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DECREASING CO_2 EMISSIONS IN METALLURGY

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Modern trends in iron metallurgy. Global carbon emissions from iron production. New solution for iron and steelmaking in a way reduce the CO_2 emission to the level, where this might be needed in the post - Kyoto period. New blast furnace technology, which operate with very low CO_2 emissions based on drastically reduced consumption of carbon containing input materials. Injection of reducing gases and oils into the furnace. Injection of materials containing hydrogen permits material recycling and influencing the coke consumption of the blast furnace process. Direct reduction and eliminating CO_2 emissions in Iron metallurgy. Technologies and processes with CO_2 removal. Cost optimized CO_2 reduction.

Key words: *metallurgy, blast furnace, CO_2 emission, decreasing*

Smanjenje emisije CO_2 u metalurgiji. Moderni putokazi u metalurgiji željeza. Ukupna ugljikova emisija u tehnologiji izradbe željeza. Nova rješenja u smanjenju CO_2 emisije na razini zahtjeva po Kyoto protokolu. Nove tehnologije visokih peći koje se temelje na jakom smanjenju rabljenja ugljena. Ubrizgavanje reduciranih plinova i ulja u peć. Ubrizgavanje materijala koji sadrže vodik omogućuje recikliranje materijala i utječe na rabljenje koksa u procesu visoke peći. Direktna redukcija i eliminacija emisije CO_2 u metalurgiji željeza. Optimizacija troškova sniženja emisije CO_2.

Ključne riječi: *metalurgija, visoka peć, CO_2 emisija, smanjenje*

INTRODUCTION

The blast furnace will remain dominant for the production route from iron ores to crude steel. In Europe, global warming is perceived by politicians, the media and the public as the results of human activity. The blame is generally pinned exclusively on CO_2 emissions. The majority of high-emission countries have not signed Kyoto Protocol up or not agreed reduction targets, with the results that the impact on the earth's climate will be negligible.

Clearly, in the 21 st century, environment will become the new driver for the Steel Industry.

We - in Czech Republic- must continue our successful efforts to make our existing production processes more material and energy efficient and we must develop new technologies that make significantly lower use of resources. Springs of CO_2 emission in Ostrava - region demonstrate Figure 1.

Along this trend, ironmaking technology will be in the first row, owing to its determinant role with respect to sustainability; new challenges regarding emissions, recycling and CO_2 issues will be a major concern, calling for innovative breakthrough technologies, new routes of production, along with the basic need for continuous improvement of performance.

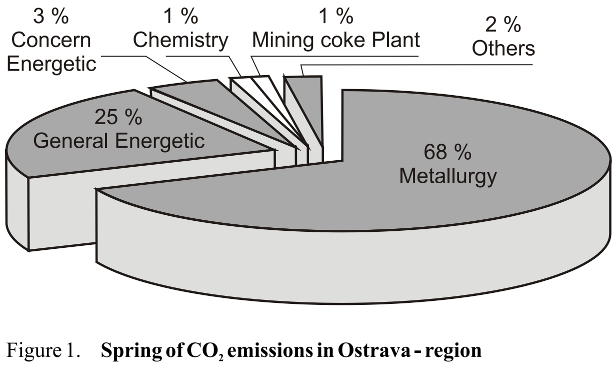

Figure 1. **Spring of CO_2 emissions in Ostrava - region**
Slika 1. **Pregled CO_2 emisije u Ostrava - području**

The blast furnace exhibits an extended flexibility with respect to the iron sources. The ferrous burden results from a local optimization between the iron sources: sinter, ores, pellets, DRI/HBI or scraps.

In West Europe, the coke rate has been divided by more than a factor 3, during the last 50 years. This trend has re-

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sulted from the improvement of the ironmaking technology and the subsequent optimalization of the process control.

Coal injection has now become the most developed way to replace coke. Blast furnaces in Europe are operated at coal rates close to 200 kg/t_{HM}. The replacement rate of coke by coal is on the order of 0,80 - 0,85. Oil injection has received renewed consideration, in the late nineties, with typical injection rates averaging at 120 kg/t_{HM}. Natural gas injection is used in the North American blast furnaces. The injection rates are in the range 40 to 110 kg/t_{HM}.Due to the high endothermic effect is required in order to maintain a suitable flame temperature at the tuyere. [1, 2].

Iron ores are chemically reduced with the aid of carbon and hydrocarbons. The gaseous products that are inevitably formed during these reducing reactions are carbon monoxide, carbon dioxide, hydrogen and steam. The consumption of reducing agents in the blast furnace is one of the major cost influencing factors in the production of hot metal. Now as before, it is therefore a chief duty of the blast furnace operator to minimize the consumption of reducing agents.

Regardless of the type and chemical composition of the auxiliary reductans injected, the total consumption of reducing agents is equivalent, by a first approximation, to the aggregate carbon dioxide emission.

The driving force for cost reduction in ironmaking has resulted in a drastic reduction of the energy consumption during the last 40 years. This has resulted in a strong decrease of the CO_2 intensity ironmaking. The potential of further decrease of CO_2 emission of the traditional route is quite limited, probably no more than 5 - 10 %. Indeed, blast furnace technology has reached such a high level of efficiency than any improvement is quite difficult. The substitution of coke by injected coal has only little effect on the total CO_2 emission, than by injected carbon gas, degasation gas, oil or tar is effect major. In addition, material and heat balance show that carbon is more effectively used in a blast furnace than in a power plant.

Possibilities of measures for CO_2 emission in iron metallurgy are illustrated in Figure 2.

SUBSTITUTE FUELS AND BLAST FURNACE TECHNOLOGY

Adding of substitute fuels causes the following technological effects:
- saving of specific coke consumption,
- increasing of richness of the mixture by lower need for basic slag formers,
- reducing of specific occurrence of slag,
- increasing of specific occurrence of gas in the lower part of the furnace (in relation to the coke),
- reducing of combustion temperature before tuyeres,
- changing of counter-current intensity in the lower part of the furnace,
- changing of the gas flowing structure in the lower part of the furnace,
- changing of lower zone permeability, especially in the area of the slag creation and flowing zone,
- changing of the fuel combustion intensity.

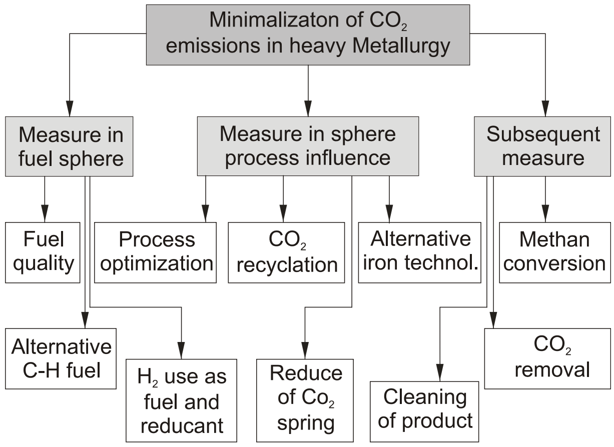

Figure 2. **Minimalization CO_2 emission from stand point of measure character**

Slika 2. **Minimalizacija CO_2 emisije s gledišta mjernih karakteristika**

The coke specific consumption reduction is the primary dominant effect. It is commonly characterized by the substitution (exchange) coefficient k_z, which is actually the ratio of the coke reduction and quantity of the added substitute fuel. It is generally known that this coefficient decreases with increased fuel specific consumption. As this decrease does not result from common balance calculations using the chemical and thermodynamic dependence, it is most likely affected by the change of the gas flowing structure in the lower part of the furnace and possibly by other common non-monitored factors. These factors cannot be determined by modelling so far; they result from evaluation of specific experiments and are the source of certain variance of results.

Connection of the effect of the coke reduction and decrease in the need for basic substances in the mixture comes out in reduction of the slag specific occurrence. This fact moderates the negative effect on the counter-current intensity in the lower part of the furnace.

An increase in the gas specific occurrence in the lower part of the furnace in relation to the coke rate is the result of reduction of the share of gas occurrence from the coke. The change of the gas specific occurrence in relation to the pig iron is dependent on the value of the substitution coefficient k_z.

The changes we have mentioned so far (reduced coke specific consumption, higher richness of mixture, reduced specific occurrence of slag, changed gas specific occurrence) can be determined based on material and thermal balances, with possible correction based on the operational verification results. The following economic calculations considered the dominant effect of the summary substitution coefficient without respecting the effect of changes on the mixture's richness.

NATURAL (CARBON) GAS IN THE CZECH REPUBLIC

Coal deposits constitute a special case when the same deposit is a coal resource as well as a gas reservoir. In the plant residues coalification process, so-called carbon gas (identified as coalbed methane CBM) was created, the main component of which is methane.

The methane release that accompanies the coal mining from the coal deposits is a risk factor, because this mixture is explosive at the concentration of 5 to 15 % of methane mixed with air. Therefore, the mixture of air with methane (degazation gas) is exhausted during the underground exploitation of coal. The importance of the mining operations degazation consists not only in reducing the explosion risk, but also in preventing emissions of methane to the atmosphere. Besides carbon dioxide emissions, methane is the second most significant component of emissions causing the greenhouse effect. Foreign studies [3, 4] document that the annual methane emissions accompanying coal exploitation reach 23×10^6 t on a world-wide scale, of which only 7 % is captured and utilized as degazation gas.

In the Czech Republic, a lot of attention was paid to the problems with providing carbon gas exploitation after 1992 [5]. In 1994, the vertical wells into productive carbon with well completion and hydraulic partitioning of promising stratum horizons verified the workable reserves of coal methane [6].

The growth of the carbon gas exploitation is also supported by the possibility of increasing the exploitation thereof by forcing the carbon dioxide through vertical wells to the coal deposit (Figure 3.). More than 57 mil. m^3 of CO_2 have been stored in the coalbeds in the USA since 1996. The worldwide potential of storing CO_2 in the coalbeds is appraised at 150 Gt [7].

Thus, the carbon dioxide injection into the coalbeds has a double meaning: enables an increase of the carbon gas exploitation and reduces the volume of emissions of CO_2 forced into the atmosphere. The test operation in the USA shows that the coal absorbs approximately twice as much carbon dioxide as methane, i.e. 2 molecules of sorbed CO_2 desorbs 1 molecule of CH_4.

Coal methane exploitation is in Czech Republic a broadly positive process, because:

- the captured methane may reduce the high costs of the natural gas import,
- a portion of the non-exploited coal methane would necessarily have to get to the air in the coal mining process, where it would contribute to the greenhouse effect,
- the coal reserves free of sorbed methane will be more secure for the prospective mining, as it will eliminate the possibility of creation of the explosive methane-air mixture.

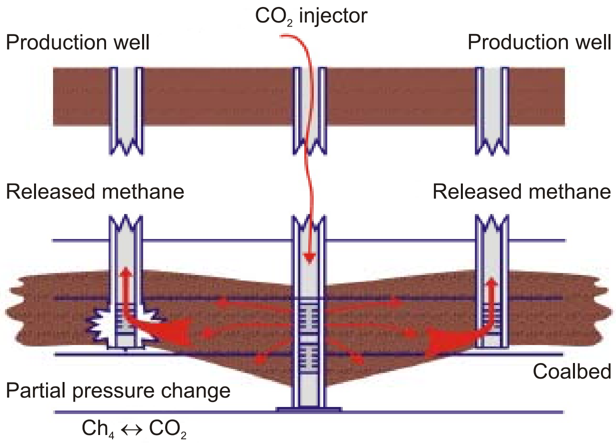

Figure 3. **Scheme of carbon gas forcing from coalbed by forcing CO_2**

Slika 3. **Shema upuhivanja ugljičnog plina iz ugljena posredstvom CO_2**

From the standpoint of fast utilization of large quantities of gas in metallurgy, the utilization of degazation gas seems to be more realistic in the nearest future.

Adding substitute gas fuels to the blast furnace affects the metallurgical processes in the blast furnace, as well as the total material and heat balance. In particular, the impact on energy consumption is significant with the aim of reducing the production costs per ton of pig iron.

Regarding the existing gas distribution systems and technological processes in the Czech iron mills, the following suitable „available" gases can be considered:

- natural gas,
- coke-oven gas from own production,
- degazation gas,
- carbon gas (1960-1961, future ?).

However, none of the above-mentioned suitable and "available" gases is available in the Czech iron mills in the required quantity, whether for one or two blast furnaces, without major investments in main gas conduits.

PREDICTION OF LIMITING FACTORS OF SUBSTITUTE C-H FUELS INJECTION TO BLAST FURNACE TUYERES

At the Ironmaking and Coking Institute of VŠB - TU Ostrava, a number of mathematical metallurgical processes were gradually developed in the past years, which in general create a so-called analytical blast furnace process evaluation system [8, 9]. The system includes sub-models of an evaluation and prediction character, e.g. prediction of theoretical combustion temperature before tuyeres, prediction of minimum coke consumption required for ensuring the reduction and thermal processes, prediction of minimum consumption of coke required to ensure the furnace permeability, prediction of coke degradation in the blast furnace, prediction of the indirect reduction course, prediction of coke reactivity, etc.

The complex prediction of the total impact of using substitute fuel/reducer also required an actual amendment of models of predicting chemical, thermal and gas-dynamic processes. The sub-models of prediction of the minimum coke consumption were worked out for the principal prediction of the main parameter - specific consumption at massive injection of one or more substitute fuel types, using determination of the marginal limits from the standpoint of consumption of C (coke) for reduction and thermal needs (Figure 4.).

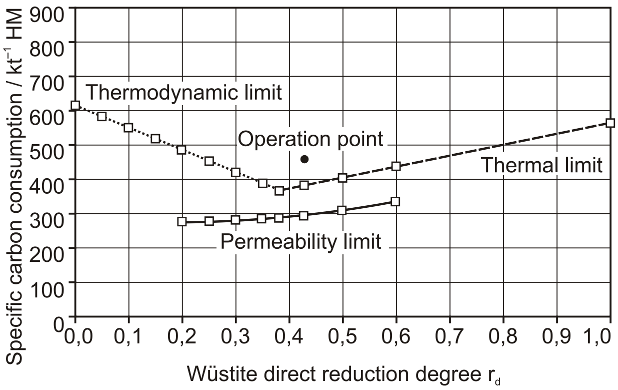

Figure 4. **Complex analysis of minimum consumption of reducer fuel - CDR diagram**

Slika 4. **Cijelovita analiza minimalne potrebe goriva - CDR dijagram**

The prediction methodology for the permeability limit is based on calculation of gas-dynamically minimum consumption of coke, which can be determined by the original model respecting the factor of dynamic balance of the gas counter-current and mixture at the bosh. The model algorithm is provided in [10].

The boundary line characterizing the permeability is calculated only as the coke carbon consumption and, as the verification calculations have proven, it is not limiting for the blast furnaces in case of small-scale adding of substitute fuels (Figure 4.). In this situation, the "permeability" limit has only a complementary significance.

However, the situation will be completely different in the case of injecting a large amount of fuels into the blast furnace tuyeres, when the thermal and kinetic limit of coke consumption will be significantly reduced, and from the standpoint of the coke specific consumption, the permeability limit will become limiting.

These considerations are based on the following question; how will the large amount of injected fuels develop at the beginning of the process and what will be the final effect of coke substitution by this fuel that is not traditional for the blast furnace?

The practical verification in the blast furnace process cannot, of course, be completed until the technical equipment for the substitute fuels preparation and injection is installed. The technical equipment cannot be installed, whether only for a test or temporary run or to the limited extent. This does not enable practical verification of how the injection technology will apply in rather different conditions, what the injection limits will be and whether the large of the injected substitute fuel required by economic factors will not endanger the furnace permeability or thermal equilibrium.

The above-mentioned model of the counter-current dynamic equilibrium model in the lower part of the furnace, where the negative impact of the increased coke degradation is mainly applied, was used for further specification of prognostic forecasts. The coke degradation extent is positively affected by a smaller extent of Boudouard reaction; however, the other degradation effects will be significantly intensified as a result of significant extension of the coke presence in the exposed area of the coke bed. Coke exposed to longer action of alkali and high temperatures will reduce its porosity, and thus the gas permeability in the critical area.

The gas permeability of the coke bed is further impaired by relative increase in volume of the smelting liquid products per unit of enforced coke, as well as by the increased volume of gases produced by conversion before the tuyeres.

The result then is that the limit coke consumption ensuring the gas permeability shown in the CDR-diagram in modified coordinates coke/direct reduction degree will exceed the limit values of the coke consumption for heat and reduction during massive injecting. These consumptions are partially covered by the injected fuel and the coke required for these decreases according to the mutual interchangeability coefficient.

Comparison of the minimum needs of coke for heat + reduction and for ensuring the permeability is shown for the current Czech conditions in Figure 5. The picture shows that permeability of the lower part of the furnace should not reach its critical value up to the injected volume of approximately 150 m^3 (115 kg) of carbon gas per ton of pig iron. In the American plants, the injection values of 100 m^3 and more of the natural gas [2] per ton of pig iron were achieved without technological difficulties.

The analysis of interim results and partial criteria received from the model calculation pointed at the factors affecting the porosity of the lower part of the furnace hightes and also critical level of the porosity. In particular, this concerned the specific volume of slag, input granulometric composition of coke and its strength and reactivity mainly at high temperatures, achieved by direct reduction of FeO and the aerodynamic conditions of the gas flow in the mixture.

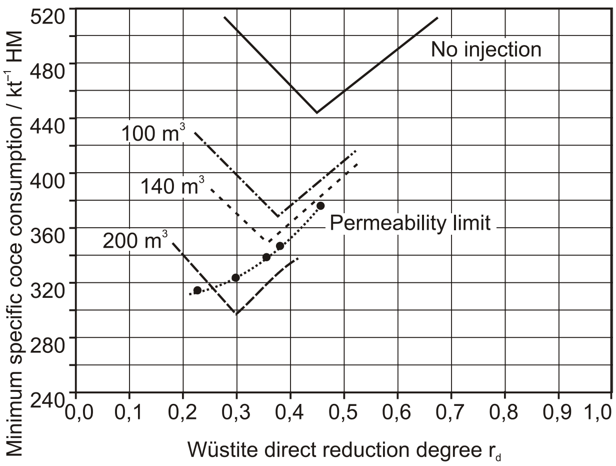

Figure 5. **The carbon gas injection limits in view of the blast-furnace process gas-dynamics**

Slika 5. **Granica ubrizgavanja ugljičnog plina s gledišta plinske propusnosti visoke peći**

The other limiting factor, which is that the injection must not change the thermal condition of the furnace, also results in the limit values of the mutual interchangeability, which is the substitution coefficient k_z, as well as the limit volume of injection from the standpoint of maintaining the technologically required temperatures before the tuyeres.

Besides the absolute values of the injected volume, the interchangeability also decides on the total amount of saved coke. The value of this coefficient says how many kilograms of coke will be replaced by 1 kg of injected gas. It cannot be determined absolutely reliably by the model calculations, as it is affected by a number of various effects.

However, it is still possible to make a rather reliable calculation on the condition of thermodynamic course of the reduction, or suitable utilization by kinetic modelling of the determined corrections, on the condition of achieving the required parameters of iron and slag, as well as correctly estimated thermal losses. The result of such calculation for technology and mixture conditions of a selected Czech blast furnace is provided in Figure 6. This Figure illustrated the comparison of the effect of various kinds of injection including the brown and black coal.

The middle coefficient of the natural gas substitution considered here is approximately 1 kg of coke per 1 kg of injected gas. In real cases, we can expect that the substitution coefficient will be relatively higher (up to 1,2) with a smaller quantity of injected gas, and relatively small with massive injection. For comparison, the Figure shows the substitution coefficient for coal as well. The substitution coefficient determined for coal by model calculation is relatively small - approximately 0,75 kg of coke per 1 kg of injected coal, however, the real coefficient does not fall so much as compared with this value for the massive injection. On the other hand, the coal injection should enable utilization of less expensive types, while the brown coal has a certain chance, as well, especially if it was possible to use carbonificated substance gained in pyrolysis or copyrolysis with still inexpensive coal. This modified brown coal, as well as suitable black coals, reduces the temperature of the oxidation area before tuyeres only a little and does not require compensation by oxygen or wind temperature.

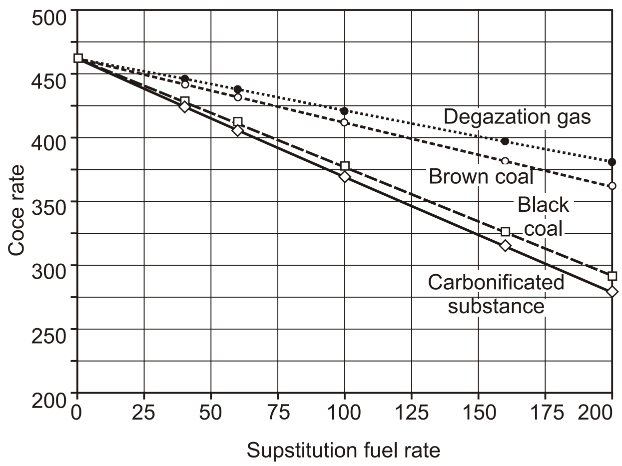

Figure 6. **Comparison of impact of the substitute fuel injection on coke consumption**

Slika 6. **Usporedba koeficijenata zamjene ubrizganog goriva na potrošak koksa**

The brown coals differ from the black coals petrologic-genetically, physically, as well as chemically. Unlike the black coal, these mostly have a high content of water (W_t^r up to 50 %), so they require drying for the related enriching processes. The high content of ashes (A^d over 20 %) and difficult modification predetermine a major part of the exploited brown coal for combustion, i.e. utilization in industrial power generation for electricity and heat production. Besides these high-ash kinds of coal, it is possible to exploit medium-ash coal from the active mines with approximately 15 % ash content, as well as low-ash kinds with the ash content close to 6 %.

Compared with the black coal, the brown coal combustible has a lower content of carbon and hydrogen and multiple content of oxygen, which belongs to the ballast elements as for the coal improvement processes.

Right after combustion, the low-temperature or, as the case may be, high-temperature, carbonization of brown coal is the second most significant technological process of its processing. It produces semi-coke - carbonificated substance, liquid products and gas products; another output is reaction water. With respect to aspects of these products, mainly the carbonificated substance comes into question for use in iron metallurgy.

ECONOMIC EVALUATION OF ECOLOGICAL UTILIZATION OF CARBON GAS OSTRAVA REGION FOR IRON PRODUCTION

The methodology of the economic evaluation of the carbon gas utilization in the pig iron production is based on the basic general relations of determining the costs of the blast-furnace process [11], as well as on the results of modeling and practical tests of adding the substitute gas fuel. The final relation for determining the specific costs combines the economic data with the data of the mixture entering the process (*M*), as well as technological values of the specific consumption of fuel *K* and fuel combustion intensity *I*. This combination immediately shows that it is not possible to objectively evaluate the final impact on the pig iron production economy without determining the effect of any change in technology, mixture or organization on the technological factors of the fuel specific consumption, as well as the furnace operation intensity. When applying this relation, we cannot omit the fact that the above-mentioned changes may modify the economic variables as well. The results of calculations of the reduced costs of the pig iron production when using the natural carbon or degazation gas, including the alternative of using the degazation gas free of nitrogen, are provided in Figure 7.

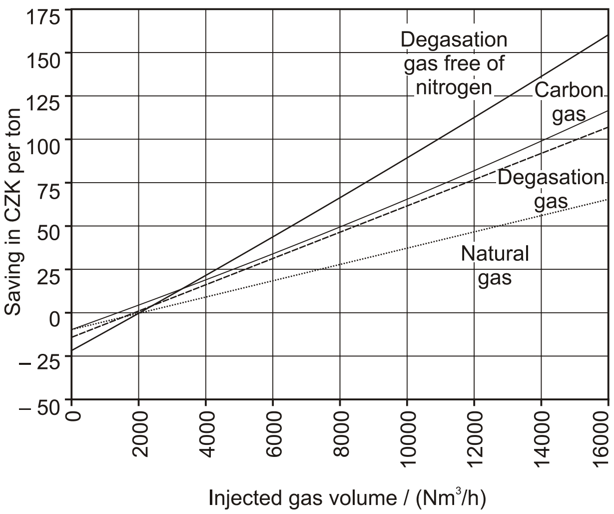

Figure 7. **Reduced costs of pig iron production**
Slika 7. **Sniženje troškova pri izradi sirovog željeza**

The curves show that the significant cost reduction occurs in all investment alternatives upon exceeding the minimum volume of injected gas of approximately 2,000 m^3/h. While adding the natural gas is perhaps less favourable because of the high price (though still advantageous), the best alternative seems to be elimination of nitrogen from regular degazation gas.

With these considerations, it is important to realize that the substitute fuel injection not only deals with the cost reduction, but also resolves the disequilibrium or lack of the need for metallurgical coke.

There was an analysis of sensitivity to changes in critical parameters conducted for the DGP alternative and injected volume of 10,000 m^3/h (cost reduction by 2 euro/$t_{pig\ iron}$). The results are:

Change in Parameter	Reduction by /$t_{pig\ iron}$
Reduced price of coke by 16,7 €/t	0,570
Decreased substitution coefficient by 0,01	0,277
Increased price of degazation gas by 0,07 €/m^3	0,541
Exceeded capital expenditures by 166 666,7 €	0,031

The prognosis of the trend in coke prices and injected gas prices will be critical for the final decision on the investment in adding the gas, while the eventual exceeding of capital expenditures should be less significant.

The achieved data was used for calculating the investment rate of return (without considering the time value of money and possible taxes on increased profit). The results shown in the Figure below show that it is necessary to inject the volume of fuel exceeding 7,000 m^3/h for the acceptable rate of return of less than two years.

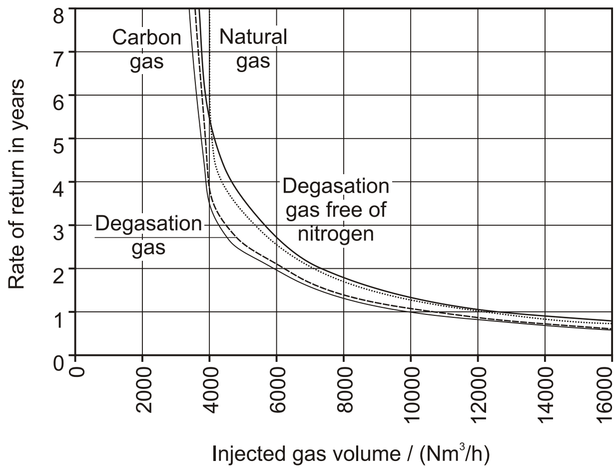

Figure 8. **Rate of investment return in years**
Slika 8. **Koeficijent povrata investicije tijekom godina**

CONCLUSION

Creation and modification of a suitable mathematical model for the computer simulation of degazation/carbon gas for the blast furnace and for the stack-type reduction reactor were required for predicting the limiting factors of the injection in the conditions of specific blast-furnace plants. The prediction calculations mainly concern the impact of injection for thermal and gas-dynamic condition in the lower part of the blast furnace. The documented results mainly show the predicted middle and marginal values of the coefficient of substituting the coke by substitute fuels for various forms of carbon gas or coal which were considered.

The final part of the paper presents economic evaluation of utilizing the carbon gas from Ostrava region in the blast-furnace production of iron. The reduction of costs was calculated, which shows that the significant cost reduction occurs at injection exceeding 2000 m^3 per hour. However, it is also important that the injection of the substitute gas fuel not only brings a cost reduction, but it also resolves the problems of CO_2 emission.

The simple rate of the investment return was also calculated. According to the results, it is necessary to inject the gas volume exceeding 7000 m^3 per hour for the acceptable rate of return of less than two years.

This report has the objective to assess whether the carbon and hydrocarbon carriers used for the reduction of iron ore represent in the main chemical raw materials or fuels from a scientific and technological point of view. This is of particular importance for materials such as heavy oil, brown coal, black coal, degasation and carbon gas which are injected via the tuyeres of a furnace as the classification of such materials depends on this assessment witch regard to CO_2 emission decreasing.

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