

## Gasification of Coke-Plant Wastes

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### **Abstract**

The main directions of processing and utilization of coke-plant wastes are considered. The method of gasification of coke-plant wastes on the example of heavy coal tars, which can be used to generate producer gas, was studied. The influence of equipment temperature and oxidant consumption on the yield of such components as:  $H_2$ ,  $O_2$ ,  $N_2$ ,  $CH_4$ ,  $CO$ ,  $CO_2$ ,  $C_2H_4$  and  $C_2H_6$  is established. Mathematical dependences of formation of separate components of gas which can be applied at forecasting of structure of producer gas depending on conditions out of experiment are developed. The use of an electrical converter allows to significantly change of gaseous products of processing with increasing content of combustible components.

**Keywords:** *Heavy coal tars; Lignite; Walnut shell; Gasification; Producer gas.*

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## **1. Introduction**

Modern coke production includes dozens of technological processes [1-6] as a result of which, unfortunately, along with the main products, a large number of by-products and wastes are formed, such as: heavy coal tars from the department of coal-tar dehydration, heavy coal tars from the tar-condensation department, acidic tars, mixture of tar and oil from biochemical wastewater treatment, residues from the cleaning of coke-plant equipment and tar storage tanks [7-9]. For many years, the main way to preserve coke-plant wastes was to store it in dumps, resulting in the formation of "resin lakes" [10-11].

Currently, many years of searching for ways to dispose of this wastes [12-25] went to the following main areas, namely:

- use in coking batch;
- use as a component of the road surface;
- development of anticorrosive protective materials on their basis;
- obtaining various furnace and boiler fuels;
- their use as a binder in the manufacture of fuel briquettes.

However, the proposed methods have a number of disadvantages, for example, road viscos materials obtained using such products, in their technological properties, although they meet the existing requirements, but due to toxicity they can be used only outside the settlements.

In work [26] the results of research of coke-plant wastes for the presence of polyaromatic hydrocarbons and benzo[a]pyrene were published. Of course, the combustion of such fuels will produce energy, but from an environmental point of view, this solution is not ideal, primarily due to the formation of additional amounts of sulfur dioxide and of nitrogen compared with petroleum fuel oil.

Fuel briquettes obtained using coke-plant wastes as a binder do not differ much from furnace and boiler fuels both in terms of their heat of combustion and in terms of their impact on the environment. In work [27], the results of studies of emissions of a wide range of pollutants (dust, oxides of sulfur, nitrogen and carbon, 3,4-benzo[a]pyrene, ammonia, hydrogen

cyanide, phenol, hydrogen sulfide) during the combustion of coal briquettes of different composition in the layer furnace on the bench boiler installation.

The addition of coke-plant wastes to the batch is carried out in the form of water-oil emulsions, which increases the moisture content of the batch, despite its small share of the loading weight, and significantly declines the sanitary conditions of workers in the coal preparation plant [8, 28-29].

In our opinion, the most attractive way to use of this waste is its gasification. The Department of Oil, Gas and Solid Fuel Processing Technologies of National Technical University "Kharkiv polytechnic Institute" has developed a method of utilization of viscous organic coke-plant wastes to obtain generator gas, which includes low-temperature gasification of a mixture of waste coke production with a solid carrier (namely, lignite or walnut shell) at a temperature of 400–500°C with the subsequent passage of the formed vapor-gas mixture through a layer of hot coke (800–1000°C) [30-31].

## 2. Experimental part

### 2.1. Raw materials

The raw materials for the research were lignite (L) of the Alexandriya field (Ukraine), walnut shell (WS) and heavy coal tars (HCT) of "ZAPORIZHKOKS", the characteristics of which are presented in Table 1. According to previous studies, the most optimal mixtures for gasification were the following ratios: 90 % (WL) + 10% (HCT) and 90% (L) +10 % (HCT). Granulometric composition of experimental samples 3–5 mm.

Table 1.Characteristics of the raw materials

Index		The raw materials					Method
		L	WS	HCT	90 % (WS) + 10 % (HCT)	90 % (L) + 10 % (HCT)	
Proximate analysis %	$W_t^r$	9,4	7,6	10,2	7,86	9,48	ISO 589:2008 [32]
	$A^d$	39,8	2,3	3,9	2,46	36,21	ISO 1171-97 [33]
	$S_t^d$	2,74	0,09	0,03	0,08	2,47	ISO 334-92 [34]
	$V^{daf}$	71,0	79,9	34,9	75,40	67,39	ISO 562:2010 [35]
Ultimate analysis %	$C^{daf}$	67,56	52,52	94,09	56,68	70,21	ISO 625-96 [36]
	$H^{daf}$	6,95	5,98	4,28	5,81	6,68	ISO 625-96 [37]
	$N^{daf}$	0,89	0,19	1,29	0,30	0,93	ISO 333-83 [38]
	$O^{daf}$	21,86	41,22	0,31	37,13	19,71	ISO 1994-76 [39]

### 2.2. Experimental equipment

The gasification of prepared materials was carried out on a two-stage laboratory installation for processing of viscous coke-plant wastes, the block diagram of which is presented in Fig. 1.

Principle of operation: the prepared material is loaded into a perforated vessel 1, which was placed at the bottom in the furnace 2, heated to a temperature of 400–500°C, fixing and temperature control of which is maintained through the control unit furnace 7. There, the oxidant-supply unit 8 serves oxidant-air. The products formed during destruction, representing a vapor and gaseous mixture, are sent for conversion into an electrical converter 3. The conversion is carried out in its lower part, heated to high temperatures of 800 and 1000°C, fixing and temperature control is maintained through the control unit 6.

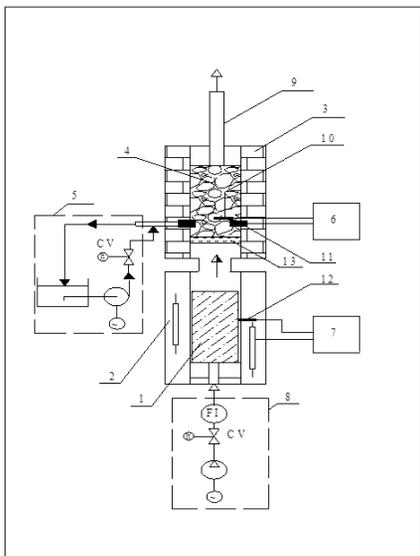


Fig. 1 Technological scheme of two-stage laboratory equipment for processing of viscous coke-plant wastes

1 – perforated vessel; 2 – furnace; 3 – electrical converter; 4 – carbon packing; 5 – water-cooling system; 6 – control unit of the electrical converter; 7 – control unit of furnace; 8 – oxidant-supply unit; 9 – tube for sampling gas produced; 10 – Chromel-Alumel thermocouple; 11 – graphite electrodes; 12 – Chromel-Copel thermocouple; 13 – grid; CV – control valve; FI – flow meter

The electrical converter 3 is a cylindrical vertical furnace. The walls of the electrical converter were made of corundum, covered with a metal casing. There is a pair of graphite electrodes 11, which creates a heating zone. Each electrode has water cooling 5. Inside the electrical converter in height on the grad 13 is loaded carbon packing 4, which used coal coke. Electric heating is carried out by applying an electric current to the graphite electrodes. During the experiment, the carbon packing (coal coke) is in a hot state.

The temperature is regulated by the supply of current to the electrodes. Microarc discharges are formed in the places of filling of the carbon packing (Fig. 2), the temperature of which can reach 2500°C [39]. The use of a quartz cover for a thermocouple allowed to engrave the above phenomena in the photo. In the upper part of the electrical converter there is a tube 9 for sampling the formed gas.

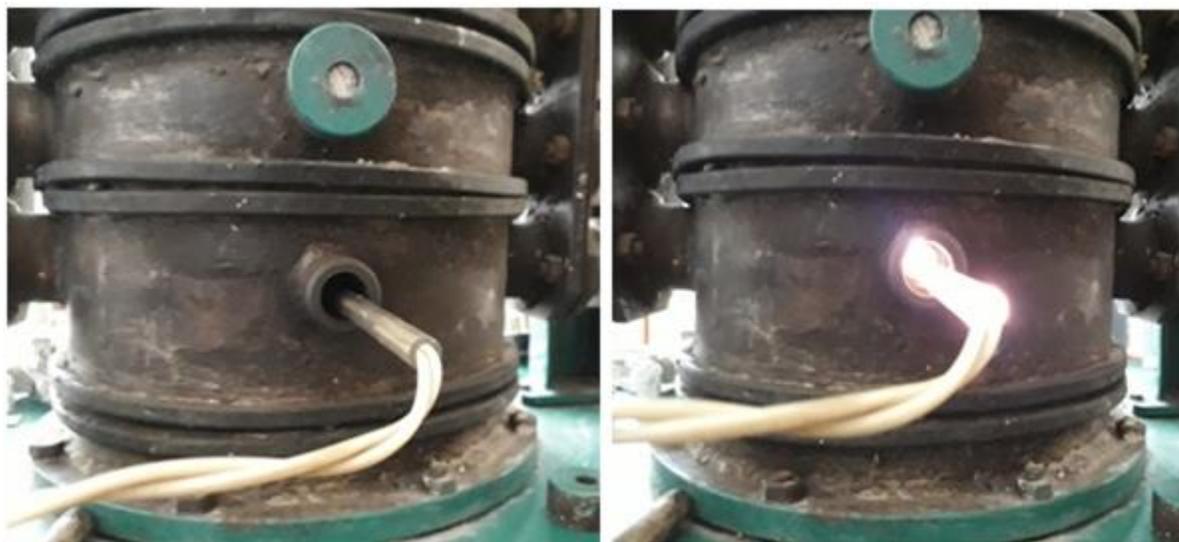


Fig. 2. Photo of the electrical converter: a) not in working condition; b) in working condition

### 2.3. Experimental procedure

Conditions for the experiment: a portion weighing  $20 \pm 0.0001$  g was introduced into the preheated furnace and kept for 40 minutes. When gasification WS + HCT furnace temperature was 400 or 500°C; when gasifying L + HCT furnace temperature was 500°C. The oxidant flow rate (V) fluctuated for WS + HCT – 0.0005; 0.001 or 0.002 m<sup>3</sup>/min; for L + HCT – 0.002;

0.003 or 0.004 m<sup>3</sup>/min. The formed vapor and gaseous compounds entered the electrical converter, the temperature of which was 800 or 1000°C for WS + HCT and L + HCT.

## 2.4. Method for characterization

Determination of gas composition was performed on a gas chromatograph "Crystal-2000" (columns with carriers "Zeolite CaA" and "Polysorb-1", helium was used as a carrier gas) by gas-adsorption separation of components and determination of the volume fraction of components by absolute calibration [40].

The principle of operation of the gas chromatograph "Crystal-2000" (Fig. 3) is as follows: the starting material is dissolved in a liquid or gaseous carrier and fed to the sorbent, which uses a solid porous body or liquid film applied to it. The sorbent together with the carrier move along the stationary phase and interact with it at different speeds. Due to physical and physicochemical processes (for example, adsorption), the components of the mixture are retained by different layers of the sorbent or leave the chromatograph together with the mobile phase. As a result, the sample is divided into components, and analysis of the speed of their exit from the device allows to establish the exact qualitative and quantitative composition.

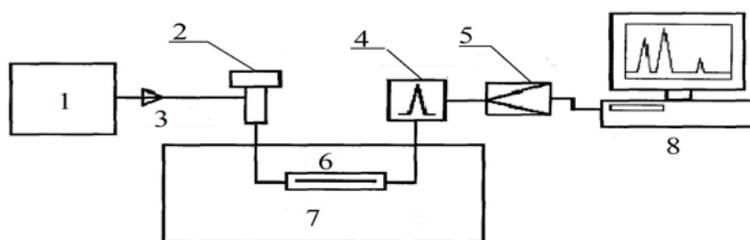


Fig. 3 Block diagram of the gas chromatograph "Crystal-2000"

1- unit for electronic control of gas flows; 2 - input device; 3 - carrier gas; 4 - detector; 5 - amplifier and analog-to-digital converter; 6 - column; 7 - thermostating device; 8 - recording device.

The columns in the chromatograph are installed in parallel. The test gas is introduced into the first column "CaA" and the first thermal conductivity detector, the dosing valve is switched to take a new gas sample and introduced into the second column with "Polysorb-1" and in the second thermal conductivity detector.

Conditions of the experiment: the temperature of the column thermostat 50°C; evaporator temperature 50°C; detector temperature 50°C; consumption of carrier gas 60 cm<sup>3</sup>/min; the volume of the injected sample is 0.125 cm<sup>3</sup>.

## 3. Results and discussion

Depending on the conditions of the experiment, the quantitative content of any component varies, because its formation is influenced by three factors: the temperature of the furnace ( $t_f$ ), the temperature of the electrical converter ( $t_{ec}$ ) and the consumption of oxidant ( $V$ ).

From the obtained data for each component, a set of linear regression was developed to determine the calculated value or predict the yield of an individual component using the constructed under given conditions, which are presented in Table 2 for WS+HCT and L+HCT.

From the mathematical dependences given in Table 2 on each component of gas received at processing of L+HCT it is visible that their maintenance depends on temperature of the electrical converter and an oxidant expense, unlike gas received at processing WS+HCT which also depends on temperature of the pyrolysis furnace. This is due to the fact that in these studies, as mentioned above, when gasifying WS + HCT, the furnace temperature was 400 or 500°C; during gasification of L+HCT the furnace temperature was 500°C, i.e. did not change.

From the obtained mathematical dependences it is seen that in the gasification of WS + HCT the formation of important components of the producer gas, such as CO and H<sub>2</sub>, is positively affected by increasing the temperature of the electrical converter, while other components such as CH<sub>4</sub>, C<sub>2</sub>H<sub>4</sub> and C<sub>2</sub>H<sub>6</sub> are affected by this factor negatively. As the temperature of the furnace increases, the H<sub>2</sub> content also increases, but the CO content decreases. This

fact must be taken into account when determining the priority of the consumer to the finished product. The increase in oxidant consumption negatively affects the formation of combustible components, except C<sub>2</sub>H<sub>4</sub>.

Table 2. Mathematical dependence of the formation of components during gasification WS + HCT and L + HCT and their statistical evaluation

Raw material	№	Type of equation	statistical evaluation		
			r	R <sup>2</sup>	σ
WS + HCT	(1)	$O_2 = 8.142042 - 157.535 \cdot V - 0.00016 \cdot t_f - 0.00629 \cdot t_{ec}$	0.807	0.622	0.821
	(2)	$N_2 = 38.03371 + 1181.635 \cdot V + 0.003526 \cdot t_f - 0.00157 \cdot t_{ec}$	0.654	0.481	1.117
	(3)	$H_2 = -3.50213 - 1572.36 \cdot V + 0.021153 \cdot t_f + 0.021238 \cdot t_{ec}$	0.674	0.454	3.799
	(4)	$CH_4 = 3.299867 - 16.5925 \cdot V - 0.00141 \cdot t_f - 0.00141 \cdot t_{ec}$	0.705	0.497	0.228
	(5)	$CO = 5.957965 - 1240.08 \cdot V - 0.0074 \cdot t_f + 0.026371 \cdot t_{ec}$	0.755	0.570	3.737
	(6)	$CO_2 = 4.651382 - 125.972 \cdot V - 0.00089 \cdot t_f - 0.00167 \cdot t_{ec}$	0.363	0.132	0.533
	(7)	$C_2H_4 = 18.49132 + 89.79153 \cdot V - 0.01123 \cdot t_f - 0.00671 \cdot t_{ec}$	0.644	0.4153	1.377
	(8)	$C_2H_6 = .247607 - 31.8803 \cdot V - 0.00103 \cdot t_f - 0.00029 \cdot t_{ec}$	0.457	0.209	0.141
L + HCT	(9)	$O_2 = 9.668586 - 654.228 \cdot V - 0.00594 \cdot t_{ec}$	0.929	0.862	0.913
	(10)	$N_2 = 39.28656 + 1596.1 \cdot V + 0.011496 \cdot t_{ec}$	0.906	0.820	2.051
	(11)	$H_2 = 0.35735 + 670.7558 \cdot V + 0.011844 \cdot t_{ec}$	0.847	0.717	1.609
	(12)	$CH_4 = -0.43369 + 123.2161 \cdot V + 0.000503447 \cdot t_{ec}$	0.882	0.777	0.138
	(13)	$CO = -0.21379 + 300.6267 \cdot V + 0.022571 \cdot t_{ec}$	0.665	0.442	3.530
	(14)	$CO_2 = 2.732742 + 1.75 \cdot V - 0.00136 \cdot t_{ec}$	0.775	0.600	0.182
	(15)	$C_2H_4 = 5.904396 + 543.0542 \cdot V - 0.00169 \cdot t_{ec}$	0.768	0.591	0.671
	(16)	$C_2H_6 = 0.032338 + 95.3 \cdot V - 3.9 \cdot 10^{-5} \cdot t_{ec}$	0.906	0.820	0.094

During the gasification of L + HCT, the increase in the temperature of the electrical converter also has a positive effect not only on the formation of CO and H<sub>2</sub>, but also on the content of CH<sub>4</sub>, except C<sub>2</sub>H<sub>4</sub> and C<sub>2</sub>H<sub>6</sub>. The increase in oxidant consumption has a positive effect on the formation of combustible components. In both cases, the logical growth of N<sub>2</sub> with the growth of the oxidant.

Using equations (1) – (8) for WS + HCT and (9) – (16) for L + HCT, the theoretical yield of component was calculated under the conditions of a real experiment. The graphical dependences of the actual on the calculated value of Fig. 4 a–h for the gas obtained during the processing of WS + HCT and Fig. 5 a–h for the gas obtained during the processing of L + HCT are constructed. Coordinate axes for each component: abscissas – the quantitative content of the component obtained as a result of the experiment; ordinates – the quantitative content of the component determined by the corresponding equation, i.e. the calculation value.

From those shown in Fig. 4 a–h and Fig. 5 a–h graphical dependences of the actual value of the component on the calculated under the same conditions show that the dependencies are linear. The values of the correlation and determination coefficients indicate the reliability of the research.

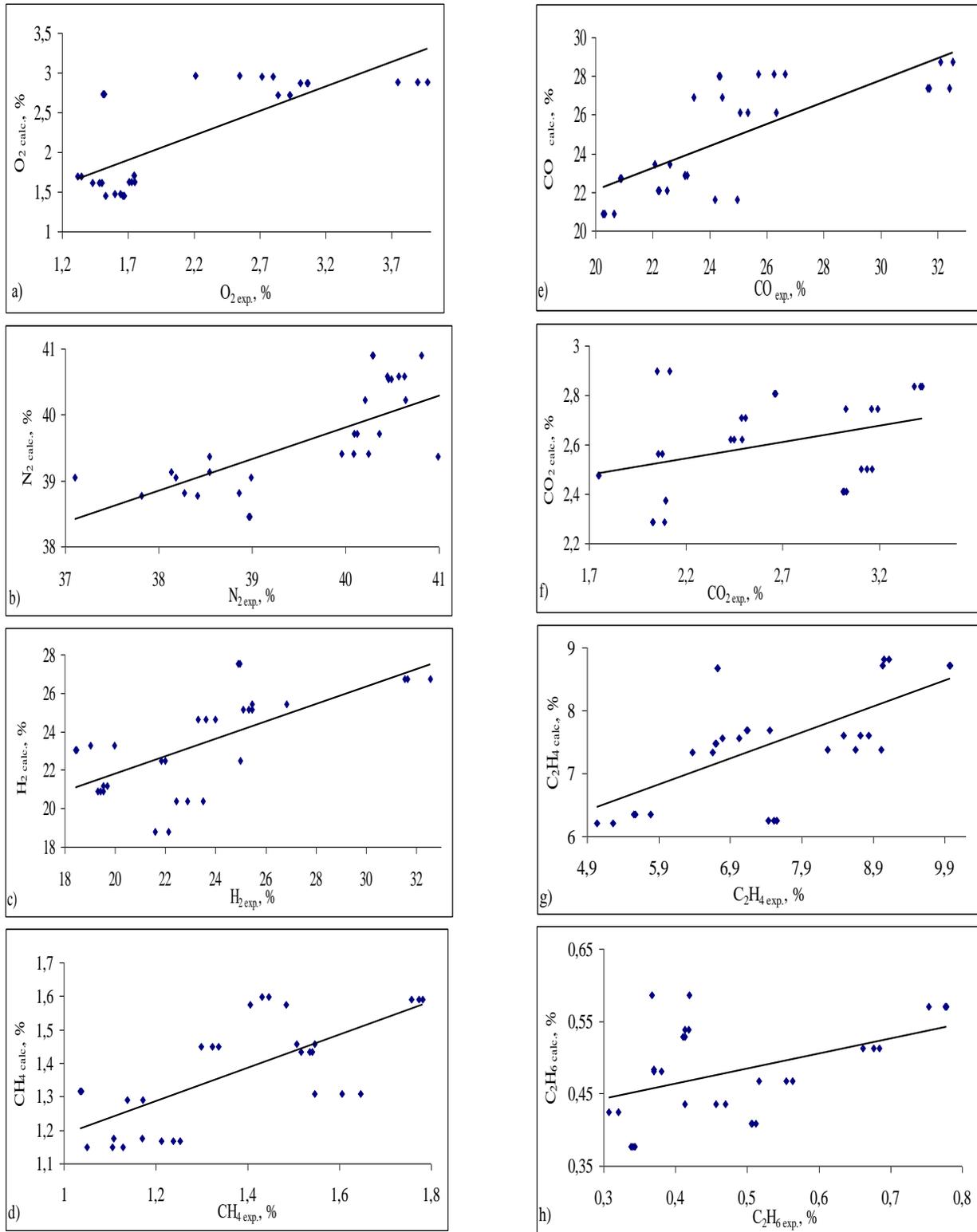


Fig. 4 a-h Graphic dependence of the actual value of the component on the calculated under the same conditions of gasification WS + HCT

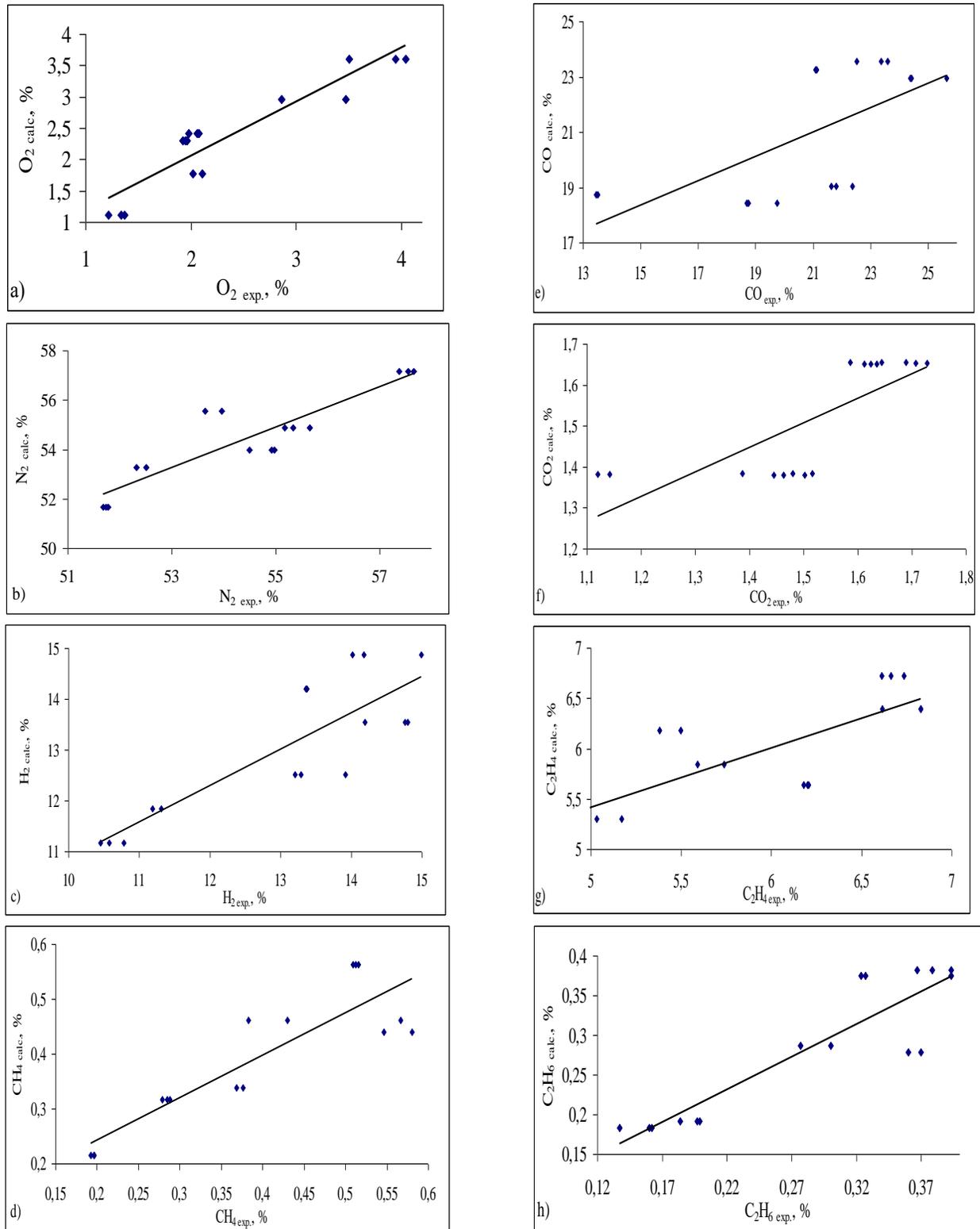


Fig. 5 a-h Graphic dependence of the actual value of the component on the calculated under the same conditions of gasification L + HCT

#### 4. Conclusions

Given the above materials and research results, we can conclude the following:

- 1) the developed method of gasification of coke-plant wastes on the example of heavy coal tars can be used to obtain generator gas;
- 2) it is established how the temperature of the equipment and the consumption of oxidant affect the yield of such components as:  $H_2$ ,  $O_2$ ,  $N_2$ ,  $CH_4$ ,  $CO$ ,  $CO_2$ ,  $C_2H_4$  and  $C_2H_6$ ;
- 3) the developed mathematical dependences of formation of separate components of generator gas can be applied at forecasting of structure of generator gas depending on conditions of carrying out experiment;
- 4) graphical dependences of the actual value of the component on the calculated under the same conditions of gasification  $WS + HCT$  and  $L + HCT$  are linear. The values of the correlation and determination coefficients indicate the reliability of the research.

#### Symbols

$L$	<i>lignite of the Alexandriya field (Ukraine);</i>
$WS$	<i>walnut shell;</i>
$HCT$	<i>heavy coal tars;</i>
$t_f$	<i>the temperature of the furnace, °C;</i>
$t_{ec}$	<i>the temperature of the electrical converter °C;</i>
$V$	<i>and the consumption of oxidant, m<sup>3</sup>/min.</i>
$R^2$	<i>determination coefficient;</i>
$r$	<i>correlation coefficient;</i>
$\sigma$	<i>standard deviation.</i>

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