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DETERMINING START PARAMETERS FOR OXYGEN–WATER STEAM GASIFICATION  
WITH AND WITHOUT RECIRCULATION

URČENÍ POČÁTEČNÍCH PARAMETRŮ PRO ZPLYŇOVÁNÍ KYSLÍK – VODNÍ PÁRA  
S A BEZ RECIRKULACE

**Abstract**

The analytical, unified selection method of the converting medium composition is presented as to apply for the oxygen – water steam gasification the recirculation of reaction products. The selected molar fractions of the converting medium components should give the assumed (needed) CO/H<sub>2</sub>-ratio of the synthetic gas by the chosen process pressure and temperature. The graphical selection methods applying the MOLLIER–HOFFMANN diagram are used for the parameters preselecting of the air – water steam gasification, e.g. if there is any contents of nitrogen from the atmosphere. The presented analytical method refers to the oxygen – water steam without and with gas recirculation. The last assumption allows the presence of CO, H<sub>2</sub>, CO<sub>2</sub> and CH<sub>4</sub> in the converting medium — new gasifying technologies need the permanent presence of carbon monoxide and hydrogen because of the iron catalyst. The determined values of its molar fractions help to choose the suitable process calculation iterative method.

**Abstrakt**

Analytická unifikovaná selektivní metoda přeměny středního složení je prezentována a aplikována pro recirkulaci reakčních produktů zplyňovacího procesu kyslík–vodní pára. Vybrané molární frakce přeměněných produktů dávají potřebný předpoklad určení poměru CO/H<sub>2</sub> syntetického plynu podle tlaku a teploty V daném procesu. Grafická unifikovaná metoda používá MOLLIER–HOFFMANN diagram pro předběžné určení parametrů vzduch–vodní pára U zplyňovacího procesu, jestliže je zde obsažen dusík Z atmosféry. Prezentována analytická metoda se odkazuje na proces kyslík–vodní pára bez a S recirkulaci. Poslední předpoklad dovoluje přítomnost CO, H<sub>2</sub>, CO<sub>2</sub> a CH<sub>4</sub> V přeměňujícím se mediu – nové zplyňovací technologie potřebují S ohledem na katalyzátor – železo, stálou přítomnost CO, H<sub>2</sub>. Určené hodnoty těchto molárních frakcí pomáhají vybrat vhodnou výpočtovou iterační metodu.

## 1 INTRODUCTION

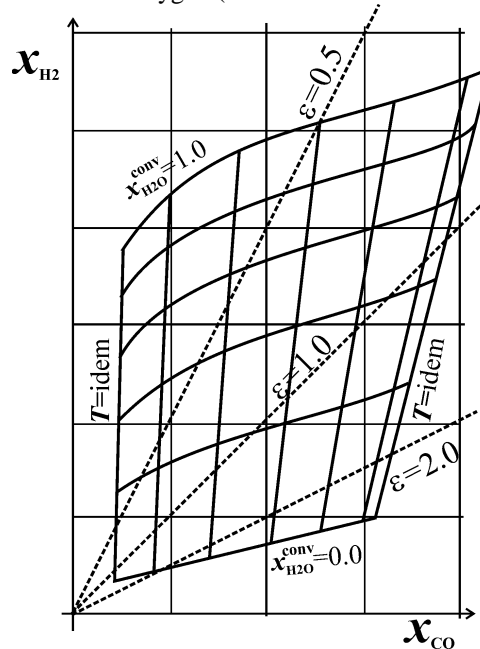
Gasifying processes are very important in the chemical industry, but also in the modern power engineering. The synthetic gas should be of needed composition, e.g. the gas used in the FISCHER–TROPSCHE method for producing synthetic liquid fuels the molar CO/H<sub>2</sub>-ratio should have the value of  $\varepsilon=2$ .

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The selection of parameters for the gasifying process can be made using the so-called MOLLIER–HOFFMANN  $x_{H_2}/x_{CO}$  diagram, which can be prepared for a chosen fuel, i.e. for hydrogen and carbon contents in it. At the diagram appropriate process temperature can be determined and the needed composition of the converting medium. The last one is the mixture of atmospheric air and water steam, with limits pure water steam and pure atmospheric air. The last component can be enriched with oxygen, but there is not possible to apply the MOLLIER–HOFFMANN diagram to the mixture of water steam and pure oxygen. In the practice, it will be taken into account, that the atmospheric air is highly enriched with oxygen (the so-called technical oxygen).



**Fig. 1** Graphical selection of the main coal gasifying process parameters [4].

An example of such a diagram is presented in Fig. 1. The  $\varepsilon$ -value lines refer to the synthetic gas composition, in detail. There is no problem to read other main process parameters: temperature  $T$  and converting medium (agent) composition  $x_{H_2O}^{conv}$  (the rest is atmospheric air with oxygen contents of 21%, which has been taken into account by preparing the diagram in Fig. 1., [4], — for the technological coal according to FRAN BOŠNJAKOVIĆ can be taken the formula  $CH_{0,64}$ , [1]–[3]).

The new fuels gasifying technologies, especially of coke (can be discussed as a pure carbon C) are realized using sophisticated iron catalysts, [5], although their real usability is discussed and not that sure, e.g. [6]–[7]. Because of the catalyst, however, the permanent presence of carbon monoxide and molecular hydrogen in the reactive mixture is needed, [8]–[9]. It can be achieved by recirculation of a certain part of the actual reactive mixture — it is led back into the mixture (gas generator) with the converting medium, e.g. [2]. Thus, the converting agent in general consists of  $H_2$ ,  $H_2O$ ,  $CH_4$ ,  $CO$ ,  $CO_2$  and  $O_2$ .

## 2 PARAMETERS OF THE $H_2O$ – $O_2$ GASIFICATION PROCESS WITHOUT RECIRCULATION

Let's assume that the needed  $CO/H_2$ -ratio of the resulting synthetic gas should be

$$\varepsilon = \frac{n_{CO}}{n_{H_2}} = \frac{x_{CO}}{x_{H_2}}$$

Following equilibrium amounts of particular species should be present in the reaction mixture:

$$x_{\text{CO}_2} = \frac{x_{\text{CO}}^2}{K_{(1)}} p = \frac{\varepsilon^2 x_{\text{H}_2}^2}{K_{(1)}} p \quad x_{\text{H}_2\text{O}} = \frac{x_{\text{CO}} x_{\text{H}_2}}{K_{(2)}} p = \frac{\varepsilon x_{\text{H}_2}^2}{K_{(2)}} p \quad (01)$$

$$x_{\text{CH}_4} = K_{(3)} x_{\text{H}_2}^2 p \quad (02)$$

which refer to chemical reactions  $\text{C} + \text{CO}_2 = 2\text{CO}$ ,  $\text{C} + \text{H}_2\text{O} = \text{CO} + \text{H}_2$  and  $\text{C} + 2\text{H}_2 = \text{CH}_4$ . It will be assumed that in the reacting mixture is not present the nitrogen from the atmospheric air and the nitrogen from the fuel. For such a mixture yields

$$x_{\text{CO}} + x_{\text{CO}_2} + x_{\text{H}_2} + x_{\text{H}_2\text{O}} + x_{\text{CH}_4} = 1 \quad (03)$$

Applying Eqs.(01)–(02) the quadratic equation with an argument  $x_{\text{H}_2}$  will be to

$$\varepsilon x_{\text{H}_2} + \frac{\varepsilon^2 x_{\text{H}_2}^2}{K_{(1)}} p + x_{\text{H}_2} + \frac{\varepsilon x_{\text{H}_2}^2}{K_{(2)}} p + K_{(3)} x_{\text{H}_2}^2 p = 1$$

or

$$\left( \frac{\varepsilon^2}{K_{(1)}} + \frac{\varepsilon}{K_{(2)}} + K_{(3)} \right) p x_{\text{H}_2}^2 + (\varepsilon + 1) x_{\text{H}_2} - 1 = 0 \quad (04)$$

where from it becomes

$$x_{\text{H}_2} = -\frac{\varepsilon + 1}{2p \left( \frac{\varepsilon^2}{K_{(1)}} + \frac{\varepsilon}{K_{(2)}} + K_{(3)} \right)} + \frac{1}{2p} \sqrt{\left( \frac{\varepsilon + 1}{\frac{\varepsilon^2}{K_{(1)}} + \frac{\varepsilon}{K_{(2)}} + K_{(3)}} \right)^2 + \frac{4p}{\frac{\varepsilon^2}{K_{(1)}} + \frac{\varepsilon}{K_{(2)}} + K_{(3)}}}$$

For the case the process pressure is moderate it can be assumed  $K_{(3)}=0$ , i.e. the existence of methane in a reaction mixture is negligible. It is an advantageous and correct assumption because equilibrium constants  $K_{(1)}$  and  $K_{(2)}$  are given as temperature functions by the standard process pressure ( $p=1$  at), and none of the reaction taken into account is an equimolar one in a gas phase. Assuming  $K_{(3)}=0$  can be treated as a first iteration step that helps the following procedure. The aim is to determine process temperature  $T$  at given process pressure  $p$ , the  $\text{CO}/\text{H}_2$ -ratio in the reactive mixture at equilibrium and the amount of hydrogen in it.

Assuming that the converting medium consists only of oxygen and water steam (with the both limits: pure oxygen and pure water steam — this was in fact the reason for these investigations) their molar ratios are  $x_{\text{O}_2}^{\text{conv}}$  and  $x_{\text{H}_2\text{O}}^{\text{conv}}$ , respectively, and it is used to gasify a pure coke (the element carbon C), following balances can be formulated for hydrogen and oxygen elements in a reactive mixture:

$$(H) = 2n^{\text{conv}} x_{\text{H}_2\text{O}}^{\text{conv}} = 2x_{\text{H}_2} + 2x_{\text{H}_2\text{O}} + 4x_{\text{CH}_4} \quad (05)$$

$$(O) = 2n^{\text{conv}} \left( x_{\text{O}_2}^{\text{conv}} + \frac{1}{2} x_{\text{H}_2\text{O}}^{\text{conv}} \right) = x_{\text{CO}} + 2x_{\text{CO}_2} + x_{\text{H}_2\text{O}} \quad (06)$$

where  $x^{\text{conv}}$  is the converting medium amount per  $n=1$  kmol of the resulting gas (or reactive mixture in equilibrium).

Because there is obviously

$$x_{\text{O}_2}^{\text{conv}} = 1 - x_{\text{H}_2\text{O}}^{\text{conv}}$$

and from Eq.(05)

$$x_{\text{H}_2\text{O}}^{\text{conv}} = \frac{x_{\text{H}_2} + x_{\text{H}_2\text{O}} + 2x_{\text{CH}_4}}{n^{\text{conv}}}$$

Additionally, taking into account Eq.(06), it yields

$$2n^{\text{conv}} \left( 1 - x_{\text{H}_2\text{O}}^{\text{conv}} + \frac{1}{2} x_{\text{H}_2\text{O}}^{\text{conv}} \right) = x_{\text{CO}} + 2x_{\text{CO}_2} + x_{\text{H}_2\text{O}}$$

and after substitution of Eq.(05)

$$2n^{\text{conv}} - (x_{\text{H}_2} + x_{\text{H}_2\text{O}} + 2x_{\text{CH}_4}) = x_{\text{CO}} + 2x_{\text{CO}_2} + x_{\text{H}_2\text{O}}$$

Rearranging this equation, it becomes finally

$$n^{\text{conv}} = \frac{1}{2}(x_{\text{CO}} + x_{\text{H}_2}) + x_{\text{CO}_2} + x_{\text{H}_2\text{O}} + x_{\text{CH}_4} = \frac{1}{2}x_{\text{H}_2}(\varepsilon + 1) + \frac{\varepsilon^2 x_{\text{H}_2}^2}{K_{(1)}} p + \frac{\varepsilon x_{\text{H}_2}}{K_{(2)}} p + K_{(3)} x_{\text{H}_2}^2 p$$

The amount of coke (element carbon C) needed for the assumed process should be higher than the stoichiometric one, i.e.

$$n^{\text{fuel}} \geq (x_{\text{CO}} + x_{\text{CO}_2} + x_{\text{CH}_4})$$

All the hitherto derived dependencies are valid for one kilomole nitrogen-free equilibrium reactive mixture, i.e. the resulting gas. If there would be supplied into the gasifying process the oxygen in an atmospheric air, where its molar ratio is  $x_{\text{O}_2}^{\text{air}}$ , then the nitrogen amount would be equal to

$$n_{\text{N}_2} = n_{\text{N}_2}^{\text{conv}} = \frac{1 - x_{\text{O}_2}^{\text{air}}}{x_{\text{O}_2}^{\text{air}}} n_{\text{O}_2}^{\text{conv}}$$

Taking into account this nitrogen amount it would be possible to determine approximate parameters of the process. The presence of carbon dioxide  $\text{CO}_2$  in the converting medium can be assumed in the same way. In general, however, assuming the presence of nitrogen from atmospheric air and carbon dioxide (or carbon monoxide  $\text{CO}$ ) makes it impossible to select appropriate process parameters in an analytical way.

The whole procedure presented above can be simplified by elimination of methane presence in the resulting synthetic gas, Eq. (04).

The molar amount ratio of element hydrogen and element oxygen according to Eqs. (05) – (06) is a parameter  $A$ :

$$A = \frac{(H)}{(O)} = \frac{2x_{\text{H}_2\text{O}}^{\text{conv}}}{x_{\text{O}_2}^{\text{conv}} + \frac{1}{2}x_{\text{H}_2\text{O}}^{\text{conv}}} = \frac{2x_{\text{H}_2} + 2x_{\text{H}_2\text{O}} + 4x_{\text{CH}_4}}{x_{\text{CO}} + 2x_{\text{CO}_2} + x_{\text{H}_2\text{O}}} \quad (07)$$

The solution due to the molar ratio  $x_{\text{CH}_4}$  of methane in a resulting gas gives

$$x_{\text{CH}_4} = \frac{A}{4}x_{\text{CO}} + \frac{A}{2}x_{\text{CO}_2} + \frac{A-2}{4}x_{\text{H}_2\text{O}} - \frac{1}{2}x_{\text{H}_2}$$

Using Eq.(03), after substituting the last quantity, it becomes

$$A = \frac{1 - x_{\text{CO}} - x_{\text{CO}_2} - \frac{1}{2}x_{\text{H}_2} - \frac{1}{2}x_{\text{H}_2\text{O}}}{\frac{1}{4}x_{\text{CO}} + \frac{1}{2}x_{\text{CO}_2} + \frac{1}{4}x_{\text{H}_2\text{O}}} = 2 \frac{2 - 2x_{\text{CO}} - 2x_{\text{CO}_2} - x_{\text{H}_2} - x_{\text{H}_2\text{O}}}{x_{\text{CO}} + 2x_{\text{CO}_2} + x_{\text{H}_2\text{O}}}$$

and taking into account Eqs.(01),

$$A = 2 \frac{2x_{\text{CO}} - x_{\text{H}_2} - 2 \frac{x_{\text{CO}}^2}{K_{(1)}} p - \frac{x_{\text{CO}}x_{\text{H}_2}}{K_{(2)}} p}{x_{\text{CO}} + 2 \frac{x_{\text{CO}}^2}{K_{(1)}} p + \frac{x_{\text{CO}}x_{\text{H}_2}}{K_{(2)}} p} \quad (08)$$

i.e. the functional dependence

$$A = f(x_{\text{CO}}, x_{\text{H}_2}, T, p)$$

Thus, knowing parameters  $x_{\text{CO}}$ ,  $x_{\text{H}_2}$ ,  $T$  (process temperature is „hidden” in an equilibrium constants  $K_{(1)}$  and  $K_{(2)}$ ) and process pressure  $p$ , the approximate numerical value of the above defined parameter  $A$  — Eq. (07) — can be determined and further the needed composition of the converting medium. There are two possible limits:

$$A \rightarrow 0 \quad \text{the converting medium is pure oxygen} \quad (x_{\text{O}_2}^{\text{conv}} = 1)$$

$A = 1$  the converting medium is pure water steam ( $x_{\text{H}_2\text{O}}^{\text{conv}} = 1$ )

### 3 PARAMETERS OF THE H<sub>2</sub>O–O<sub>2</sub> GASIFICATION PROCESS WITH RECIRCULATION

The balance of the element hydrogen in a resulting equilibrium reactive mixture of the amount of  $n=1$  kmol is

$$(H) = (2x_{\text{H}_2} + 2x_{\text{H}_2\text{O}} + 4x_{\text{CH}_4}) = n^{\text{conv}}(2x_{\text{H}_2}^{\text{conv}} + 2x_{\text{H}_2\text{O}}^{\text{conv}} + 4x_{\text{CH}_4}^{\text{conv}})$$

and the element oxygen balance

$$(O) = (x_{\text{CO}} + 2x_{\text{CO}_2} + x_{\text{H}_2\text{O}}) = n^{\text{conv}}(x_{\text{CO}}^{\text{conv}} + 2x_{\text{CO}_2}^{\text{conv}} + 2x_{\text{O}_2}^{\text{conv}} + x_{\text{H}_2\text{O}}^{\text{conv}})$$

Just like in the case of a gasifying process without recirculation, the appropriate characteristic parameter  $A$  can be defined and calculated

$$A = \frac{(H)}{(O)} = \frac{2x_{\text{H}_2}^{\text{conv}} + 2x_{\text{H}_2\text{O}}^{\text{conv}} + 4x_{\text{CH}_4}^{\text{conv}}}{x_{\text{CO}}^{\text{conv}} + 2x_{\text{CO}_2}^{\text{conv}} + 2x_{\text{O}_2}^{\text{conv}} + x_{\text{H}_2\text{O}}^{\text{conv}}} = \frac{2x_{\text{H}_2} + 2x_{\text{H}_2\text{O}} + 4x_{\text{CH}_4}}{x_{\text{CO}} + 2x_{\text{CO}_2} + x_{\text{H}_2\text{O}}}$$

After rearranging, it becomes

$$x_{\text{CH}_4} = \frac{A}{4}x_{\text{CO}} + \frac{A}{2}x_{\text{CO}_2} - \frac{1}{2}x_{\text{H}_2} + \frac{A-2}{4}x_{\text{H}_2\text{O}}$$

and taking into account the normalizing relation

$$x_{\text{CO}} + x_{\text{CO}_2} + x_{\text{H}_2} + x_{\text{H}_2\text{O}} + x_{\text{CH}_4} = 1$$

the formula

$$x_{\text{CO}} + x_{\text{H}_2} + x_{\text{CO}_2} + x_{\text{H}_2\text{O}} + \frac{A}{4}x_{\text{CO}} + \frac{A}{2}x_{\text{CO}_2} - \frac{1}{2}x_{\text{H}_2} + \frac{A-2}{4}x_{\text{H}_2\text{O}} - 1 = 0 \quad (09)$$

where from

$$A = \frac{1 - x_{\text{CO}} - \frac{1}{2}x_{\text{H}_2} - x_{\text{CO}_2} - \frac{1}{2}x_{\text{H}_2\text{O}}}{\frac{1}{4}x_{\text{CO}} + \frac{1}{2}x_{\text{CO}_2} + \frac{1}{4}x_{\text{H}_2\text{O}}} = 2 \frac{2 - 2x_{\text{CO}} - x_{\text{H}_2} - 2x_{\text{CO}_2} - x_{\text{H}_2\text{O}}}{x_{\text{CO}} + 2x_{\text{CO}_2} + x_{\text{H}_2\text{O}}}$$

Substituting equation for appropriate chemical equilibrium constants Eq. (01) – (02) and Eq.(09) yields

$$A = 2 \frac{2 - 2x_{\text{CO}} - x_{\text{H}_2} - 2 \frac{x_{\text{CO}}^2}{K_{(1)}} p - \frac{x_{\text{CO}}x_{\text{H}_2}}{K_{(2)}} p}{x_{\text{CO}} + 2 \frac{x_{\text{CO}}^2}{K_{(1)}} + \frac{x_{\text{CO}}x_{\text{H}_2}}{K_{(2)}} p} \quad (10)$$

This parameter is analog to the  $A$ -parameter derived for the nitrogen-free gasifying process without recirculation. It is easy to solve. The only trouble is that the chemical reactions taken into account do depend very strongly on process pressure (because of their non-equimolarity) and the approximations for  $K_{(1)}$ ,  $K_{(2)}$  (and eventually  $K_{(3)}$ ) are usually given for the chemical normal pressure  $p^0=1$  ata.

The special advantage of the  $A$ -parameter as a ratio of the element hydrogen and element oxygen amounts in a converting medium is the fact, it does not contain the equilibrium constant  $K_{(3)}$ , which means the methane formation in a process. The methanization, even by high hydrogen concentrations is not fully known chemical process, e.g. [11], and hitherto trials did not give any applicable method for producing the so-called strong synthetic gas.

From Eq. (10) three characteristic cases of the converting medium composition can be formulated:

$A \rightarrow 0$  converting medium is pure oxygen ( $x_{\text{O}_2}^{\text{conv}} = 1$ )

$A = 0$	converting medium is water steam	$(x_{\text{H}_2\text{O}}^{\text{conv}} = 1)$
$A \rightarrow \infty$	converting medium is pure hydrogen	$(x_{\text{H}_2}^{\text{conv}} = 1)$

## 5 CONCLUSION

The analytical method of selecting main parameters of the coke (carbon C) gasifying process has been presented in detail. For needed amounts of hydrogen and carbon monoxide in a resulting synthetic gas, the concentration of oxygen and water steam in a gasifying medium can be determined for chosen  $T$  and  $p$ . In fact the method is valid only for the case no atmospheric nitrogen is present. The last case can be proceeded using the MOLLIER–HOFFMANN diagram, which, however, cannot be applied to the nitrogen-free gasification. The pressure in Eq. (08) and Eq. (10) is in fact a dimensionless quantity  $p/p_0$  with  $p_0$  as a reference pressure (usually 1 ata). The equilibrium composition of the resulting gas (reactive mixture) is presented as a function of process pressure (not such often, e.g. [4]), process temperature (very often, e.g. [1] – [4], [9]), but also the parameter  $A$ , characterizing the gasifying agent (e.g. [11]). The main concentration parameter  $a$  determined using the presented above analytical method is very convenient as a start value in iteration procedure of computing gasifying processes, discussed in detail e.g. in [4]. To the problem also [12] – [13].

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