Application of Wave Technologies for the Preparation of Fuels Containing Organic Waste Pyrolysis Products

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Abstract: Products of fast pyrolysis of organic materials (bio-oil) have such advantages as ease of storage and transportation, production from renewable raw materials, but have not yet found wide application as motor and boiler fuels. The article describes the results of research aimed at obtaining such fuel containing petroleum fuel oil and bio-oil as the main components. Direct mixing of these components is a difficult task due to their chemical incompatibility. Its solution required the development of technology and devices, which was the purpose of the presented research results. The research methodology was based on mathematical modeling of the processes of mixing fuel components, analysis of its results, their transfer to natural samples and their experimental study. As a result, a technology and design of mixers were developed that made it possible to obtain a stable mixed fuel containing fuel oil and bio-oil in the following proportions of the mass ratio up to 50:50. The obtained results confirmed the correctness of the chosen approach, which made it possible to develop new technology and equipment to achieve the stated research goal.

Keywords: Motor fuels, Blended fuels, Biofuels, Wave technologies.

1. INTRODUCTION

Currently, active research is being conducted in the global scientific community and technical practice to create mixed fuels based on oil and plant components [1]. This trend is due to the need to reduce dependence on minerals in the context of rising world prices for hydrocarbons, the emergence of laws regulating carbon dioxide emissions into the atmosphere, as well as the desire to obtain products with high added value.

Plant biomass occupies a special place among other renewable energy sources, since it can be processed to produce motor and boiler fuel [2]. Therefore, the problem of using components obtained from plant materials, including wood and agricultural waste, in mixed fuels is of particular importance [3]. Unlike the use of agricultural crops for these purposes, these materials do not lead to an increase in food prices, soil depletion, deforestation in order to create new areas for "fuel" crops, and, as a consequence, climate change.

One of the types of wood processing for the purpose of obtaining fuel is fast pyrolysis, which is a process of thermal decomposition of organic compounds without oxygen access at a temperature of 400–500 °C. Its main feature is the high heating rate of raw material particles (several thousand degrees per second), rapid cooling of intermediate decomposition products and removal of these products from the reaction zone [4].

As a result of wood pyrolysis, the following are formed: charcoal and a steam-gas mixture, which in turn is divided into non-condensable gases and liquid products (bio-oil) [5]. It should be noted that there is still no uniform terminology regarding liquid products of thermal conversion of plant materials. They are called pyrogenic resins, bio-oil, biomass, pyrolysis liquid, etc. [6]. In this article, we use the term bio-oil.

The advantage of bio-oil is that it is easy to store, transport and is produced from renewable raw materials.

Depending on the temperature of the wood pyrolysis process, the ratio of the products obtained changes. To increase the yield of liquid products during wood pyrolysis, it is necessary to increase the heating rate and reduce the duration of the products' stay in the destruction zone. This is ensured by a processing time of less than 2 seconds [4].

There are many scientific studies and examples of industrial implementation of technologies for obtaining pyrolysis products. However, unlike traditional biofuels - biodiesel and bioethanol, products of fast pyrolysis of organic materials have not yet found wide application as motor and boiler fuels or additives to them. There are only isolated examples of private use of pyrolysis

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biofuel in stationary diesel engines, furnace and boiler units, for example, at asphalt-concrete plants. The problem of expanding the areas of application of pyrolysis fuel by developing new technologies for the preparation of mixed hydrocarbon fuel on its basis is the subject of this work.

2. MATERIALS

Marine internal combustion engines have relatively low requirements for fuel quality, therefore, at the considered stage of research, it was chosen to produce mixed emulsion fuels for these purposes. The emulsification of bio-oil into heavy petroleum fuels (fuel oil) with the addition of surfactants (SF) was studied.

Petroleum fuel used in marine equipment, as well as in stationary boiler installations, is a composition consisting of residual products of direct distillation of oil (fuel oil, tar) with or without the addition of residues of destructive processes - the residue of thermal cracking of fuel oil and the residue of visbreaking tar - and components that provide the required viscosity of fuel compositions [7-9].

The basis of this fuel is fuel oil or residual (heavy) fuel obtained after extraction of light fractions from oil. By mixing it in certain proportions with diesel fractions, naval fuel oil is obtained. Such fuel oil is burned in ship boiler rooms, used for the operation of diesel and gas turbine installations.

The classification of naval fuel oil is based on its most important characteristic, viscosity. Density, coking capacity, ash content and other indicators are also interrelated with it.

The physico-chemical properties of fuel oil depend on the chemical composition of the source oil and the degree of its processing – distillation of distillate fractions. In percentage terms, fuel oil contains the following elements: carbon (C) – 83-87%; hydrogen (H) – 10-12%; sulfur (S) – 1-3.5%; nitrogen and oxygen (N+O) – 0.2–1.9%; other impurities (including water and mechanical impurities) – 0.5-1.0% [10].

Fuel oil refers to petroleum products that are insoluble in water and are nonpolar liquids, unlike liquids that are polar substances that are easily soluble in water and form azeotropic mixtures with it (methyl, ethyl, butyl alcohol, acetone, glycerin).

The improvement of this type of petroleum fuel is aimed at reducing the sulfur content in it, improving operational properties, and reducing environmentally hazardous emissions into the atmosphere.

The mixing of fuel oil with biofuels, in particular, obtained by pyrolysis, is aimed at solving these problems.

The composition of pyrolysis bio-oil depends not only on the type of wood (coniferous, deciduous, a mixture of rocks), but also on the type and mode of thermal exposure, humidity of the feedstock and other parameters. It can contain more than 180 chemicals [11, 12]. Liquid products obtained by high-speed pyrolysis contain organic compounds of various classes - acids, phenols, alcohols, ketones, esters, carbonyl compounds, depolymerization and dehydration products of carbohydrates [13, 14].

The results of studies of pyrolysis fuel have shown that it has acceptable fuel characteristics and can be used as furnace and boiler fuel to produce thermal energy [11]. However, it is necessary to take into account some disadvantages of bio-oil compared to traditional petroleum fuels, namely:

a) corrosion activity due to its high content of water and acid (especially formic and acetic acids) [15, 16];

b) relatively low specific calorific value, determined by the high oxygen content, which is, as a rule, ~ 50 wt.% [17];

c) chemical instability due to the abundance of reactive functional groups such as carbonyl groups and phenolic compounds, which can lead to polymerization during storage [18]. Over time, its viscosity and molecular weight increase simultaneously with the increase in water content. The aging process accelerates with increasing temperature;

d) relatively high viscosity and tendency to phase separation under high shear conditions, for example, in a fuel system nozzle. This creates problems with atomization and smooth combustion of fuel [19];

e) incompatibility, due to insolubility, with traditional hydrocarbon-based (petroleum) fuels [20].

These negative properties do not allow direct use of liquid pyrolysis products as motor and boiler fuels. Therefore, in recent decades, significant efforts have been directed towards the search for practical technologies that can neutralize the above-mentioned disadvantages of bio-oil. In particular, the following methods were used: esterification and acetalization of bionefty with alcohols such as ethanol and butanol [22, 17], direct hydrotreating of bionefty [22-24], co-processing with a hydrocarbon solvent [22, 25], stabilization of bionefty by hydrogenation [26, 27], godroreforming [28]. All these technologies of chemical refining of bio-oil are quite high-tech, labor-intensive in terms of hardware and technical implementation.

From this point of view, the most promising direction is the use of bio-oil as a component of a mixed fuel. At the same time, to overcome incompatibility (insolubility) with petroleum fuels and obtain a stable emulsion, it is necessary to select the appropriate SF, which is not an easy task due to the complex chemical composition of bio-oil. In [17], a large number of surfactants (about 100) were tested, including commercial blends and chemically pure SF formulations. Among them are cationic, anionic, zwitterionic and nonionic stabilizers. Moreover, polymer and non-polymer additives were also tested for each of these SF classes.

Literature analysis showed that the following types of surfactants are used in the emulsification of Bio-Oil: Span 60, Span 80, Span 85, Span 100, Tween 80, Lecithin, Atlox 4914, Lignin, PGO, Lanolin, Hypermer B246, Brij 58 and Brij 72, PEG-DPHS [29-35].

Heavy petroleum fuels contain asphaltenes, resins, paraffins, naphthenic acids and other components that are a kind of natural emulsifiers: on the one hand, they reduce the force of surface tension at the interface, and on the other hand, they stabilize the formed droplets of the dispersed phase due to a strong adsorption-solvate layer at the interface. This makes it possible to partially or completely eliminate the use of SF in the manufacture of mixed fuels based on fuel oil [36].

The emulsion obtained by mixing fuel oil, bio-oil and SF will contain a continuous phase and a dispersed phase of fine droplets. Most of the properties of emulsion systems (stability, viscosity, etc.) depend on the droplet size and their size distribution, which usually covers a fairly wide range, from hundreds of nanometers to tens and hundreds of micrometers [37, 38].

In our case, the continuous phase is liquid hydrocarbon fuel (fuel oil), and the dispersed phase is products of rapid pyrolysis of wood. The emulsion obtained in this case is called a reverse emulsion or a water-in-oil type emulsion. It contains: fuel oil in an amount sufficient to form a continuous phase in the composition; pyrolysis liquid forming an intermittent phase in the composition; at least one emulsifier selected from nonionic hydrophilic surfactants.

Such fuel compositions can contain up to 50% by weight of pyrolysis liquid in the mixed fuel. The surfactant is usually present in an amount of about 0.5 to 5% by weight of the fuel composition [7].

3. METHODS OF RESEARCH

The analysis of the current level of technical developments, development trends and the patent and licensing situation in the field of methods and devices for preparing mixed fuels containing pyrolysis products of organic waste has allowed us to determine the main task in the form of developing a new technology based on nonlinear wave mechanics of multiphase media.

It is based on fundamental scientific achievements in the field of nonlinear wave mechanics, a new field of mechanics developed by the staff of the Scientific Center for Nonlinear Wave Mechanics and Technology of the Russian Academy of Sciences [39-41].

In relation to the research objectives, the scientific basis of the new technology for the preparation of mixed fuels are resonant phenomena and effects in multiphase media, leading to processes of powerful spatial mixing and dispersion of the treated media at low energy consumption. In addition, cavitation-wave processes implemented in flow-through hydrodynamic wave apparatuses can contribute to mechanochemical activation and increase the rate of chemical reactions.

Let's highlight the key points that were taken into account when developing the technology:

1. Pyrolysis bio-oil is a liquid with a complex chemical composition. In its direct form, it does not mix with carbohydrate petroleum fuels due to its polar properties and a large amount of oxygen-containing substances.

2. Bio-oil is quite unstable. This is reflected in the change in the ratios of its chemical components during storage. The viscosity increases, polymerization and precipitation processes occur.

3. Bio-oil is quite chemically aggressive, due to the content of a large amount of water and acids, and has a higher density than petroleum fuels.

With this in mind, the research was focused on:

the development of hydrodynamic cavitation-wave devices (CVA) designed for the preparation of stable finely dispersed fuel emulsions;

selection of effective SF ensuring the stability of an emulsion consisting of fuel oil and bio-oil;

determination of the optimal concentration of the emulsion components.

The research and development of the technology was carried out on the basis of theoretical and experimental methods. In the process of theoretical research, mathematical models of the physical processes occurring in CVA when mixing bio-oil and fuel oil are constructed. Based on them, optimal design parameters were determined, which were later used in the manufacture of experimental equipment. During the experiments, the CVA design was corrected, optimal concentrations of bio-oil, fuel oil and SF in the mixed fuel were determined, and emulsion processing modes were established.

4. RESEARCH RESULTS

4.1. Theoretical Research

To determine the optimal geometry and operating modes of the CVA, a mathematical model was developed and numerical calculations of the flows of viscous liquids (bio-oil and fuel oil), their fine mixing and dispersion were performed.

Hydrodynamic cavitation-wave devices create vibrations and waves when liquid flows through channels of a certain geometry. CVAs do not have any moving parts, which ensures their reliability and durability. The mechanisms of oscillation and wave generation in such generators are diverse. They are caused by complex hydrodynamic processes occurring in the working part when liquid flows through it:

- the occurrence of vortices and their disruption into the flow;
- the occurrence of low static pressure zones in the flows, leading to evaporation of the liquid and the release of gas dissolved in the liquid, followed by the appearance of pulsating vapor-gas-liquid structures with significant compressibility significantly exceeding the compressibility of the surrounding liquid;
- intense collapse of steam and gas bubbles arising in the liquid.

These processes can be described by nonlinear equations that allow us to explore various practical problems [42]. The developed theoretical and experimental method for calculating generators is based on simplified theoretical approaches that make it possible to assess the conditions for the occurrence of highly compressible zones in currents. Numerical analysis of the flows makes it possible to clarify these conditions. However, conducting numerical experiments for the purpose of mathematical modeling of all factors is difficult because it is necessary to take into account many high-frequency and small-scale effects, the identification of which requires significant amounts of machine time and memory. This is especially important for modeling flows of multiphase liquids and emulsion dispersion processes.

In this case, it is much more effective to solve the problem of fine mixing and dispersion using simplified theoretical approaches using limited numerical calculations, as well as conducting field experiments.

When creating a theoretical model of wave generation, the results published in monographs [43, 44] were used.

For practical purposes, it is sufficient to limit ourselves to fluctuations and waves arising in currents containing highly compressible (vapor-gas) zones, as well as zones with high gradients of velocities and pressures. It is assumed that the crushing of droplets of the dispersed phase in this case will occur both due to their passage through zones with high gradients of velocities and shear stresses in a viscous medium, and due to the effects of hydrodynamic and acoustic high-frequency vibrations and waves, leading to the collapse of combined-cycle gas cavities and bubbles.

To calculate hydrodynamic cavitation-wave apparatuses, a mathematical model was used to study cavitation processes occurring in CVA [45]. In this model, a system of Navier-Stokes continuity equations for an axisymmetric flow, a two-parameter turbulence model written in a cylindrical coordinate system, and the equation of vapor mass fraction transfer are jointly solved. Its description is given in [46].

Various types of wave-flow dispersing and mixing apparatuses have been studied: with direct-flow liquid flow, vortex-type apparatuses with swirling liquid movement, plane-type hydrodynamic apparatuses with flow bodies and combined CVA of complex geometry using the energy of the vortex motion of the liquid [43]. The study of the flow patterns of liquids under various

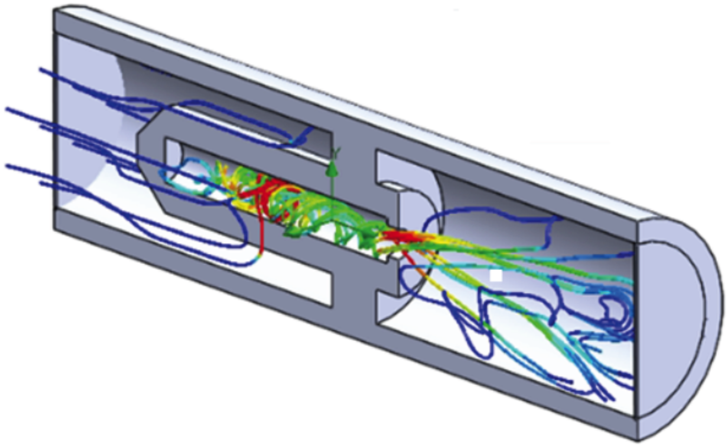


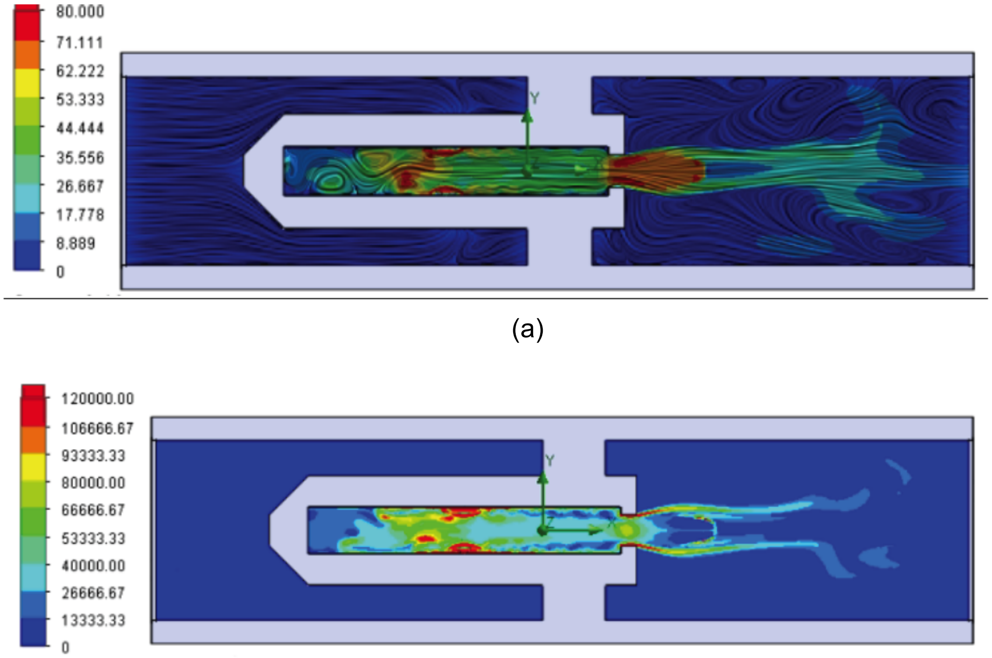
Figure 1: Calculation model of vortex type CVA.

turning conditions and mechanisms that affect the internal characteristics of the flow are important in applied problems [47].

One of the types of developed CVA is a vortex type cavitation-wave dispersing device. Its calculation model is shown in Figure **1**. The device is a cylindrical channel of a certain length, into which a working fluid is supplied tangentially through the holes. Under certain conditions, a break in the continuity of the medium may occur on the axis of the swirling flow under the action of centrifugal forces with the formation of a precessing steam-gas cavity. A nozzle is located at the end of the cylindrical channel.

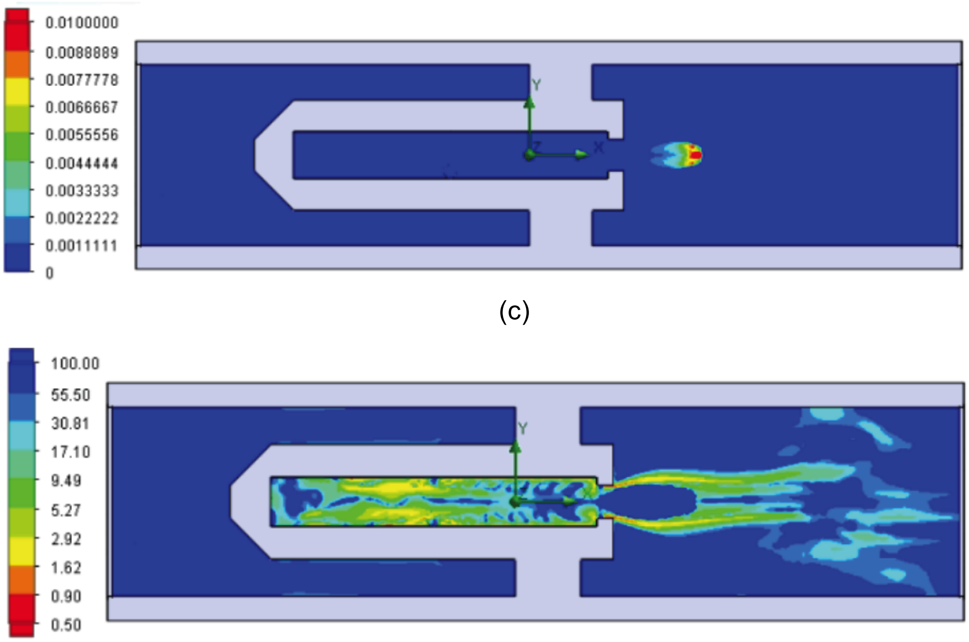
The simulation was performed at an inlet pressure of 100 atm and a static outlet pressure of 2 atm. The geometric parameters of the device were varied during the calculations to find the optimal design.

In the process of theoretical modeling, the following flow characteristics were studied: velocity and pressure fields, velocity gradients – vorticity (characterizes vortex formation and shear gradient), shear stresses (deformation rates) of viscous forces, and the diameter of a dispersed bio-oil droplet. The obtained distribution patterns of the marked parameters for one of the geometry variants are shown in Figure **2**.



(a)

(b)



(c)

(d)

Figure 2: Pictures of numerical calculations in the longitudinal section of the vortex-type CVA: **a**) velocity fields (m/s) and liquid flow lines; **b**) generator vorticity fields (1/s); **c**) volumetric content of the vapor-gas phase; d) minimum diameters (in the figure, the scale is logarithmic, in micrometers) of droplets of the dispersed phase of the bio-oil emulsion in fuel oil (without SF).

In Figure **2** it can be seen that the most intensive fragmentation of droplets due to the tangential stresses of viscous forces occurs in the region of inflow of tangential jets, as well as near the outlet nozzle, where the peripheral velocity of the flow increases sharply.

The theoretical minimum size of bio-oil dispersion in fuel oil without using SF under given design conditions and studied geometry variants can reach a minimum of about 0.5 μm. This occurs in the feed channel zone and near the nozzle as a result of shear deformations acting on a drop of bio-oil in fuel oil.

Расчётное время активной обработки эмульсии в CVA вихревого типа составляет порядка 30 мс при расходе жидкости 40 LPM.

The estimated active processing time of the emulsion in a vortex type CVA is about 30 ms at a liquid flow rate of 40 LPM.

As a result of mathematical modeling and numerical calculations of viscous media flows in flow-through wave dispersing devices of the vortex type, their optimal geometry was determined from the point of view of mixing and dispersing effects.

4.2. Field Studies

Taking into account the results obtained at the previous stage, CVA were manufactured and studies were conducted to optimize the parameters of the components of the mixed fuel and the required concentration of SF, as well as the modes of their processing. A series of experiments was conducted to prepare mixed fuel with different concentrations of bio-oil.

The processing was carried out in two stages. At the first stage, a low-frequency wave resonance mixing mode was performed. For this purpose, the components of fuel oil and bio–oil were loaded into the reactor, where they were mixed by rotating the mixer at a speed of 50-100 rpm while the reactor itself oscillated with a frequency of 50 Hz and an angular amplitude of 5°. The processing time was 3-7 minutes.

The resulting pre-mixed fuel sample was then evaluated using a Zeiss Axio Scope optical microscope using the bright field transmission method. It showed that already at this stage a sufficiently high degree of dispersion is achieved (the size of bio-oil droplets is 0.5 - 20 microns).

The next stage involved cavitation-wave treatment of the pre-obtained emulsion in the fuel circulation mode through a vortex-type CVA. The treatment time was from 1 to 3 minutes. The liquid was heated only by the operation of the pump and the hydrodynamic cavitation-wave apparatus.

Three series of experiments were performed to obtain mixed fuel in the following proportions of the mass ratio of fuel oil and bio-oil: 95:5, 85:15 and 50:50. In the second and third series of experiments, SF was added to the emulsion in a concentration of 0.05 and 0.5% mass %, respectively.

Figure **3** shows microphotographs of the obtained samples of mixed fuel.

It is evident from the figures that:

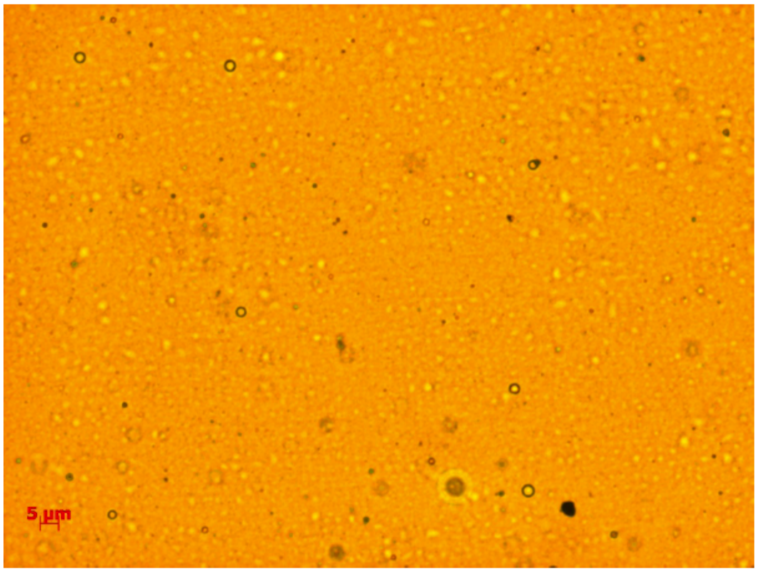
After processing on CVA, the size of the droplets of the dispersed phase of bio-oil decreases by an order of magnitude compared to the first stage of processing and becomes submicron (Figure **3.a**).

Bio-oil is in the fuel oil in a dispersed-colloidal state (Figure **3.b**).

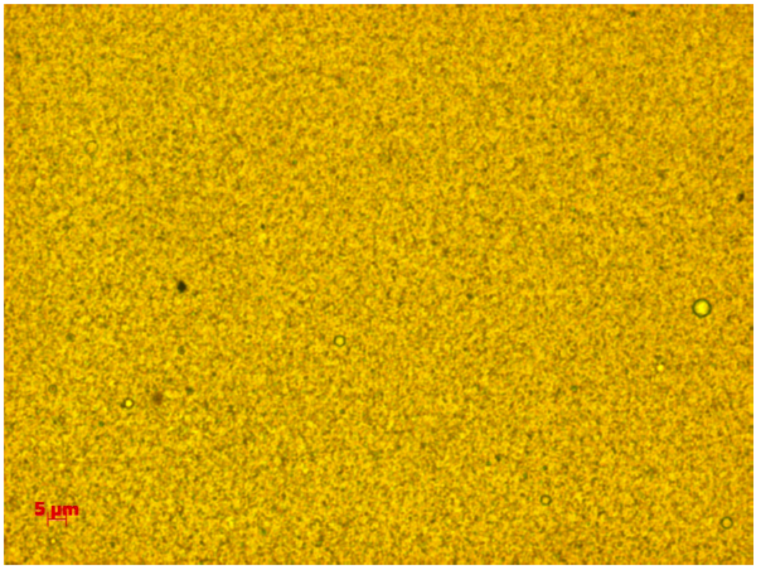
After processing, the resulting composition acquires the properties of a continuous emulsion, when the dispersed phase (bio-oil) and the dispersion medium (fuel oil) are distributed along thin channels of irregular geometry, separated by a layer of SF (Figure **3.c**).

5. DISCUSSION OF THE RESULTS

Theoretical and experimental studies have shown that hydrodynamic cavitation-wave devices can produce mixed emulsion fuels with a concentration of bio-oil in fuel oil up to 20% by weight. without using SF



(a)



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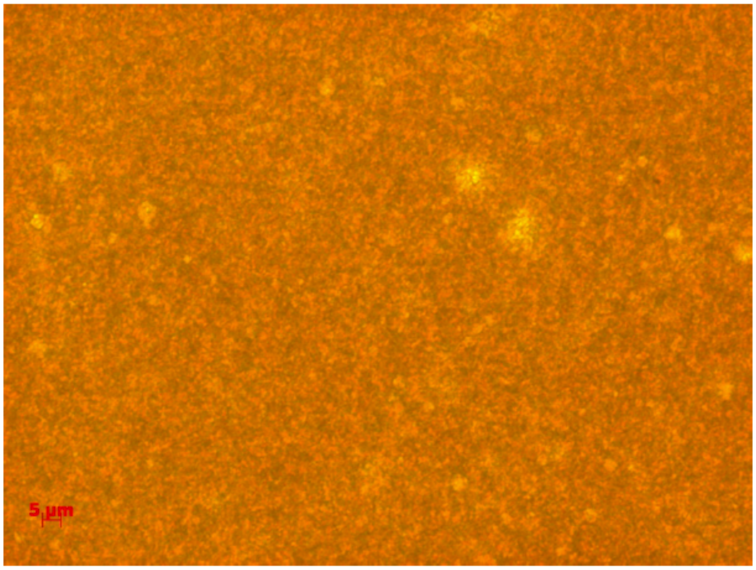

(c)

Figure 3: Optical micrographs with 500x magnification of fuel samples obtained after processing at CVA with the following component ratio: **a**) 95:5; **b**) 85:15+0.05%; **c**) 50:50+0.5%.

and up to 50 wt. % with the addition of SF at a concentration of 0.5% by weight. They have a high degree of stability. After 6 months of storage of the obtained mixed fuels at temperatures of 18-25 °They have not been stratified into components and the overall picture of dispersion has not changed dramatically. A small part of the biooil droplets coagulated, while the maximum droplet size did not exceed 5 microns.

The stability of the obtained mixed fuels is explained by a number of reasons:

Heavy oil residues, including fuel oil, contain asphaltenes, resins, carbenes and other components, which are a kind of natural emulsifiers: on the one hand, they reduce the surface tension force at the phase boundary, and on the other, they stabilize the droplets of the dispersed phase due to the formation of a strong adsorption-solvate layer at the interphase boundary. The chemical and rheological properties of fuel oils are significantly higher than those of other petroleum fuels. In this regard, the resulting level of dispersion and homogeneity of bio-oil mixtures with fuel oil is higher than that of mixed fuels based on other petroleum components.

The acidity of bio-oil contributes to increased stability: in an acidic environment, the adsorption layers on the surface of the droplets are rigid, solid-like and, accordingly, water-in-oil emulsions are more stable.

The high viscosity of fuel oil also contributes to increased kinetic stability of the fuel.

To obtain mixed fuels with a bio-oil concentration in fuel oil of more than 20% by weight, effective surfactants were selected. SF based on a mixture of non-ionic polyglyceryl polyricinoleate and polysorbate showed the best efficiency in stabilizing fuel emulsions. In this case, SF on this basis must be introduced into the oil fraction in a concentration not exceeding 0.5% by weight.

The production of mixed fuels on CVA is possible in two modes: in a flow mode in one pass through the disperser and in a circulation mode.

The developed laboratory CVAs provide a productivity of about 40 l/min when producing emulsion fuels in a flow mode in one pass through the disperser.

If the concentration of bio-oil in the mixed fuel based on fuel oil exceeds 15 wt.%, it is necessary to implement a circulation mode of processing on CVA to increase the degree of dispersion and stability of the emulsion.

6. CONCLUSION

The results of the conducted studies showed the high efficiency of CVA in obtaining emulsion mixed fuels with products of fast pyrolysis of wood.

The hydrodynamic dispersers and mixers used in this work were created on the basis of the theory of nonlinear wave mechanics of multiphase media and were developed at the Branch of Mechanical Engineering Research Institute of the Russian Academy of Sciences (IMASH RAN) – Research Center for Nonlinear Wave Mechanics and Technology (NWMTC)

Taking into account scaling, CVA can be recommended for use in industrial conditions to produce emulsion mixed fuels.

CONFLICT OF INTEREST

No conflict of interest.

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