

# СУДОВЫЕ ЭНЕРГЕТИЧЕСКИЕ УСТАНОВКИ, СИСТЕМЫ И УСТРОЙСТВА

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## THE CATALYTIC NEUTRALIZATION OF EXHAUST GASES IN SHIP POWER PLANTS

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*Since 1 January 2020 all world transport, according to the decision of the International Maritime Organization, transfers to the low-sulfur fuel (no more than 0.5% of mass S-compounds in marine fuel). From such a transition, the decision of the International Maritime Organization exempts ships which are equipped with systems for the purification of exhaust gases of ship power plants from sulfur compounds, as well as those vessels that conduct research on the development of effective methods of purification from sulfur compounds. In this article, a catalytic method for the purification of exhaust gases from ship power plants is suggested.*

*It should be noted, that the transition to low-sulfur fuel can only reduce emissions of sulfur and / or sulfuric anhydride, while reducing the damage to the air basin, and the cost of low-sulfur fuel is 2-5 times higher than the cost of conventional marine fuel, in which the content of sulfur compounds does not exceed 3,5% mass.*

*For the neutralization of exhaust gases of car engines, the catalysts which contain precious metals and their compounds – platinum, palladium, rhodium, ruthenium, rare earth elements are used. The aim of this work is to create the catalysts for neutralization of toxic substances which are contained in exhaust gases of ship power plants that do not contain precious metals and their compounds, but the catalytic activity and duration of action is not inferior to catalysts, which are based on use of precious metals. To reduce the amount of work on the selection of effective and stable catalysts, it is suggested to use only carbon monoxide instead of studying a wide nomenclature of harmful toxic substances contained in the exhaust gases of ship power plants. A new technology for the preparation of catalysts for the neutralization of exhaust gases of power plants has been developed in our study. This technology is based on the use of a cheap raw materials source with a large stock ratio, as well as secondary material resources and technological systems wastes. For the real vessel OXL “Samurai” in the engine room technological calculations were performed to optimally locate the catalytic neutralization installation for the purification of toxic gases of ship power plant and heat exchange equipment for the heat utilization of exhaust gases. The technology fully meets the requirements of resource-saving and environmental protection. The management of the integrated technology is based on automation of the process using modern software.*

*Keywords: non-precious metals, exhaust gases neutralization, ship power plant, carbon monoxide, heat utilization, new purification installation, exhaust gases, catalytic cleaning, International Maritime Organization, toxic substances.*

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## КАТАЛИТИЧЕСКАЯ ОЧИСТКА ОТРАБОТАННЫХ ГАЗОВ В СУДОВЫХ ЭНЕРГЕТИЧЕСКИХ УСТАНОВКАХ

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*С 1 января 2020 г. весь мировой транспорт, согласно решению Международной морской организации, переходит на низкосернистое топливо (не более 0,5% масс. S-соединений в судовом топливе). От такого перехода, по решению Международной морской организации, освобождаются суда, укомплектованные системами очистки отработанных газов судовых энергетических установок от сернистых соединений, а также научно-исследовательские суда, занимающиеся разработкой систем очистки отработанных газов судовых энергетических установок от сернистых соединений. В настоящей статье предлагается каталитический способ очистки отработанных газов судовых энергетических установок. Отмечается, что переход на низкосернистое топливо позволяет снизить только эмиссию сернистого и / или серного ангидрида, при этом снижается ущерб воздушного бассейна, а стоимость низкосернистого топлива в 2–5 раз выше стоимости обычного судового топлива, в котором содержание серосоединений не превышает 3,5 % масс. Для нейтрализации отработанных газов двигателей транспортных средств используют катализаторы, содержащие драгоценные металлы и их соединения: платину, палладий, родий, рутений, а также редкоземельные элементы.*

*Целью данной работы является создание катализаторов нейтрализации токсических веществ и соединений, не содержащих драгоценных металлов и их соединений, а по каталитической активности и длительности действия, не уступающих катализаторам на основе драгоценных металлов. Для снижения объема работ по подбору эффективных и стабильных катализаторов предложено вместо исследований широкой номенклатуры вредных токсических веществ, содержащихся в отработанных газах судовых энергетических установок, использовать только монооксид углерода. Разработана новая технология приготовления катализаторов нейтрализации отработанных газов энергосиловых установок. Технология основана на использовании исходного дешевого сырья с большой кратностью запаса, а также вторичных материальных ресурсов и отходов техногенных систем. Для реального судна OXL «Samurai» в машино-котельном отделении были выполнены технологические расчеты по оптимальному размещению каталитической установки нейтрализации отработанных газов судовой энергетической установки и теплообменного оборудования для утилизации теплоты отработанных газов. Технология полностью отвечает требованиям ресурсосбережения и защиты окружающей среды. Управление комплексной технологией основано на автоматизации процесса с использованием современного программного обеспечения.*

*Ключевые слова: недорогие металлы, очистка отработанных газов, судовая энергетическая установка, монооксид углерода, утилизация теплоты, новая система очистки, отработанные газы, каталитическая очистка, Международная морская организация, токсические вещества.*

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## **Introduction**

Nowadays, the role of Industrial Ecology which specializes in the degree of harm assessment caused by the industrialization and the intensive growth in mankind's requirements is considerably increasing. Undoubtedly, transport is known as the main factor of air pollution [1]–[2]. In Cosmopolitan cities the contribution of air pollution caused by transport reaches 85–90%. Over the past 50 years the World Merchant Fleet has been increased more than in 25 times. Moreover, the role of maritime, railway and air transport has been highly intensified. Although the impact of air and maritime transport on the environmental pollution and the increase in “greenhouse effect” is considerably high (currently it is more than 10%) and has a growing tendency, marine and air transport industries have not been included into Outcomes of 2015 United Nations Climate Change Conference, Le Bourget in the suburbs of Paris, France.

The exhaust gases of ship power plants (further the EG SPP) are consisted of more than 200 harmful toxic components which are carcinogenic, mutagenic, irritating and poisonous by their nature, relate to the I–IV classes of dangerous impact on human health. Furthermore, besides chemical pollution, the EG SPP possess high enthalpy that leads to the thermal pollution of air basin. According to the all mentioned factors, from the Ecology safety and Environmental Protection perspective the concern of the safe EG SPP neutralization and purification remains relevant and highly-significant [3].

In order to reduce the “pressure” of man-made systems on the environment and the biosphere, recently engineering and technical means of environmental protection are being developed and improved.

The scientific basis for the creation of closed, low-waste, resource-saving and waste-free technological processes are evolving.

In the research [4] the requirements of the International Normative Legal Acts to the permissible level of toxicity of exhaust gases of car engines (the EG CE) and the EG SPP are analyzed and generalized. As a result, it is established that the requirements to the level of toxicity of the EG CE are much 'striker' in comparison with the requirements to the level of toxicity of the EG SPP. However, these differences in the requirements for the level of toxicity of the EG CE and the EG SPP are particularly paradoxical against the fact, that the macro-composition, the physiological effects on humans and the biosphere, as well as their impact on the quality of the environment are resulted equally both by the EG SPP and the EG CE. These emissions, regardless of their origin, are almost congruent and in both cases are very dangerous.

In the research [5] to reduce the emission of 'greenhouse' gas components from ships, operating in seaport areas and populated areas adjacent to the ports, up to 2030, three scenarios for environmental protection are suggested, namely:

1. Development and application of alternative marine fuels.
2. Development of new designs of ships and ship power plants.
3. Analysis and development of new technical solutions, and as a result, their implementation.

In order to reduce the EG SPP content of harmful components to the maximum permissible standards, particularly to EURO-VI requirements [6], the following methods of the EG SPP purification are used:

1. Physical:
  - low temperature condensation;
  - membrane separation.
2. Physicochemical:
  - absorption;
  - adsorption;
  - high temperature – thermal oxidation, pyrolysis;
  - chemical – conversion, destruction;
  - catalytic.
3. Biological:
  - biochemical oxidation by microorganisms, anaerobic decomposition.
4. Combined.

This research work includes:

- the development of a simple, unified method of catalytic activity of catalysts determination;
- the study of catalytic activity of oxidation catalysts;
- the development of the catalyst preparation technology for the EG SPP neutralization without expensive precious metals or their compounds in its structure.

Catalytic reactions can be divided into three classes:

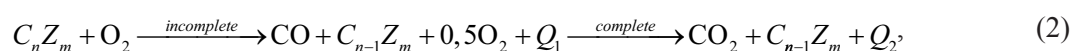
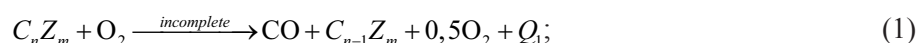
1. Catalytic oxidation.
2. Catalytic reduction.
3. Catalytic conjugated chemical reactions.

In this research work the cluster of catalytic oxidation of the harmful, toxic compounds which contain in the EG SPP is investigated.

The catalytic oxidation reaction can be divided into two subclasses:

1. Incomplete catalytic oxidation.
2. Complete catalytic oxidation.

For instance, any carbon-containing organic compounds may be subjected to catalytic oxidation according to two routes (subclasses) according to the following equations:



where  $Z_m$  — can include hydrogen, oxygen, nitrogen, sulphur, chlorine, etc.

On the one hand, the second subclass – complete catalytic oxidation – equation (2) is more profitable from the perspective of energy, sanitary and environmental constituent parts. Moreover, the heat generated while the complete catalytic oxidation is more ( $Q_1+Q_2$ ) than the heat generated while the incomplete catalytic oxidation ( $Q_1$ ) by the amount of  $Q_2$  heat. On the other hand, carbon monoxide is a toxic compound while carbon dioxide is a neutral compound.

Due to the wide range of harmful toxic substances contained in the EG SPP studying the catalytic activity of catalysis for each of them individually is definitely a very time-consuming and costly procedure.

### Methods and materials

Based on the theoretical assumptions and practical results, the carbon monoxide is suggested by us as the main organic, carbonaceous substance for the catalytic activity of oxidation catalysts study. Moreover, carbon monoxide has the only one route of catalytic oxidation – complete catalytic oxidation subclass – with only carbon dioxide formation.

The experiments were conducted on the two oxide catalysts:  $C_1$  and  $C_2$  in Boreskov Institute of Catalysis as part of Siberian Branch of the Russian Academy of Sciences, fraction  $1 \times 1$  mm (table 1). The study of the activity of the catalyst was carried out in running unit (reactor of ideal displacement) with a fixed catalyst.

The temperature was controlled by thermocouple. The temperature measurement was ranging between 20-450 °C. The carbon monoxide content analysis was carried out by using the detector of the gas chromatograph at the inlet in the reactor apparatus and at the output from it. The CO concentration measurements were taken at least three times. The average value was considered as the measurement result.

Table 1

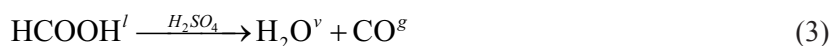
$C_1$  and  $C_2$  catalyst characteristics

The catalyst	Chemical composition, mass %	Specific surface area, m <sup>2</sup> /g
$C_1$ in the granule form	CuO – 10 Cr <sub>2</sub> O <sub>3</sub> – 10 $\gamma$ -Al <sub>2</sub> O <sub>3</sub> – 80	170
$C_2$ in the granule form	Co <sub>2</sub> O <sub>3</sub> – 11 Cr <sub>2</sub> O <sub>3</sub> – 6 Cr(+VI) no more than 0,05 SiO <sub>2</sub> – 82,95	110

Laboratory installation has a block-modular design and consists of five main parts:

1. The carbon monoxide receiving system.
2. The preparation and batching of initial reaction mixture system (a mixture of carbon monoxide and air).
3. The reactor unit.
4. The cooling gas system.
5. The system of condensation and separation – liquid phase – water and gaseous mixture – carbon monoxide, carbon dioxide, nitrogen and oxygen.

The carbon monoxide used in the experiment was received as a result of formic acid decomposition in the presence of a catalyst – Sulphur acid, at a temperature of 75-100 °C according to the following reaction:



where  $l$ ,  $v$ , and  $g$  are liquid, vapor and gas phases respectively.

## Results

Study of the catalyst activity in the carbon monoxide oxidation was carried out under the following circumstances:

- the catalyst grain was 1×1 mm;
- volumetric flow of the reaction mixture was 9 and 11 cm<sup>3</sup>/s;
- the carbon monoxide content in the air was 123 mg/m<sup>3</sup>;
- the excessive pressure in the reactor was 1000 mm Hg.

To study the catalytic activity, the catalyst sample was heated in a stream of air to a predetermined temperature beforehand, and then instead of air in the reaction mixture (a mixture of carbon monoxide and air) it was dosed out.

In the testing process we determined the degree of conversion of carbon monoxide (X, %) and the chemical reaction rate constant (K, mm<sup>3</sup>/s×mg).

The catalytic activity of the carbon monoxide oxidation on the catalyst C<sub>1</sub> was studied at the gas mixture flow of 9 and 11 cm<sup>3</sup>/s, at the temperature range 160–260 °C with the initial carbon monoxide concentration of 123 mg/m<sup>3</sup>.

The mass of catalyst which was loaded in the reactor was 90 mg, size of investigated catalyst fractions was 1×1 mm.

The degree of carbon monoxide conversion was changed from 17% to 72%. The results are shown in a table 2 and table 3.

Table 2

### Results of research at the 9 cm<sup>3</sup>/s gas mixture flow on the C<sub>1</sub> catalyst

№ пп.	T, °C	C <sub>co</sub> (input), mg/m <sup>3</sup>	C <sub>co</sub> (output), mg/m <sup>3</sup>	X <sub>co</sub> , %	K*, mm <sup>3</sup> /s×g	LnK
1	160	123	98	18	25,25	3,23
2	160	123	100	17	23,12	3,14
3	210	123	68	43	71,39	4,27
4	260	123	43	64	129,14	4,86

Table 3

### Results of research at the 11 cm<sup>3</sup>/s gas mixture flow on the C<sub>1</sub> catalyst

№ пп.	T, °C	C <sub>co</sub> (input), mg/m <sup>3</sup>	C <sub>co</sub> (output), mg/m <sup>3</sup>	X <sub>co</sub> , %	K*, mm <sup>3</sup> /s×g	LnK
1	160	123	89	28	33,76	3,56
2	160	123	90	26	31,78	3,46
3	210	123	60	51	74,83	4,32
4	260	123	34	72	132,88	4,89

The catalytic activity of the carbon monoxide oxidation on the catalyst C<sub>2</sub> was studied at the gas mixture flow of 9 and 11 cm<sup>3</sup>/s, at the temperature range 150–250 °C with the initial carbon monoxide concentration of 123 mg/m<sup>3</sup>. The loaded in the reactor catalyst mass was 1700 mg, size of investigated catalyst fractions was 1×1 mm.

The degree of carbon monoxide conversion was changed from 28% to 82%. The results are shown in table 4 and 5.

Table 4

### Results of research at the 9 cm<sup>3</sup>/s gas mixture flow on the C<sub>2</sub> catalyst

№ пп.	T, °C	C <sub>co</sub> (input), mg/m <sup>3</sup>	C <sub>co</sub> (output), mg/m <sup>3</sup>	X <sub>co</sub> , %	K*, mm <sup>3</sup> /s×g	LnK
1	150	123	89,00	27,78	2,18	0,78
2	150	123	87,29	29,17	2,31	0,84
3	200	123	42,79	65,28	7,10	1,96
4	250	123	27,39	77,78	10,10	2,31



Table 5

**Results of research at the 11 cm<sup>3</sup>/s gas mixture flow on the C<sub>2</sub> catalyst**

№ пп.	T, °C	Cco (input), mg/m <sup>3</sup>	C <sub>CO</sub> (output), mg/m <sup>3</sup>	X <sub>CO</sub> , %	K*, mm <sup>3</sup> /s×g	LnK
1	150	123	73,60	40,28	2,83	1,04
2	150	123	73,60	40,28	2,83	1,04
3	200	123	34,23	72,22	7,03	1,95
4	250	123	22,25	81,94	9,40	2,24

According to the study of the carbon monoxide oxidation on the C<sub>1</sub> and C<sub>2</sub> catalysts following points should be mentioned:

1. The oxidation of carbon monoxide on the C<sub>1</sub> and C<sub>2</sub> catalyst has a clear dependence. The temperature growth increases the CO degree of conversion; the rate constant of chemical oxidation in the reactor increases as well.

2. The C<sub>1</sub> catalyst is the most active one, because at equal temperatures the rate constant of a chemical reaction in the presence of C<sub>1</sub> catalyst was more, than on the C<sub>2</sub> catalyst.

3. The work on the C<sub>1</sub> catalyst allows to save catalyst mass which is loaded in the reactor, meanwhile operations under the same conditions on the C<sub>2</sub> catalyst require the 19 times more catalyst mass loaded in than the used on the C<sub>1</sub> catalyst.

Thus, based on the performed studies, the catalyst C<sub>1</sub> is recommended by us for catalytic purification of the EG SPP from all toxic components.

### Discussion

Based on the conducted research, it can be assumed that the suggested method allows taking measurements of the catalytic activity of different catalysts which are used in the heterogeneous catalytic oxidation of carbonaceous compound processes with acceptable accuracy. The error in the catalytic activity of the catalysts determination does not exceed more than 5% relative.

Therefore, studies on the catalytic activity of carbonaceous compound "C<sub>n</sub>Z<sub>m</sub>" oxidation catalysts, for instance, the study of carbon monoxide oxidation, allow us to determine the most effective oxidation catalyst fast and cost-effectively.

The activity of the catalyst should be determined for specifically selected carbonaceous, toxic substances with the aim of increasing the reliability of the selected catalyst, namely:

At the point of the optimal temperature which is obtained as a result of the carbon monoxide oxidation on the selected catalyst.

At the point of the real oxygen concentration in the EG SPP. It is advisable to specify the technological parameters which are obtained during the carbon monoxide oxidation on the selected catalyst.

In the developed European Union countries for exhaust gases neutralization and purification from toxic compounds which are produced by power plants and, particularly by cars, the expensive catalysts based on platinum, rhodium, rare earth elements and its compounds are used [4].

The purpose of the exhaust gases neutralization and purification from toxic compounds which are produced by power plants is to be in compliance with the health standards which are established by EURO-VI requirements to limit the level of toxicity of exhaust gases.

In the research [7] the catalysts for the carbon monoxide oxidation to carbon dioxide which contain expensive precious metals such as platinum, europium, cerium, titanium were obtained by the extraction-pyrolytic method. The active CO to CO<sub>2</sub> oxidation catalysts were obtained, particularly such catalytic systems as Pt + CeO<sub>x</sub> + Eu<sub>2</sub>O<sub>3</sub>/γ – Al<sub>2</sub>O<sub>3</sub>, Eu<sub>2</sub>O<sub>3</sub>/SiO<sub>2</sub>, Pt/SiO<sub>2</sub> and Pt/TiO<sub>2</sub>/Ti. The Pt/TiO<sub>2</sub>/Ti system was obtained by combination of plasma-electrolytic oxidation and extraction-pyrolytic methods.

In the research [8] the catalysts which are based on expensive materials such as Pd, Pt, Rh, La, Ce/γ – Al<sub>2</sub>O<sub>3</sub> in the three-way neutralization reaction: (1) carbon monoxide – (2) hydrocarbons – (3) nitrogen oxides were analyzed. It must be noted, that simultaneous neutralization of all three compounds is not possible, because the carbon monoxide and hydrocarbons are neutralized by the catalytic oxidation

reactions, whereas nitrogen oxides are neutralized by the catalytic reduction reactions. As far as we are concerned, various catalysts which are separated in time and space, placed in different reactor apparatus should be implemented for these neutralization processes.

In the research [9] the issue which relates to the hydrogen usage, namely its production, storage and transportation was analysed. The most common and widely-spread technologies of hydrogen production are considered, such as the hydrogen production in the process of a steam catalytic conversion and water electrolysis which require the usage of expensive catalysts based on platinum.

In the research [10] the technology of the precious metals extraction (the platinum, palladium, rhodium extraction) from the spent catalysts of EG SPP neutralization is suggested by means of manifold smelting in plasma-arc furnace with the use of self-generated steel manifold.

We have conducted a scientific-research work which was directed on expensive, precious, and scarce metals replacement in production of exhaust gases neutralization catalysts.

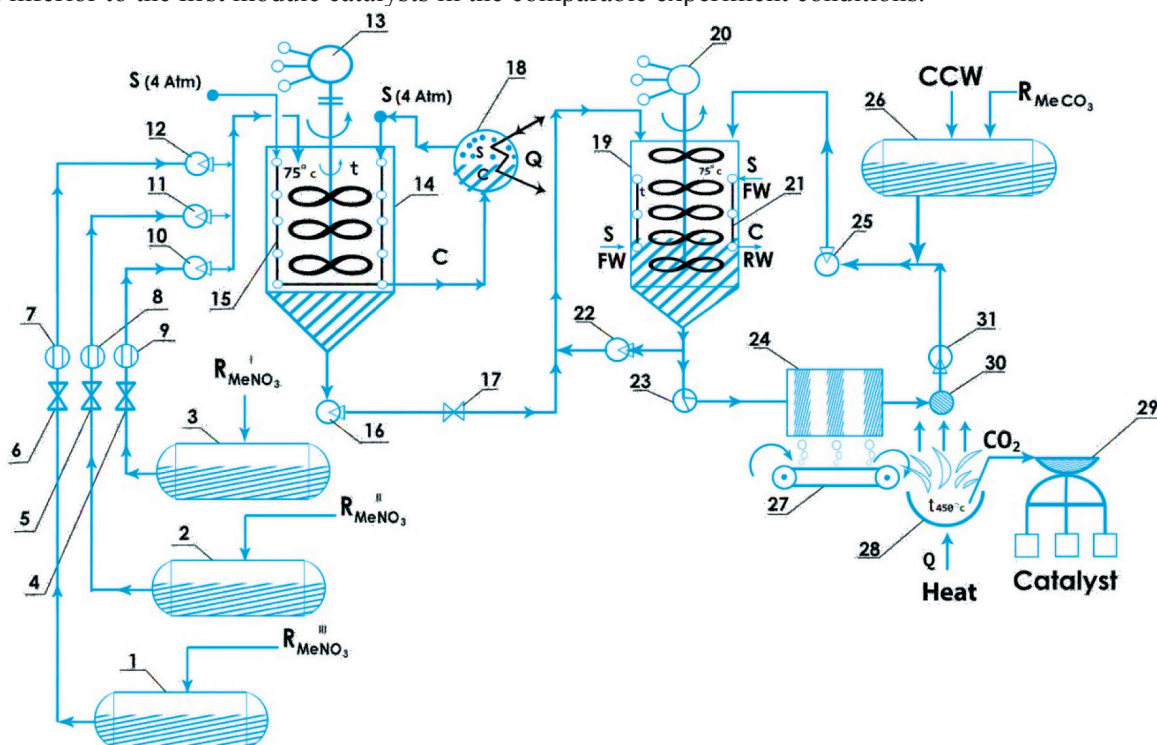
We have conditionally divided the technology of catalysts preparation into three modules according to the principle of the feedstock usage, namely:

1. The present technologies with the use of precious metals.
2. The classical technologies without the use of precious metals.
3. New integrated technologies without the use of precious metals.

The catalyst samples were prepared according to three modules and were tested on the flow type installation of ideal displacement in comparable conditions through the above mentioned method. Carbon monoxide was used as the initial toxic substance.

The most active catalysts were selected from the second and the third modules and were further compared with the first module catalysts.

As a result, the most active catalysts of the second and the third modules were identified which are not inferior to the first module catalysts in the comparable experiment conditions.



**Specification of appliance:** 1–3, 26 — catchment tanks; 4–6, 17 — shut-off and control valves; 7–12, 16, 22, 25, 31 — pumps; 13, 20 — an electric engine of three-phase alternating current; 14 — a mixer; 15, 21 — a steam and condensate manifold; 18 — a heat exchanger; 19 — reactor apparatus; 23 — a slurry pump; 24 — a frame filter; 27 — a conveyor belt; 28 — a calcining furnace; 29 — a tableting machine; 30 — a stabilizer; S — steam; Q — energy; C — condensate vapours of steam; CCW — chemically cleaned water; R — material flows; RW — returned water / wastewater; FW — fresh water; t — temperature

Fig. 1. The unit of catalysts preparation

The resource-conserving technology of catalyst preparation based on cheap materials, without expensive precious metals in their structure has been developed by us based on the third module catalyst formula. The scheme of this technology is shown on the figure 1.

The developed by us technology includes the following stages:

1. The preparation of reaction systems (positions 1–15).
2. The conversion of the reaction systems in the reactor apparatus – device mixer (positions 19–22, 25, 26).
3. The reactions precipitate dehydration, followed by drying and calcining (positions 24, 27, 28).
4. The tableting and / or granulation of the catalyst (position 29).

In the research [4] the complex technology of the warmth utilization from the EG SPP was suggested with the further exhaust gases treatment / neutralization from toxic compounds. The layout for the apparatus and the pipe manifold assembly installation which will utilize the excess heat and purify / neutralize the EG SPP in the engine department of the real ship OXL “Samurai” has been developed [11].

The financial support for the creation of an integrated installation is not significant. Moreover, the payback period of financial cost will take three years. The catalytic reactor apparatus of the toxic compounds oxidation which are contained in the EG SPP with the tube assemblies requires less space and the payback period of financial cost will take less than three years. Meanwhile, the passport-mode of ship power plant operation is not contravened in normal exploitation conditions and during forced modes of manoeuvring the vessel.

### Conclusion

The simple method of testing catalysts on the catalytic activity in the heterogeneous catalytic oxidation reactions of one-carbon compounds is suggested.

The correlation between the catalytic carbon monoxide oxidation and organic one-carbon compounds in catalytic oxidation reactions is established.

The analysis and generalization of scientific and technical publications in the field of the catalysts of the EG SPP neutralization, particularly the analysis of catalytic systems which contain expensive, precious metals in their structure are held.

The tests of the catalysts of the EG SPP neutralization which contain expensive, precious metals in their structure and which excludes expensive, precious metals in their structure under comparable experimental conditions are conducted. It is determined that catalytic activity of catalysts is almost comparable within the inaccuracy of experiments.

The resource-conserving technology of catalyst preparation based on cheap materials, without expensive precious metals in their structure has been suggested.

The layout for the apparatus and the pipe manifold assembly installation which will utilize the excess heat and purify / neutralize the EG SPP in the engine room of the real ship OXL “Samurai” has been developed.

The financial support for the creation of an integrated installation is not insignificant. Moreover, the payback period of financial cost will take three years.

### СПИСОК ЛИТЕРАТУРЫ

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