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ESTABLISHMENT OF THE PATTERNS OF NITROGEN INERTIZATION OF CYCLOPENTANE VAPOR-AIR MIXTURES

The object of research is the process of phlegmatization of the gaseous medium of vapor-air mixtures of cyclopentane when nitrogen is introduced into this mixture. The problem of using cyclopentane for the production of thermal insulation is to ensure explosion and fire safety, since the technology for creating foamed urethane thermal insulation includes the introduction of a liquid foaming agent, such as cyclopentane, into the reaction mass. It is characterized by a low boiling point and the creation of explosive mixtures with air. In this regard, the coordinates of the points of the phlegmatization curve were experimentally determined, the value of the lower concentration limit of flame propagation was obtained, which is 1.34% vol. of cyclopentane in a mixture with air, the upper concentration limit, which is 8.64% vol., the minimum phlegmatization concentration of nitrogen, which is 48.1% vol. and the value of the combustible substance at point D (1.45% vol.) and E (6.31% vol.). Adding a phlegmatizer to the "cyclopentane – air" system gradually increases the lower branch and lowers the upper branch of the phlegmatization curve, which converge at the point of the minimum phlegmatizing nitrogen concentration. The area intersecting the phlegmatization curve is capable of flame propagation, outside of which the gas mixture is not explosive and fire-hazardous. A method for calculating the minimum safe ratio of phlegmatizer and combustible gas (steam) in technological devices based on analytical dependencies has been developed and tested. Based on the results of determining the smallest ratio of volumetric concentrations of the phlegmatizer and the combustible substance in the gas mixture, at which it is impossible to ignite it in the environment of this oxidant, the coordinates of the contact point L (45.945% vol.; 1.7445% vol.) were established, as well as the numerical value of the coefficient k (k = 26.4). The practical significance lies in the fact that the results obtained were taken into account when creating a fire-safe technology for manufacturing thermal insulation.

Keywords: *phlegmatization of flammable gas environments, nitrogen, cyclopentane, thermal insulation, fire protection efficiency.*

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1. Introduction

In the production of refrigeration and heating equipment, as well as in the production of pipelines for heat carriers, one of the problems is the use of thermal insulation is the degradation of polyurethane foam during operation. In particular, the use of carbon dioxide for foaming polyurethane foam leads to its rapid diffusion and replacement by air in the pores of thermal insulation, which increases thermal conductivity. Therefore, a technology was proposed for creating polyurethane foam thermal insulation, which includes the introduction of a liquid component (foaming agent) such as cyclopentane, which is characterized by a low boiling point, into the reaction mass. In this case, foam is formed that fills the cavities and, as a result of polymerization, hardens. Polyurethane foam, which is made on the basis of cyclopentane as a foaming agent, has fairly high thermal insulation properties, which significantly improves the operation of refrigeration and heating equipment and reduces the cost of energy production.

Until recently, freons R11 ($CFCl_3$) and R141b ($C_2H_3FCl_2$) were used for foaming the reaction mass, which do not pose a significant fire hazard, but they are characterized by high values of ozone depletion potential and have been discontinued. In this regard, the problem of replacing them with environmentally safe analogues arose, in particular in the field of thermal insulation production and for the production of polyurethane foam. In this regard, cyclopentane (boiling point 49°C), which is close to them in terms of physicochemical properties, has been proposed, which does not belong to ozone-depleting substances. This is explained by the favorable combination of low thermal conductivity, good solubility in polyols and an acceptable boiling point of cyclopentane compared to other hydrocarbons. The thermal conductivity of cyclopentane is not only lower than that of most other hydrocarbons, but also has a low global warming potential [1]. However, it is a flammable liquid that forms flammable vapor-air mixtures. Therefore, in the case of its use, it is necessary to take additional measures to ensure fire and explosion safety of both the technological process of polyurethane foam formation and auxiliary operations related to unloading, storage and transportation of cyclopentane. The most effective measure is phlegmatization of the gaseous medium by introducing a gaseous fire extinguishing agent, such as nitrogen, into its composition.

Thus, there is a need to develop work to determine the conditions for phlegmatization of gaseous cyclopentane combustible media with nitrogen.

The paper [2] discusses the use of large-capacity batteries in energy storage systems, thermal discharge control and the associated fire and explosion risks. Liquid nitrogen, known for its extremely low boiling point and oxygen displacement properties, was evaluated for its cooling and inertizing effect. In this research, an experimental chamber was set up inside an energy storage container to conduct liquid nitrogen inerting experiments on several 280 Ah LFP batteries during thermal discharge events. Comprehensive experimental results confirmed that liquid nitrogen effectively suppressed thermal discharge by rapidly reducing both temperature and oxygen concentration, thereby preventing ignition of combustible gases. The experiments covered both simulated gas environment tests and real battery thermal discharge scenarios, revealing significant differences in gas composition and reactivity. In the simulated experiments, the injection of 14.58 kg of liquid nitrogen reduced the oxygen concentration from 18.3% to 6.2%. In real battery scenarios, the injection of liquid nitrogen ensured safety by maintaining a lower oxygen volume fraction, effectively preventing explosions. These results highlight the dual suppression capabilities of liquid nitrogen, both cooling and inerting, and demonstrate its potential to enhance the safety of large-capacity LFP-based energy storage applications. However, due to the dynamic nature of thermal runaway in real batteries, controlling the inerting environment remains challenging, indicating the need for further research to optimize safety measures and liquid nitrogen application systems.

In [3], the temperature after adiabatic compression of common combustible gases such as methane and oxygen is estimated, and it is assumed that adiabatic compression under negative pressure of impactors can lead to accidental ignition and explosion. In addition, the explosion hazard associated with gas combustion devices is discussed from several perspectives, including the explosion temperatures and pressures of common combustible gases in the laboratory and the effects of explosion overpressure on human health. From the perspective of inherent safety, the following recommendations are proposed: Relatively safe combustible gases should be selected for experimental schemes whenever possible. Inert gases should be selected based on their inertization capacity. Normal pressure burners or combustion chamber volume and gas should be selected. The mixing tank in deflagration/detonation devices should be minimized as much as possible. Experimental devices should be equipped with appropriate valves, the number of impact parts should be minimized, and a monitoring and condition control system should be used to promptly respond to emergency situations. In addition, filters, flame arresters, safety valves and other necessary safety devices should be installed. In [4], the inert effect of gaseous inhibitors on the ignition process of dust clouds in $O_2/N_2/CO_2$ atmospheres was experimentally and theoretically studied, with an emphasis on the role of the CO_2/N_2 ratio. 10 different combustible carbon dusts were selected, including grain dust, biomass dust and coal dust. Experimental results showed that the inhibitory effect of CO_2/N_2 is closely related to the ignition mechanism of dust clouds. In particular, a higher CO_2/N_2 ratio gives a stronger inhibitory effect on the ignition process of dust samples with a relatively low content of volatile substances, where heterogeneous ignition prevails. In addition, two new models of the steady-state ignition mechanism were developed to interpret the experimental observations. The Maxwell-Stefan equation was used to describe the diffusion coefficient in ternary gas mixtures $O_2/N_2/CO_2$. The analytical results were in good agreement with experimental data on the minimum ignition temperature of a dust cloud (MITC) in low-oxygen atmospheres. The mechanism modeling can be used to estimate the critical ignition temperature of all carbonaceous dust clouds with a wide range of volatiles in different inert atmospheres. However, it is not known whether it will provide a guideline for assessing the explosion hazard of dust generated by a hot surface in the processing industry. However, no relevant physicochemical data on the change in their properties during operation are provided.

In [5] addresses the flammability of fuels and oils and hydraulics, fuel tank fires and explosions, engine fires, post-accident fires, flammability of polymeric materials, cabin fires. As well as cargo and concealed area fires, composite structure flammability, lithium battery fires (cabin and cargo), external hazards that may affect the aircraft, and fire suppression systems (detection and suppression). Topics include minimum performance standards for aircraft hand-held fire extinguishers, halon replacement agents, cargo and nacelles fire suppression/extinguishing systems, and fuel tank explosion protection. Including: nitrogen inerting, onboard inert gas generation system/onboard oxygen generation system (OBIGGS/OBOGS), aircraft concealed area fire suppression systems, and fire/smoke detection systems. However, the scope of application of the given products is not specified.

As noted in [6], heterogeneous, stratified combustible gas mixtures and the propagation of flame in such mixtures are encountered in many situations, including numerous technical applications, as well as in various potentially hazardous circumstances. Leakage of fuel from storage tanks or pipelines, formation of combustible mixtures in building spaces, formation of gas pockets in coal mines, and inerting of combustible mixtures by addition of a diluent gas are some examples. Here, some aspects of flame propagation and mass transfer in a confined, stratified gas environment are considered. The diluents nitrogen and helium were alternately used to coat initially combustible methane-air or hydrogen-air mixtures. Stratification of the mixture was achieved by mutual diffusion of two initially homogeneous gas systems at constant temperature in a long, vertical, smooth, closed circular tube. Only upward flame propagation was investigated, as this regime is expected to have the widest flammability limits and the fastest propagation for both homogeneous and stratified methane-air mixtures when contained in pipes. However, no relevant physicochemical data on the change during operation are provided. As stated in [7], the mixture will not become flammable regardless of the amount of fuel added as long as the oxygen content is below the so-called limiting oxygen concentration (COC). Therefore, it is appropriate to determine the inertization-related characteristics of two different inert gases: nitrogen (N_2) and carbon dioxide (CO_2). Therefore, the COC of a number of alcohols and ketones was measured. The experimental determination of the COC was carried out according to EN 1839 using the tube method at an initial ambient pressure and an appropriate initial temperature for the evaporation of the liquid sample. Based on the results, triangular diagrams were constructed, depicting the explosion ranges, and additional parameters related to inerting were obtained. In particular, the minimum inert gas to air ratio (IAR) and the minimum inert gas to combustible gas ratio (ICR). It is well known that carbon dioxide has a stronger inerting effect than nitrogen. Accordingly, the ratio $LOC(CO_2)/LOC(N_2)$ has now been determined to be a constant value of 1.33 ± 0.04 in the applicable temperature range at atmospheric pressure. In inerting applications, this ratio will in the future help to calculate $LOC(CO_2)$ from the much more commonly known $LOC(N_2)$. However, the effect of environmental changes on its degradation over time is not mentioned.

Ternary systems containing a flammable gas, an inert gas and air, as specified in [8], were investigated in order to give the user an assessment of the method for calculating the flammability of gas mixtures according to ISO 10156. While part 1 of this article focused on the fire potential of flammable gases, the effect of inert gases on the flammability of gas mixtures was studied in part 2. The inertization capacity of an inert gas is expressed by a dimensionless value *K*, the so-called "nitrogen equivalent factor". The experimental determination is demonstrated by means of explosion diagrams. The aim of this research was to compare the calculated results given by ISO 10156 with the measurements of

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the explosion ranges based on the German standard DIN 51649-1 provided by CERN and CHEMSAFE. However, in a number of cases, ISO underestimates the inertization capacity, so non-flammable gas mixtures are considered flammable.

In [9], the results of experimental and numerical researches of CO formation in a kinetic flame of natural gas under oxy-fuel combustion conditions, when it is impossible to provide a completely nitrogen-free atmosphere, are presented. The aim of these researches was to determine the effect of adding nitrogen to the furnace chamber on changes in CO concentration. Adding nitrogen to oxy-fuel combustion reflects the sources of nitrogen in industrial furnaces, which can be: additives used to accelerate the melting of the charge in glass production, insufficient oxygen purity, and a leaky chamber. It was experimentally demonstrated that by adding nitrogen to an oxidizing atmosphere consisting of 27 vol./vol. % O_2 and CO_2, the CO concentration in the flame region decreases. For this purpose, an analysis of the reaction rates that have a significant effect on the CO concentration in the flame was carried out, and the predominant reactions responsible for CO formation were listed. It was noted that both an increase in the residence time of combustion products and an increase in the nitrogen content in the combustible mixture lead to a decrease in the CO concentration in the flame.

In [10], the effect of using premixed inert gas (N_2 was 50%, 60%, 70% and 80%) instead of CO_2 on the inertization effect of a nearly horizontal and shallow produced space was studied by establishing an experimental platform to simulate physical similarity. The experimental results showed that the inertization effect of the premixed inert gas was better than that of CO_2. For example, in a nearly horizontal produced space, the inertization effect of the premixed inert gas was optimal when N_2 was 70%. The average O_2 concentration in the controlled zone decreased from 9.7% with CO_2 to 6.4%. In addition, in the shallow produced space, the premixed inert gas exhibited a storage state similar to CO_2, mainly in the lower part adjacent to the face. In addition, the storage state of the premixed inert gas was inversely proportional to its inertization effect. This research has important reference value for the application of inert gas fire prevention and extinguishing technology in mines. However, the mechanism is not specified and the operating conditions are not revealed.

In [11], it is noted that in order to assess the fire/explosion risks and ensure the safety and optimal operation of reactors, tanks or storage tanks, it is necessary to recognize the flammability characteristics of flammable materials. Using a 20-liter apparatus, it is possible to investigate the flammability and explosive characteristics of methane with three different inert gases (CO_2, N_2 and Ar) at a pressure of 1 atm, temperatures of 30 and 100°C and different air concentrations. Our test results showed that the inert effect of CO_2 on methane is better than that of N_2 and Ar. The results of this paper include graphs showing the lower and upper flammability regions and explosive characteristics for these inert systems. Flammability ranges include results for 30 and 100°C, not stated as for industrial compatibility and production.

In [12], experiments were conducted on three different process burners (a staged fuel burner, a staged air burner and a low-calorie burner) to describe the effect of inert gases on flame characteristics, emissions and heat flux to the combustion chamber wall. Natural gas was used as the reference fuel, and a thermal power of 500 kW was maintained throughout all tests. N_2 and CO_2 were used as diluents to simulate the combustion of alternative fuels with lower calorific values (LHV). Inert gas in hydrocarbon fuels can, under certain conditions, reduce NO_x emissions (up to 80%) and increase heat flux (up to 5%). As soon as non-combustible compounds are present in the fuel, a larger amount of fuel flowing through the nozzles affects the flow in the combustion chamber, increasing the Reynolds number. This can change the shape of the flame and the temperature field, and it can be both positive and negative, depending on the actual conditions.

Thus, it has been established from the literature that inert gases are able to neutralize the flammability of gaseous combustible mixtures during operation, but their quantity needs to be established. Therefore, the establishment of phlegmatization parameters and the influence of inert gases on this process necessitated the need for research in this direction.

The object of research is the process of phlegmatization of the gaseous medium of cyclopentane vapor-air mixtures when nitrogen is introduced into this mixture.

The scientific hypothesis is the inhibition of the flammability indicators of the gaseous combustible medium during phlegmatization with gaseous fire extinguishing agents.

The research adopted simplifications that relate to the process of phlegmatization of the gaseous combustible medium as a model. These include features that determine the effect of changing conditions on the object of research and the passage of the phlegmatization process: external and internal influences, temperature, humidity, atmospheric pressure.

The aim of research is to identify the regularities of phlegmatization of the cyclopentane vapor-air medium with nitrogen under the influence of an ignition source. This makes it possible to substantiate the requirements for the use of inert diluents for phlegmatization of gaseous combustible media and expand the scope of their application.

To achieve the aim, the following objectives were solved:

– to establish the concentration limits of the phlegmatization region in the "cyclopentane – air – nitrogen" system and, based on the results obtained, to substantiate the most acceptable conditions for phlegmatization of gaseous media with nitrogen;

– to develop a method for calculating the lowest volumetric concentration of nitrogen in a mixture with cyclopentane vapor, at which stable (torch) combustion of this mixture in air is impossible.

2. Materials and Methods

To study the process of phlegmatization of cyclopentane vapor-air media, balloon nitrogen was used, and cyclopentane was used as a combustible substance. It is a flammable liquid, with a flash point of minus 37°C, an autoignition temperature of 320°C, a density of 0.745 g/cm^3 (+20°C), insoluble in water, miscible with organic solvents (ethanol, acetone, diethyl ether). Vapors can form explosive mixtures with air (concentration limits of flame propagation: lower 1.52% vol., upper 8.71% vol.) [13].

To establish the regularities of phlegmatization of vapor-air flammable media, nitrogen-air mixtures of a certain concentration with flammable cyclopentane vapors were used, which were obtained on a device for determining the minimum phlegmatizing concentration of gaseous fire extinguishing agents (ISO 14520-1). The scheme of the device for determining the minimum phlegmatizing concentrations of gaseous fire extinguishing agents is shown in Fig. 1.

The determination of the minimum phlegmatizing concentration is based on a method based on the sequential preparation in a test chamber of gas mixtures from a combustible substance, air and a gaseous fire extinguishing agent of a given composition. And with subsequent measurement of the excess pressure that is created after introducing an ignition source with a given energy into them.

Determination of the concentration limit of flame propagation and the minimum phlegmatizing concentration:

1) evacuate chamber 1 to a residual pressure of no more than 1 kPa;

2) prepare a gas mixture of a given quantitative composition in chamber 1 according to the partial pressures of the components by sequentially introducing a gaseous combustible substance and an oxidizer;

3) allow the gas mixture to homogenize and reach ambient temperature for at least 15 minutes;

4) charge the capacitors of the ignition unit 4 and create an electric discharge in chamber 1;

5) measure the peak value of the excess pressure P_e in chamber 1. The mixture is capable of flame propagation if this value exceeds 7 kPa;

6) if the value of P_e obtained in point 5) does not exceed 7 kPa, by partially evacuating chamber 1 and subsequently introducing an equivalent amount of combustible substance, the oxidant concentration is reduced (by no more than 4% relative) and the operations according to points 1)–5) are performed;

7) the procedure according to point 6) is repeated until, after creating an electric discharge in chamber 1, a value of P_e of more than 7 kPa is recorded;

8) the lower concentration limit of flame propagation is taken to be the average value between the concentration of the combustible substance in the mixture with the oxidant at the point of transition of P_e in the chamber.

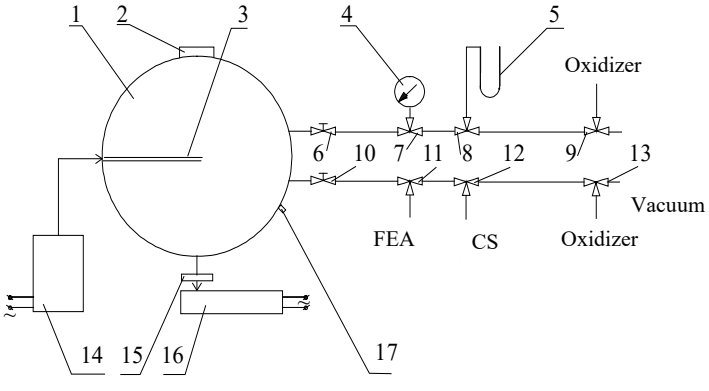

Fig. 1. Scheme of the installation for determining the minimum phlegmatizing concentration: 1 – test chamber; 2 – inspection window; 3 – discharge device; 4 – vacuum gauge (standard vacuum gauge, manufactured by ROSS, Ukraine); 5 – mercury vacuum gauge (manufactured by DEYS, Ukraine); 6 – outlet valve; 7, 8, 9, 11, 12, 13 – three-way valves; 10 – inlet valve; 14 – ignition unit; 15 – pressure transducer; 16 – oscilloscope (digital oscilloscope FNIRSI 1014D, MikroAmpermetr, Ukraine); 17 – safety valve. FEA – fire extinguishing agent, CS – combustible substance

The disadvantage of this method of determining the phlegmatizing concentration of nitrogen is the significant labor costs associated with determining the points of the phlegmatization curve.

For this purpose, a research methodology is presented, which combines elements of experiment and mathematical calculations in a ratio sufficient to obtain reliable results with relatively small labor costs.

In the general case, the region of explosive compositions of gas mixtures of the "combustible substance – air – phlegmatizer" system is limited by the ACB phlegmatization curve (Fig. 2).

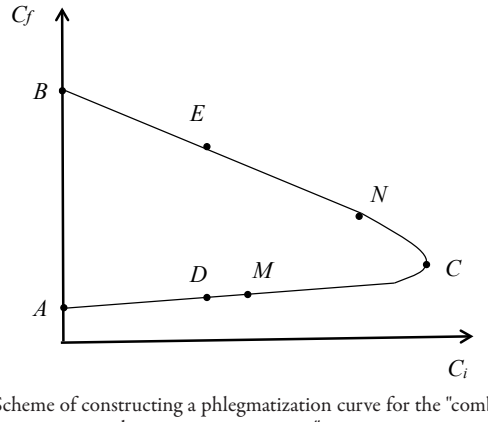

Fig. 2. Scheme of constructing a phlegmatization curve for the "combustible substance – air – nitrogen" system

Points *A* and *B* correspond to the lower and upper concentration limits of flame propagation in mixtures of a combustible substance with an oxidant. A point *C* (the peak of the phlegmatization curve) is the smallest sufficient content of the phlegmatizer at the most unfavorable ratio of the combustible substance and the oxidant. Considering that each of the two branches of the mentioned *ACB* curve (both upper and lower) consists of a linear section adjacent to the axis OC_i, and a nonlinear section described by a power dependence of at least the 2nd degree. Linear and nonlinear sections converge at hypothetical points *M* and *N*.

In order to obtain analytical expressions for all four sections, it is necessary to experimentally determine the coordinates of at least 5 points: *A*, *B*, *C*, as well as arbitrary points *D* and *E*, which belong to the lower and upper branches of the phlegmatization curve, respectively.

In general, these dependencies have the following form.

For the lower branch of the *ACB* curve

$$c_i\left(c_f\right)=\begin{cases}\dfrac{c_{i(D)}}{c_{f(D)}-c_{f(A)}}\left(c_f-c_{f(A)}\right), & c_f\in\left[c_{f(A)};c_{f(M)}\right],\\ & c_i\in\left[0;c_{i(M)}\right];\\ a\cdot\left(c_f-c_{f(C)}\right)^n+c_{i(C)}, & c_f\in\left[c_{f(M)};c_{f(C)}\right],\\ & c_i\in\left[c_{i(M)};c_{i(C)}\right].\end{cases}\quad(1)$$

For the upper branch

$$c_i\left(c_f\right)=\begin{cases}\dfrac{c_{i(E)}}{c_{f(E)}-c_{f(B)}}\left(c_f-c_{f(B)}\right), & c_f\in\left[c_{f(A)};c_{f(N)}\right],\\ & c_i\in\left[0;c_{i(N)}\right];\\ b\cdot\left(c_f-c_{f(C)}\right)^m+c_{i(C)}, & c_f\in\left[c_{f(N)};c_{f(C)}\right],\\ & c_i\in\left[c_{i(N)};c_{i(C)}\right].\end{cases}\quad(2)$$

Coefficients *a*, *b*, *n*, *m* ($n, m \geq 2$) are parameters that depend on the physicochemical properties of the combustible substance and the phlegmatizer, which must be found in the process of solving the problem.

Thus, the dependences that describe the concentration limits of the phlegmatization region for gas mixtures of the "cyclopentane – air – nitrogen" system were obtained.

3. Results and Discussion

3.1. Determination of the concentration limits of the phlegmatization region in the "cyclopentane – air – nitrogen" system

The results of researches on determining the concentration of the combustible substance (C_f) and phlegmatizer (C_i) for mixtures corresponding to points *A*, *B*, *C*, *D*, *E* of the phlegmatization curve are given in Table 1 in the form of coordinates of the corresponding points.

Table 1

Experimental data necessary for calculating the phlegmatization curve

Name of the flammable substance	Coordinates of the points of the phlegmatization curve, % vol.							
	C_{fa}	C_{fb}	C_{fd}	C_{id}	C_{fe}	C_{ie}	C_{fc}	C_{ic}
Cyclopentane	1.34	8.64	1.45	22.0	6.31	22.0	2.09	48.1

Based on the experimental data in Table 1, using equations (1) and (2), the dependences describing the phlegmatization curve of a mixture of cyclopentane with air when nitrogen is used as a phlegmatizer were obtained:

$$C_i\left(C_f\right)=\begin{cases}221\left(C_f-1.34\right);\ C_f\in\left[1.34;1.42\right];\ C_i\in\left[0;13.9\right];\\ -184\left(C_f-2.09\right)^4+48.1;\ C_f\in\left[1.42;2.09\right];\ C_i\in\left[13.9;48.1\right];\end{cases}\quad(3)$$

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$$C_i\left(C_f\right)= \\ =\begin{cases}9.35\left(8.66-C_f\right); C_f\in\left[4.96;8.66\right]; C_i\in\left[0;34.6\right]; \\ -1.62\left(C_f-2.09\right)^2+48.1; C_f\in\left[2.09;4.96\right], C_i\in\left[34.6;48.1\right].\end{cases} \quad (4)$$

Fig. 3 shows the phlegmatization curve of cyclopentane with nitrogen, which was calculated using equations (3) and (4).

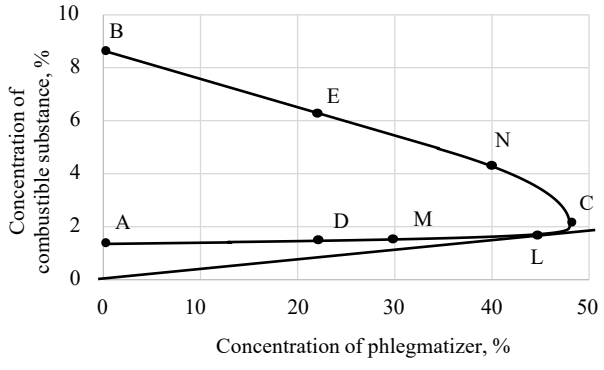

Fig. 3. Phlegmatization curve for the system "cyclopentane – air – nitrogen"

Thus, the mechanism of formation of concentration limits of flame propagation in the system "combustible substance – air – gaseous fire extinguishing agent" is substantiated. Namely, the addition of a phlegmatizer to the system "combustible substance – oxidizer" gradually increases the lower branch and lowers the upper branch of the phlegmatization curve, which converge at the point of minimum phlegmatization concentration. The area intersecting the *ABC* curve is capable of flame propagation, outside its limits the gas mixture is not explosive and fire hazardous.

3.2. Development of a method for calculating the diffusion combustion limit in a mixture of nitrogen and cyclopentane

In practical terms (for fire protection of technological processes), the lower part of the phlegmatization curve is most noteworthy, especially point *L* (Fig. 3), through which a straight line tangent to the *ABC* curve on the *MS* section passes from the origin

$$C_N^0=k\cdot C_f. \quad (5)$$

Having established the coordinates of point *L*, it is possible to determine the value of *k* – the smallest ratio of volumetric concentrations of the phlegmatizer and the combustible substance in the gas mixture, at which it is impossible to ignite it in the environment of a given oxidant. This ratio is important to know to prevent the ignition of combustible gas or vapor in the event of their leakage from the technological apparatus into the air atmosphere.

In terms of physical content, the indicator of the explosion and fire hazard of combustible substances ("diffusion combustion limit") is the lowest volumetric concentration of a gaseous fire extinguishing agent (phlegmatizer) in a mixture with a combustible substance, at which stable (torch) combustion of this mixture in air is impossible. This indicator can be determined with sufficient accuracy for any system "combustible gas (steam) – phlegmatizer" based on the research results of the phlegmatization curve.

To find the coordinates of point *L* of the phlegmatization curve of gas mixtures of the "cyclopentane – air – nitrogen" system (Fig. 3), as well as the numerical value of k, let's solve the system of equations:

$$\begin{cases}C_i=-184\left(C_f-2.09\right)^4+48.1; \\ C_i=k\cdot C_f; \\ k=-742\left(C_f-2.09\right)^3,\end{cases} \quad (6)$$

the first of which is the equation of the *MC* curve, the second – of the *OL* line, and the third is the condition of equality of the angular coefficients of the *OL* line and the tangent to the *MC* curve.

In the process of solving this system, an algebraic equation of the 4th degree arises, which is solved by one of the approximate methods (for example, Newton's method). As a result, let's obtain the coordinates of the point of contact: *L* (45.945% vol.; 1.7445% vol.), as well as the numerical value of the coefficient *k* (*k* = 26.4).

Thus, a method for calculating the minimum safe ratio of phlegmatizer and combustible gas (vapor) in technological devices has been developed and tested based on analytical equations that describe the concentration limits of the phlegmatization region of the corresponding combustible gas medium.

Having analytical dependencies (3) and (4), it is possible to calculate the minimum concentration of nitrogen sufficient for phlegmatizing a mixture of cyclopentane vapors with air of any quantitative composition. If this composition is unknown, it is necessary to ensure that the minimum phlegmatizing concentration $C_{i(C)}$ 48.1% vol. is achieved (Fig. 3). In practice, when designing fire protection systems, the value of the normative phlegmatizing concentration should be used, which takes into account the safety factor of 1.1.

Having determined the coefficient *k* of equation (5), it is possible to calculate the above-mentioned "diffusion combustion limit" C_l for mixtures of combustible vapors of cyclopentane with nitrogen

$$C_l=\frac{k}{k+1}\cdot 100\%\text{ vol.} \quad (7)$$

However, in practical calculations for a sealed space, it is more convenient to operate not with the volumetric concentrations of the components of the gas mixture, but with the values of their partial pressures. If the saturated vapor pressure of cyclopentane p_f is known, then the partial pressure of nitrogen p_N^0, necessary to ensure fire protection of technological equipment, can be found from the equation

$$p_N^0=k\cdot p_f. \quad (8)$$

Considering that the saturated vapor pressure of cyclopentane at a temperature of 20°C, according to [14, 15], is 0.042 MPa. If *k* = 26.4, then p_f^0 = 26.4 × 0.042 MPa = 1.108 MPa. Therefore, at this temperature, the absolute (total) pressure in the reactor, where cyclopentane is stored under a nitrogen "cushion", must be at least 1.108 MPa, and the excess pressure must be at least 0.946 MPa (provided that the atmospheric pressure is 0.100 MPa).

In general, the dependence of determining the concentration limits of the phlegmatization region in the "cyclopentane – air – nitrogen" system is described by equations that depend on the physicochemical properties of the combustible substance and the phlegmatizer, which are given by the phlegmatization curve of cyclopentane with nitrogen, Fig. 3.

Based on the results of determining the smallest ratio of volumetric concentrations of the phlegmatizer and the combustible substance in the gas mixture, at which it is impossible to ignite it in the environment of a given oxidant, the coordinates of the contact point *L* (45.945% vol.; 1.7445% vol.), as well as the numerical value of the coefficient *k* (*k* = 26.4).

3.3. Discussion

When studying the area of explosive compositions of gas mixtures of the "combustible substance – air – phlegmatizer" system, which is limited by the *ACB* phlegmatization curve (Fig. 2), it is natural that the combustion process is inhibited outside its limits. Since points *A* and *B* correspond to the lower and upper concentration limits of flame propagation in mixtures of a combustible substance with an oxidant.

And point *C* (the peak of the phlegmatization curve) is the smallest sufficient content of the phlegmatizer for the most unfavorable ratio of the combustible substance and the oxidant. Each of the two branches of the *ACB* curve consists of a linear section adjacent to the axis OC_i, and a nonlinear one, which is described by a power dependence of at least the 2nd degree. Linear and nonlinear sections converge at hypothetical points *M* and *N*. To obtain analytical expressions for all four sections, it is necessary to experimentally determine the coordinates of at least 5 points: *A*, *B*, *C*, as well as arbitrary points *D* and *E*, which belong to the lower and upper branches of the phlegmatization curve, respectively. According to the results of experiments on determining the coordinates of the points of the phlegmatization curve, the value of the lower concentration limit of flame propagation was obtained, which corresponds to the value of 1.34% vol. of cyclopentane in a mixture with air. As well as the upper concentration limit of flame propagation, which is equal to 8.64% vol., the value of the minimum phlegmatizing concentration of nitrogen, which is 48.1% vol. and the value of the combustible substance at point *D* (1.45% vol.) and *E* (6.31% vol.). In general, these dependencies are described by equations that depend on the physicochemical properties of the combustible substance and the phlegmatizer, which are given by the phlegmatization curve of cyclopentane with nitrogen, Fig. 3.

According to the results of determining the smallest ratio of volumetric concentrations of the phlegmatizer and the combustible substance in the gas mixture, at which it is impossible to ignite it in the environment of a given oxidant, the coordinates of the contact point *L* (45.945% vol.; 1.7445% vol.) were established, as well as the numerical value of the coefficient *k* (*k* = 26.4).

This indicates that it is possible to calculate the minimum concentration of nitrogen sufficient for phlegmatizing a mixture of cyclopentane vapors with air of any quantitative composition. If this composition is unknown, it is necessary to ensure that the minimum phlegmatizing concentration $C_{i(C)}$ 48.1% vol. is achieved (Fig. 3). This reflects the participation of nitrogen in the phlegmatization of cyclopentane vapor-air mixtures, which can be identified directly by high-temperature exposure to gas-air mixture samples [16, 17].

Unlike the researches presented in [2, 4, 5], where the main attention is paid to the use of liquid nitrogen or combustion inhibitors that suppress the combustion of a gaseous combustible medium, this research considers nitrogen, which is widely available on the market.

However, unlike the results obtained in [18–20] regarding the inhibition of the combustion process of combustible substances, the following can be stated:

– the regulator of extinguishing combustible substances with nitrogen is not only the retention of the oxidant in the volatile and condensed stages, but also the cooling of the fire center when supplying a gaseous extinguishing agent;

– a significant contribution to the inhibition of the flame propagation process occurs in the direction of inhibition and dilution of the gaseous combustible medium and the formation of such an amount of gaseous fire extinguishing agent in a mixture with a combustible substance that stable combustion in air is impossible.

The obtained experimental data contain certain limitations when the limits of diffusion combustion are set due to the unpredictability of vaporization of the combustible substance, which can be taken into account provided that the minimum phlegmatizing concentration of $C_{i(C)}$ 48.1% vol., which is formed during the supply of nitrogen, is guaranteed.

Such imprecision is difficult to establish during this research, since the need to conduct additional experiments to obtain more data becomes important. This, for example, requires new data to study the minimum phlegmatizing concentration of nitrogen and establish the stage at which the loss of the minimum safe ratio begins. Such an action makes it possible to transform the gaseous medium towards shifting the concentration boundary of the phlegmatization region. This also makes it possible to detect differences that affect the volumetric concentrations of the components of the gas mixture.

4. Conclusions

1. The coordinates of the points of the phlegmatization curve were experimentally determined, the value of the lower concentration limit of flame propagation was obtained, which corresponds to the value of 1.34% vol. of cyclopentane in a mixture with air. As well as the upper concentration limit of flame propagation, which is equal to 8.64% vol., the value of the minimum phlegmatization concentration of nitrogen, which is 48.1% vol. and the value of the combustible substance at point *D* (1.45% vol.) and *E* (6.31% vol.).

The mechanism of the formation of the concentration limits of flame propagation in the "cyclopentane – air – nitrogen" system was substantiated. Namely, the addition of a phlegmatizer to the "cyclopentane – air" system gradually increases the lower branch and lowers the upper branch of the phlegmatization curve, which converge at the point of the minimum phlegmatization concentration of nitrogen. The area that intersects the phlegmatization curve is capable of spreading the flame; outside its limits the gas mixture is not explosive and fire-hazardous.

2. A method for calculating the minimum safe ratio of phlegmatizer and combustible gas (vapor) in technological devices has been developed and tested based on analytical dependencies that describe the concentration limits of the phlegmatization region of the corresponding combustible gas environment. Based on the results of determining the smallest ratio of volumetric concentrations of the phlegmatizer and combustible substance in the gas mixture, the coordinates of the contact point *L* (45.945% vol.; 1.7445% vol.) were established. As well as the numerical value of the coefficient *k* (*k* = 26.4) at which it is impossible to ignite in the environment of a given oxidant.

Conflict of interest

The authors declare that they have no conflict of interest in relation to this research, whether financial, personal, authorship or otherwise, that could affect the research and its results presented in this paper.

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Manuscript has no associated data.

Use of artificial intelligence

The authors confirm that they did not use artificial intelligence technologies in creating the submitted work.

Authors' contributions

Yuriy Tsapko: Conceptualization, Methodology, Formal analysis, Writing – review and editing, Supervision; ***Oksana Berdnyk***: Investigation, Data analysis, Formal analysis, Writing – original draft; ***Aleksii Tsapko***: Conceptualization, Investigation, Data analysis, Writing – original draft; ***Ruslan Likhniovsky***: Methodology, Data curation, Validation; ***Anastasiia Kovalova***: Resources, Validation, Funding acquisition; ***Kostiantyn Sokolenko***: Resources, Supervision, Validation; ***Kostiantyn Kaveryn***: Validation, Funding acquisition, Visualization; ***Anna Borysova***: Funding acquisition, Validation, Visualization; ***Oleksandr Dotsenko***: Investigation, Resources, Funding acquisition.

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