shape matter which covers ore particles and affects reduction rate. Under these conditions conversion rate is limited by carbon diffusion through the barrier to reaction surface.

**Table 2:** Values of k at different temperatures for each reductant

| **t, °C** | **Reductant used** | **k** |
|---|---|---|
| 1200 | Special coke | $1{,}252\cdot10^{-4}$ |
| | Coke | $5{,}847\cdot10^{-5}$ |
| 1300 | Special coke | $2{,}637\cdot10^{-4}$ |
| | Coke | $2{,}124\cdot10^{-4}$ |
| 1500 | Special coke | $1{,}183\cdot10^{-3}$ |
| | Coke | $8{,}140\cdot10^{-4}$ |
| 1600 | Special coke | $2{,}344\cdot10^{-3}$ |
| | Coke | $1{,}898\cdot10^{-3}$ |

Compared to coke chromite reduction with special coke has highest k and lowest E (191.3 kJ/mole and 168.8 kJ/mole respectively). Special coke advantage is attributable to its high surface area which improves ore and reductant contacts at the initial stage. Since diffusion barrier is formed at ore surface and reaction rate is limited by carbon diffusion through carbide and chromium, type of reductant has insignificant impact on the process.

Silicon reduction. Type of reducing agent effect on FeSi75 mixture reduction has been investigated by continuous weighing method at 1600°, 1650° and 1700°C. The reductants under investigation were coke and special coke. All materials were ground and sized to 0.315-1.000 mm. Reductants were grounded for equality of their surface areas and carbonaceous material chemical activity estimation. According to curves behavior from figure 2 mass losses of FeSi 75 mixtures with special coke are higher at all temperatures. The higher temperatures, the more mass losses and the less the differences between reductants; nevertheless special coke maintains the lead in reactivity. This proves high chemical activity of special coke towards $SiO_2$ reduction.

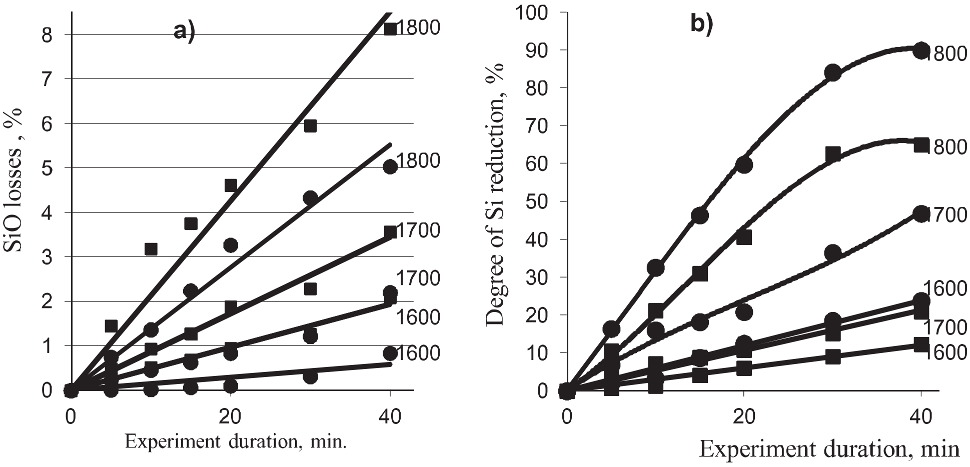
The next experiments stage was to investigate the influence of reductant type on $SiO_2$ reduction. Quartz (99.5% purity) was sized to 0.5-1.6 mm and reductants to 3-6 mm. Reducing agent and quartz ratio was stoichiometric according to $SiO_2+2C=Si+2CO$ reaction.

Four graphite sample holders (two of them with reaction mixture and the other two only reductants) are simultaneously placed into preheated furnace. After isothermal processing at 1600-1800°C samples are recovered from furnace, weighed and chemically analyzed, and mass balance is estimated. Degree of Si reduction is identified as ratio of silicon mass in Si and SiC forms divided by initial silicon quantity.

Compared to coke, Si and SiC formation ability of special coke is higher, this means that special coke has high reactivity towards silica. Special coke has a high surface area, which allows favorable conditions for SiO to penetrate deep into reductant piece. SiC formation occurs whichdecreases the Si losses (figure 3). The higher temperature, the higher degree of Si reduction and the more SiO losses. Increase of SiO losses with treatment time correlates with available carbon content decrease due to SiC layer formation on reductant surface.

For experimental data interpretation a number of kinetic models, which describes diffusion controlled reactions, has been involved. If experimental data are fitted in Jander equation plot, points are nearest to straight line. According to this model diffusion layer thickness is proportional to covered matter conversion degree and it is expressed by equation:

$$(1-(1-\alpha)^{1/3})^2=k\tau$$



Numbers under the curves are process temperatures °C;
■– mixture with coke; ●– mixture with special coke

**Figure 3:** SiO losses (a) and degree of Si reduction (b)

Under these conditions diffusion barrier is silicon carbide. Since diffusion coefficient of Si is an order higher than carbon diffusion, layer of reaction product penetrates into reductant depth, therefore reducing agent is a covered matter. Degree of conversion is expressed through ratio of consumed carbon mass to initial C quantity. Table 3 shows that reaction rate constant increases by 3-5% per 1° and this fact indicates that reduction proceeds at combined mode. At this mode both the diffusion rate and the reaction rate effect on the reduction process. However, this investigation has considered total reducing ability of reductant, which integrates different factors including SiC formation and consumption reactions. Silicon carbide covers carboneus material and creates a diffusion barrier for process evolution. Reaction carries on by carbon diffusion through SiC layer after all reductant surface would have been covered by SiC. Due to free carbon absence SiO reacts with SiC and Si is formed at carbide surface. Concentration gradient improves conditions for Si diffusing through carbide layer to SiC-C boundary and for silicon and carbon interaction with additional carbide formation.

**Table 3:** Values of silica reduction rate constant at different temperatures for each reductant

| **t, °C** | **Reductant used** | **k** |
|---|---|---|
| 1873 | Special coke | $2{,}50\cdot10^{-4}$ |
| | Coke | $8{,}09\cdot10^{-5}$ |
| 1973 | Special coke | $9{,}59\cdot10^{-4}$ |
| | Coke | $4{,}51\cdot10^{-4}$ |
| 2073 | Special coke | $5{,}78\cdot10^{-3}$ |
| | Coke | $2{,}55\cdot10^{-3}$ |

SiC consumption decreases diffusion barrier and improves conditions for further carbon consumption. At 1700-1800 °C quartz is melted with enlarging of $SiC-SiO_2$ interaction surface and consequently increasing carbide consumption rate. In summary, from our point of view, reacting substances diffusion, that is to say SiC thickness, is of great importance for total Si reduction rate.

All other things constant, the activation energies of quartz reduction by special coke and coke are 505.6 and 556.2 kJ/mole respectively. Since reaction rate is inversely proportional to diffusion barrier (SiC) thickness, it can be suggested that compared to coke special coke has lower thickness of SiC layer per carbon unit. High specific surface area of special coke provides this advantage.

## 2.2. Electrical resistance

Specific electrical resistance of various reductants used in practice in comparison with the new one has been measured at a high-temperature laboratory facility [12, 13]. Experiments have been carried out under the following conditions: upper electrode was pressed under 0.4 kg per square cm, heating rate was 400-500°C per hour, material layer depth was 40 mm, reductants were sized to 3-6 mm, internal diameter of sample holder was 50 mm, and test indication was recorded every 50°C. The reductants under investigation were Russian coke, China coke, Russian char and special coke.

As it can be seen from figure 4, reductants being tested at low temperatures can be divided as follows: cokes have lowest resistivity, special coke has resistivity which is in 13-18 times higher than previous two cokes and char resistivity is highest. Temperature rise reduces all reductans resistances. It should be noted that each reductant has unique behavior of resistivity reduction. Within the range of 700-900°C reductants have approximately equal resistivity, and at temperatures above 1000°C reductants are sequenced in resistivity decreasing order, as follows: China coke, special coke, Russian coke and char with resistivity levels at 1600 °C of 3.62; 2.69; 2.04 and 1.66 Ohm-cm respectively.

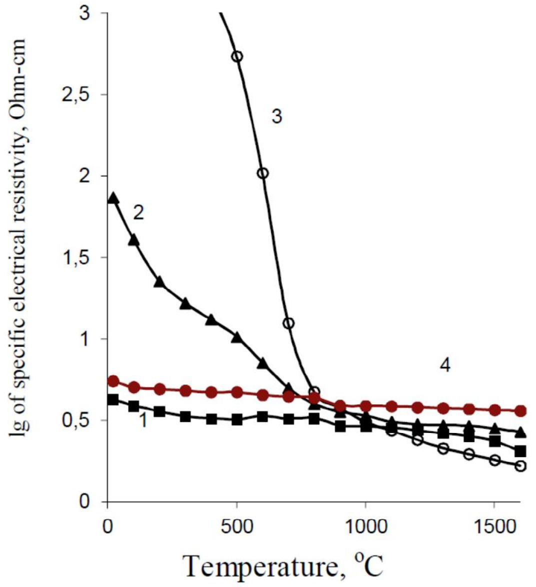
The investigation has proved sufficient electrical properties of special coke: its resistivity is only lower than for char at low temperatures and lower than china coke at temperatures above 1000°C.

Electrical resistivity of ferrosilicon mixtures. The investigation has been carried out under the same conditions. To prevent consumption each electrode had been protected by a molybdenum tip. Pre-dried materials under investigation were: quartzite(98% $SiO_2$), fractions 3-15 mm, iron turnings, special coke, Russia coke, China coke, also special coke mixed with coke where coke content is 75, 50 and 25% (weight). Reductant is sized to 2-5 mm. Mixture content is given in table 4. Figure 5 exhibits relationship between FeSi 75 mixture's resistivity and temperature.

**Table 4:** Mixture content for FeSi 75, g

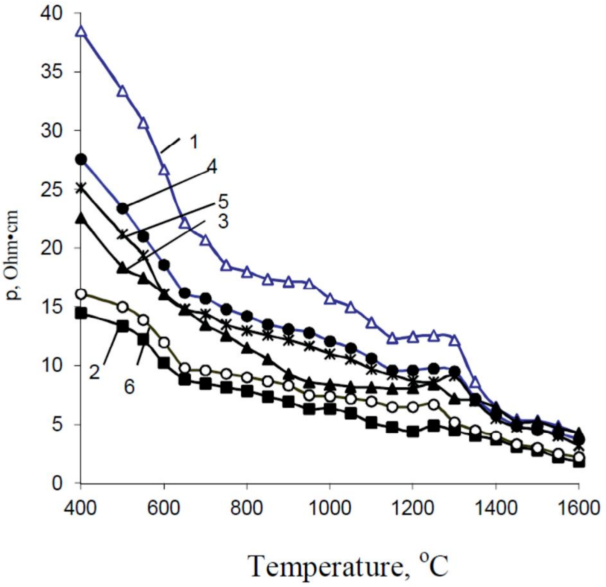
| **Material** | **Mixtures** | | | | | |
|---|---|---|---|---|---|---|
| | **1** | **2** | **3** | **4** | **5** | **6** |
| Quartzite | 50 | 50 | 50 | 50 | 50 | 50 |
| Steel turnings | 6,5 | 6,5 | 6,5 | 6,5 | 6,5 | 6,5 |
| Special coke (dry) | 22,70 | - | - | 17,02 | 11,35 | 5,67 |
| Russia coke (dry) | - | 23,00 | - | 5,75 | 11,50 | 17,25 |
| China coke (dry) | - | - | 22,70 | - | - | - |

At relatively low temperatures special coke mixture has the maximum resistivity and Russia coke mixture –the minimum one. Special coke increases mix charge resistivity the more its content becomes. Special coke addition has greater effect if it is more 50%. Temperature decreases all mixtures resistivity. Within the range of 400-1200°C mixtures curves concerning their resistivity are arranged in decreasing order: Special coke mixture has the maximum resistivity followed by mixture containing 25% coke and 75% special coke, and then comes mixture with coke and special coke mix used as a reductant (1:1).



1 - Russian coke, 2 - special coke, 3 - char, 4 - China coke

**Figure 4:** Relationship between temperature and resistivity of reductants



1 - special coke mixture; 2 - Russia coke mixture; 3 - China coke mixture; 4-reducer mixture comprising special coke and coke 3:1; 5 - reducer mixture comprising special coke and coke 1:1; 6 - reducer mixture comprising special coke and coke 1:3

**Figure 5:** Relationship between FeSi 75 mixtures resistivity and temperature

Slightly below there are data on China coke mixture. Coke mixture and a mixture using blend of special coke (25%) and coke (75%) as a reducer have the minimum resistivity.

Within the range of 1150-1300°C, resistivity level equalization can be observed, iron turnings having high conductivity are melting which results in some resistivity increase.

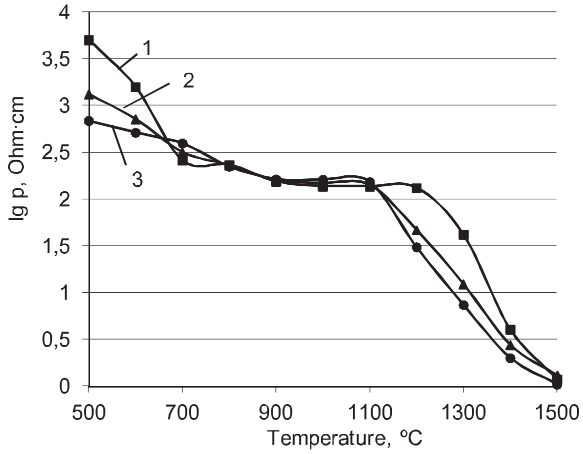
Temperature increase above 1300°C reduces mixtures subsequent resistivity also levels resistivity of the mixtures containing 50-100% of special coke and China coke. Resistivity values of mixtures containing 100 and 75% of coke, get equalized, however their resistivity being 1,5-2 times less locate them somehow below than others.

Thus, special coke addition into mixture for ferrosilicon smelting raises its resistivity upon special coke share increase.

Electrical resistivity of carbon ferrochrome mixture. The investigation has been performed within the range of 400-1500°C . Molybdenum tips have been installed onto graphite electrodes carbon to prevent their interaction with chromite ore. Top electrode was pressed under 0,5 kg/cm$^2$ and heated up to 50-60°C/min which confirms to commercial furnace conditions. Chromite ore comprised two fractions: fines (0,2-2,0 mm) and lumps (3-10 mm) in the ratio 2:3 (by weight).

Russia, China and special coke was sized 2-5 mm. Charges bed depth amounts to 40 mm. Pre-dried materials are being stirred up and charged into a preheated up to 400°C furnace. Figure 6 summarizes the investigation results.

Within the range of 500-600°C coke mixture proves to have the highest resistivity, following by China coke mixture, and special coke mixture have the lowest electrical resistivity. Temperature increase rapidly decreases mixture resistivity, charge and reduce slowly upon reaching 650-1000°C. Temperature of more than 1100°C is indicated by rapid reduction of special coke and China coke mixtures. Rapid reduction of Russia coke mixture electric resistivity occurs if temperature is considerably higher (higher than 1200 °C). Upon reaching 1400-1500°C temperatures all charges electrical resistivity levels get equal and total to 1-2 ohm·cm.



1 - Russia coke mixture; 2 - China coke mixture; 3 - special coke mixture

**Figure 6:** Specific electrical resistivity of HCFeCr mixture

Special coke mixture giving low electrical resistivity at temperatures 500-600°C can be explained by special coke light bulk weight. Chromite ore having high electrical resistivity at low temperatures breaks a contact between electrically conductive carbon material and increases charge general resistance. Chrome ore concentration by volume in special coke charge makes 46%, same concentration in coke charge-54%. Special coke charge has high electrical conductivity due to its low volume rate of chrome ore. Chrome ore volume in China coke charge is 49%and, consequently, specific electrical resistivity value of charge with china coke within the range 500-600° C takes intermediate position between specific electrical resistivity values of mixtures with special coke and coke.

Within the range of 650-800°C special coke mixture has the highest specific electrical resistivity. This phenomenon is associated with decrease of special coke volume in a mixture which is indicated by charge shrinkage, observed in the specified temperatures range, also start of reductants`s own conductivity effect onto general charge electrical resistivity.

Mixture high electrical conductivity at more than 1000°C can be explained by recovery and metallization processes. China coke and special coke mixture resistance decreases rapidly at 1100°C, and Russian coke mixture - at 1200°C. Charge shrinkage occurs at the same temperatures indicating reduction processes commencement. The lowest electrical resistivity of special coke mixture at temperatures above 1000°C results from reduction processes progression.

Thus, at relatively low temperatures special coke mixture have no considerable electrical resistance advantages over coke mixture. Specific electrical resistance of special coke considerably exceeds coke resistivity however this difference levels up, since ore resistivity in carbon ferrochrome charge is several orders greater than the one of carbon reductants. Minimum electrical resistance at 1100-1400°C provides increase of fraction of power being released at solid-phase processes zone. At 1400-1500°C specific electrical resistivity of charge with various reductants stay at one level. According to [14] while melting ferrochrome in the range of 1400-1500°C, charge resistivity is to be sufficiently provided within the range of 0,3-0,6 ohm·cm. The specific electrical resistivity using the special coke mixture at the mentioned temperatures is 1-2 ohm·cm and meets the requirement.

## 3. CONCLUSION

Laboratory scale investigation evaluated electrical and reactivity properties of the new type reducing agent– special coke derived during high speed pyrolysis reaction of lump Shubrakol coal (Kazakhstan) over conventionally used reductants. Obtained results proved advantages of special coke and enabled successful tests on industrial scale. Using this new reductant, efficient technology of ferrochrome and ferrosilicon smelting was developed [15-17].

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