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Use of Tyre Pyrolysis Products as Recycled Raw Materials for the Production of Composite Suspension Fuel

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Abstract

Composite suspension fuel (CSF) is a viable alternative not only to solid coal but also to fuel oil and diesel fuel. The issue with combined systems is the low stability and heterogeneity of the distribution of solid particles, which causes an increase in the viscosity of dispersed systems. Studies have shown that it is possible to use pyrocarbon, which is made from used car tyres, as a fuel for boats. Systems based on pyrocarbon without additives have high viscosity values. To regulate the rheological properties and stabilize the CSF, additives such as dispersants, plasticisers, and stabilisers are used. Research has been done into how dispersants affect the properties of composite suspension fuel (CSF). They used additives like carboxymethylcellulose (CMC), sodium lignosulfonate (LSTNa), naphthalene formaldehyde (NF), the condensation product of β-naphthalene sulfonic acid and formaldehyde (C-3), and humic acids. Studies of the flow of CSF based on pyrocarbon are in line with studies of the surface of pyrocarbon using electrokinetic methods. It is estimated that the surface of unmodified pyrocarbon has a positive charge at a pH of 2-10. The addition of reagents causes a change in the surface charge of the pyrocarbon and surface recharging. The best results have been obtained in CSF formulations with additives containing a sulfur group, such as C-3, NF, and LSTNa in a mass concentration of 2%. The resulting suspensions have an apparent viscosity of no more than 0.8 Pa·s, which makes it possible to use the obtained CSF as a liquid fuel suspension with a solid phase concentration of up to 60% by mass.

1. Introduction

The disposal of used car tyres is a significant environmental concern. In industrialised countries, on average, there is one used car tyre per person per year. One of the most common methods of recycling tyres is their pyrolysis [1,2]. The rubber components are transformed into a mixture of volatile and liquid thermal degradation products in this process. The solid pyrolysis residue is pyrocarbon, which forms in approximately 30% of the mass of the raw material. Pyrocarbon is made up of amorphous carbon, has a porous structure, and is most often used as a rubber filler. However, the problem is that its properties are not stable. This means that they depend on the composition of the raw material and the pyrolysis technology. It is quite difficult to get a product with stable properties during thermal destruction [3]. In addition, pyrocarbon often contains a significant amount of ballast impurities of inorganic nature. This means that much of the carbon produced during the process is unused. Pyrocarbon is not as easy to use as coal because it is more porous, it ages quickly, and it cakes easily. However, it can be used as a fuel on its own or added to other hydrocarbon suspensions. This kind of energy carrier is a real alternative to coal and liquid (fuel oil, diesel fuel) and gaseous fuels [4]. Another advantage of this approach is that it can be used as a dispersion medium for liquid distillates, which contain water-organic mixtures and are removed from the pyrolysis process as wastewater. Preparing suspension fuel from these liquids stops them from being put into water sources used in industry and homes, and increases the energy value of the fuel [5].

Due to the high degree of dispersity, the presence of a significant number of submicron carbon particles, and the much higher porosity compared to coal, pyrocarbon is advisable to use as an additive in the production of composite suspension fuel (CSF). Its advantages are a much better degree of carbon combustion (residue < 0.04%), and a reduction of atmospheric pollution (SOx, NOx). This is a consequence of the catalytic activity of finely dispersed carbon [5-7].

To be successfully used in energy-generating devices, CSF must meet several technological and operational requirements that ensure its storage, transportation, and burning. The main ones are apparent viscosity, which should not exceed 1.5-1.6 Pa·sec, and sedimentation resistance, which should be 2-3 days under the condition of direct preparation before burning and 10-14 days under the condition of transportation and storage of CSF.

The problem of combined systems of the CSF type is the low stability and inhomogeneity of the distribution of coal particles in organic liquids, which leads to an increase in the viscosity of dispersed systems, therefore, regulating the rheological properties and stabilise the CSF, additives of chemical reagents (dispersants, plasticisers, stabilisers) are used [5,8,9]. The most widely used additives include sodium, calcium, and magnesium lignosulfonates, naphthalene sulfonic acids, sulfonated melamine formaldehyde, and polycarboxylates (C-3, NF, Dofen, Melment, Melflux), which effectively improve the properties of CSF. Emulsifiers should also be introduced for organo-mineral systems. Non-inorganic surfactants have proven themselves well in this capacity. The use of anionic surfactants should include control of the pH of CSF since surfactants are effective at high pH. The problem of combined systems of the CSF type is the low stability and inhomogeneity of the distribution of coal particles in organic liquids, which leads to an increase in the viscosity of dispersed systems, therefore, regulating the rheological properties and stabilise the CSF, additives of chemical reagents (dispersants, plasticisers, stabilisers) are used [5,8,9]. The most widely used additives include sodium, calcium, and magnesium lignosulfonates, naphthalene sulfonic acids, sulfonated melamine formaldehyde, and polycarboxylates (C-3, NF, Dofen, Melment, Melflux), which effectively improve the properties of CSF. Emulsifiers should also be introduced for organo-mineral systems. Non-inorganic surfactants have proven themselves well in this capacity. Anionic surfactants are effective at high pH 7-9, which is why the pH of CSF should be controlled when using them.

Therefore, the purpose of the work was to study the possibility of using technical pyrocarbon in CSF formulations as a carbon-containing energy carrier for its partial or complete replacement of coal. The nature of the pyrocarbon surface, the pH of the dispersion medium, and the effect of various plasticisers and stabilisers on the rheological properties of technical pyrocarbon-based CSF were also studied.

2. Materials and Methods

At the first stage, the chemical composition of samples and surface groups was studied, and the technical, structural, and sorption characteristics of “technical pyrocarbon” as a solid fraction of the product of tyre pyrolysis - in the further is refer to as " pyrocarbon".

The sample of pyrocarbon belongs to medium-ash materials and has a high content of volatile substances and a high calorific value as shown. According to the ash content (A^d = 19.5%), the pyrocarbon sample does not meet the technological parameters of suspensions. A process called oil agglomeration has been used to reduce the ash content of pyrocarbon. Technical characteristics of pyrocarbon from tyre pyrolysis are presented in Table 1.

Table 1 *Technical characteristics of technical pyrocarbon*

Component	Component content (mass.%)	Method of determination
Humidity, W^a (%)	2.38	DSTU EN 14774-1
Ash content, A^d (%)	19.5	DSTU EN15403, ISO 1171
Volatile substances, V^{daf} (%)	18.3	DSTU EN15148:2012
Higher heat of combustion in the dry state, Q_c (kcal/kg)	6678	DSTU ISO 1928, calorimeter IKA C2000
Higher heat of combustion for the working state, Q_p (kcal/kg)	6519	

The chemical composition of the surface groups of pyrocarbon was determined by IR spectroscopy on the device "Avatar 370FT-IR, Thermo Nicolet" in the range of 4000-400 cm^{-1} (Fig. 1) on Zn-Se glass (dry film). Iso-C_3H_7OH, ortho-$C_6H_4(CH_3)_2$ and C_7H_{14} were used as extractants. Samples were obtained by extraction of organic substances with pyrocarbon at the rate of 10% pyrocarbon for the entire system for 7 days at a temperature of 20°C with daily stirring for 2-3 minutes. The mass loss of pyrocarbon samples after filtration, drying at 110 °C, and for 1 hour was 11.97%, 14.0% and 10.87%, respectively. To identify the components, the obtained spectra were compared with the reference and standard spectra of the instrument database (EZ Omnic v. 7.0 programs) and the spectra given in the literature data [10].

The spectra of extract samples practically do not differ despite the different polarity of the solvents. Fig. 1 shows that the spectrum has absorption bands characteristic of aliphatic compounds (2925cm^{-1}, 2856cm^{-1}, 1456cm^{-1}, 1376cm^{-1}, 754cm^{-1}), carbonyl compounds (1703cm^{-1}), aliphatic sulfides (1076cm^{-1}, 699cm^{-1}), as well as weakly expressed absorption bands mass loss of pyrocarbon samples after filtration and drying of aromatic compounds (1601cm^{-1}, 1032cm^{-1}, 877cm^{-1}). These data are consistent with the spectra obtained for coals [11].

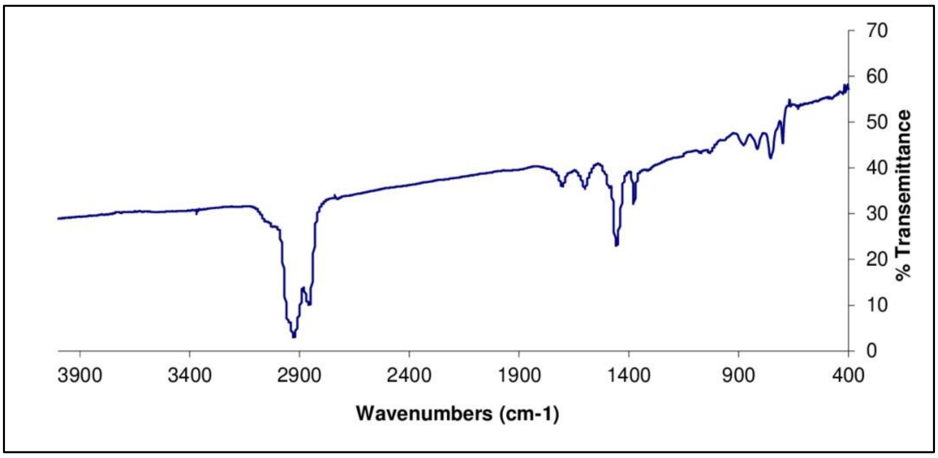

Fig. 1 *IR spectrum of a* sample *of technical pyrocarbon*

Structural and sorption characteristics of mesopores (2-50 nm) of pyrocarbon: specific surface area (Ssp=35.3 m^2/g), specific pore volume (Vpore=0.0365 cm^3/g) and effective pore diameter (dpore =26.8 nm) were determined by the DFT method based on low-temperature N_2 adsorption data (77 K) on a Quantachrome Autosorb specific surface area analyser with subsequent data processing by Quantachrome Instruments software. v. 3.2. The number of surface functional groups: COOH - 0.0085 mg-eq/g; OH - 0.0206 mg-eq/g; >C=O - 0.00955 mg-eq/g) was determined by titration according to Boehm with 0.05 M NaOH, Na_2CO_3 and $NaHCO_3$ [12], the number of basic groups was determined according to [13] and was 0.053 mg-eq/g.

The apparent viscosity (η) of the samples was measured on a Rheotest-2 device at t=20^{0}C. The range of measurement of shear rates (D_r) is limited to values of 1 – 250 sec^{-1}. Determination of the value of zeta–potential (ξ) was carried out at the installation and according to the method given in [14]. To achieve the required pH values, 0.1 M solutions of NaOH and HCl were used. The sedimentation stability St was studied at the time of delamination of the CSF sample in the measuring cylinder.

In the second stage, the influence of the concentration of dispersants, stabilisers, and plasticisers on the rheological properties of pyrocarbon suspensions was studied. To get the CSF, the pyrocarbon powder was mixed with a dispersion medium.

Most particles are very small, so the best way to collect them is to use something called oil agglomeration. [15]. The main advantages of the oil agglomeration method include being able to separate particles smaller than 100 µm, which takes into account a wide range of ash content of the enriched material.

First, pyrocarbon is mixed with industrial water for 1 minute using a paddle mixer. After that, the liquid fraction of tyre pyrolysis was added in the amount of 10% to the weight of the carbon residue and stirred for another 6 minutes. As a result of pulp tribulation, pyrocarbon aggregates are formed, which are compacted into spherical granules. At the same time, the ash content of the concentrates did not exceed 10-12% by mass, which satisfies the technological standards and allows them to be used for the preparation of CSF. The obtained concentrate with granules of 2-3 mm is separated from water and mineral impurities on sieves with cells of 0.5 mm.

Then wet grinding is carried out in a ball mill lasting 5-10 minutes with the addition of chemical reagents. Wet grinding was carried out to particles < 0.25 mm in size. CSF with a solid phase concentration of 50-52% was obtained.

As chemical reagents, we used CMC - carboxymethylcellulose Sigma-Aldridge (CAS: 9004-32-4), C-3 - condensation product of β-naphthalene sulfonic acid and formaldehyde (TU 5870-005-58042865-05), humic acids Sigma-Aldridge (CAS: 1415-93-6), OP-10 (product of processing a mixture of mono- and dialkylphenols with ethylene oxide which has 10 degree of ethoxylation). LSTNa – sodium lignosulfonates (TU - 2455-002-00281039-00) and NF – condensation product of β-naphthalene sulfonic acid and formaldehyde (TU 5870-005-58042865-05).

3. Results and Discussion

When obtaining highly concentrated suspensions of the CSF type, the decisive factor in their application is the compliance of their characteristics, in particular viscosity (less than 1.5 Pa·s) and sedimentation resistance (at least 7 days) with the necessary operational requirements. Aggregative and sedimentation stability in systems with submicron particles of the dispersed phase of pyrocarbon is largely provided by electrostatic repulsion. Electrokinetic potential (ζ-potential) is used to characterise electrostatic interactions, which occur at the sliding boundary between the adsorption and diffusion layers. Electrokinetic processes that occur at the phase interface between the dispersed phase and the dispersed medium affect the contact interactions between the components of the dispersed system and determine its stability and rheological properties [5]. Since the sliding plane can be located at a different distance from the interphase surface, which is characterised by certain physicochemical properties, the value of the ζ- potential depends on all these factors. To determine the effect of pH on the value of ζ-potential, highly concentrated suspensions based on pyrocarbon were obtained. The nature of the curves of the dependence of the ζ-potential on pH for samples without the addition of anionic reagents is positive in the pH range from 2 to 10 (Fig. 2, curve 1).

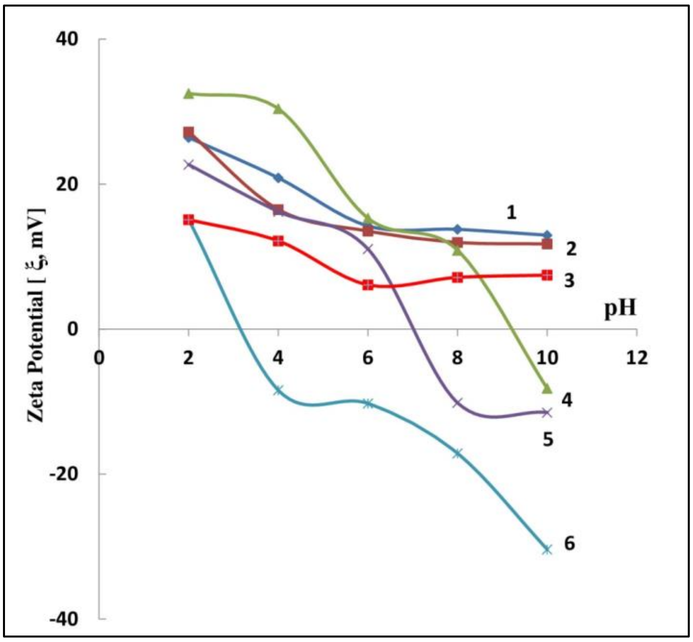

Fig. 2 *Dependences of the ζ- potential of CSF based on pyrocarbon pyrolysis of tyres with the addition of various additives on the pH of the dispersion medium: 1- without additives, 2- 10% liquid pyrolysis, 3- 1% OP-10, 4- 0.1% CMC, 5- 2 % HA (humic acids), 6 - 2% NF*

As pH decreases, the positive surface charge of pyrocarbon increases. In a strongly acidic environment, the absolute values of the ζ- potential reach 30-40 mV, which can provide the desired stability for submicron particles. Unfortunately, a strongly acidic environment does not meet the operational requirements for CSF, as it will inevitably cause increased corrosion of fuel equipment, pipelines, and liquid fuel storage tanks. Dispersants based on anionic surfactants should be added to the pyrocarbon-based CSF composition to regulate surface properties. The surface-active anion of such compounds is adsorbed by the positively charged active centres of the pyrocarbon surface and effectively recharges the surface. In the presence of additives such as surfactants and pH adjustment, the desired surface properties of the particles of the dispersed phase can be achieved.

In Fig. 2 it is demonstrated that adding anionic dispersants recharges the surface of pyrocarbon, with this effect being more pronounced at higher pH levels. This phenomenon is linked to the increased dissociation of anionic surfactants and a rise in the surface activity of anions.

Another benefit of using dispersants is the creation of a solvate layer around particles, composed of polar H_2O molecules that are adsorbed with surfactants. This adsorption-solvate layer, as illustrated in Fig. 3, possesses both elasticity and strength, effectively preventing the aggregation of pyrocarbon particles. It helps ensure the desired stability of the colloidal system even when the ionic-electrostatic stabilisation factor is insufficient.

Moreover, in the presence of larger and branched surfactant and polyelectrolyte molecules, steric stabilisation is further enhanced. This enhancement occurs because additional energy is required to compress the structured layers of water and polyelectrolytes located between the particles.

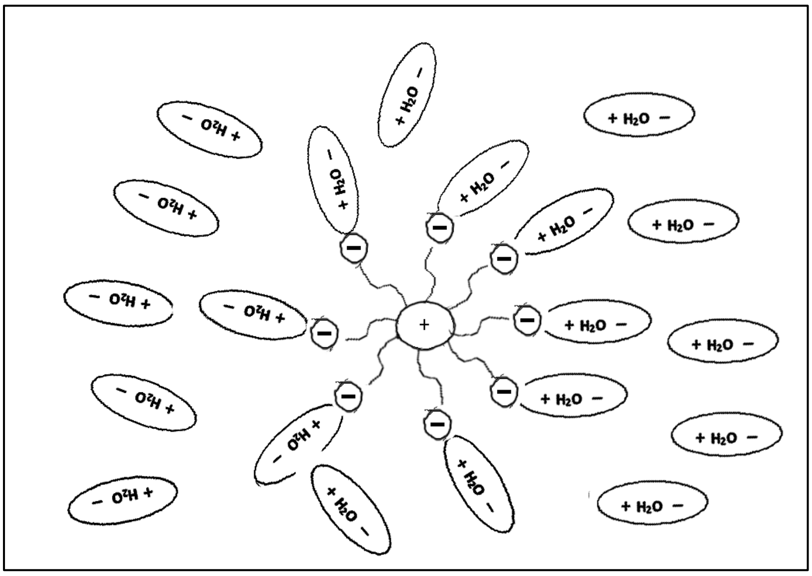

Fig. 3 *Recharging of the surface of a pyrocarbon particle and the formation of an adsorption-solvate layer around a pyrocarbon particle in the presence of anionic dispersants*

The stability of suspensions according to the DLVO theory [16] is determined by the balance between van der Waals forces, which ensure particle attraction and electrostatic repulsion. The surface of pyrocarbon has hydrophobic properties (wetting angle > 90°), so the classic theory of DLVO cannot be applied here due to the significant predominance of attractive forces, due to the instability of the wettability of the surface of particles of the dispersed phase. To correct this, together with dispersants, it is desirable to use hydrophobic surface-wetting agents. Several compounds tested by us have both dispersing and wetting properties.

In the initial experiments, it was established that suspensions with pyrocarbon concentration > 40% are unsuitable for use due to strong thickening. Such systems do not show signs of flow under the influence of external forces. Therefore, to give them properties of stable fluidity, modification with the help of chemical reagents should be used.

To select the most effective plasticisers and stabilisers of CSF, the effect of various reagents with a concentration of 0.5-2% by mass was studied on dynamic viscosity: LSTNa, NF, CMC, C3, OP-10, sodium humate. CSF has the lowest viscosity with plasticisers LSTNa, NF, and C3, which is 0.26 - 0.39 Pa·sec, as shown (Table 2). Other systems were obtained as a paste with high viscosity values and were not used in further studies. In Figure 2 it is shown that the addition of anionic dispersants alters the surface charge of the pyrocarbon, with this effect being more pronounced at higher pH levels. This phenomenon is linked to the increased dissociation of the anionic surfactants and an increase in the surface activity of the anions.

Another advantage of using dispersants is the formation of a solvate layer around the particles, composed of polar water (H_2O) molecules adsorbed by the surfactants. This adsorption-solvate layer, as illustrated in Fig. 3, exhibits both elasticity and strength, effectively preventing the aggregation of pyrocarbon particles. It helps to maintain the stability of the colloidal system, even when ionic-electrostatic stabilisation alone is insufficient.

Additionally, the presence of larger and branched surfactant and polyelectrolyte molecules enhances steric stabilisation. This improvement occurs because more energy is required to compress the structured layers of water and polyelectrolytes situated between the particles.

However, systems with the addition of a plasticiser do not provide sedimentation stability. It is known that sodium humates are used as a plasticiser for the production of CSF based on the coal of various degrees of metamorphism, as well as pyrocarbon [17]. However, in our systems with pyrocarbon, the sedimentation resistance with sodium humates was insufficient (1-3 days). As can be seen from Table 2, an increase in the concentration of sodium humate, in the range from 1% to 3%, does not affect the rheological properties of CSF (with a solid phase share of 52%) and can be used more as a suspension stabiliser.

Table 2 *Rheological properties of CSF based on pyrocarbon at* different *concentrations of rheological* additives *(pyrocarbon content 52%)*

Rheological additive with different concentrations (% mass)	Apparent viscosity (Pa·s)	Sedimentation resistance (days)
2% C3	0.36	<1
2% LSTNa	0.26	<1
2% NF	0.39	<1
1% HA	0.39	
2% HA	0.39	1-3
3% HA	0.39	
2% NF+0,1% CMC	0.49	5-7
1% OP-10	2.63	25-30
2% HA+1%OP-10	0.73	25-30

The addition of just stabilisers CMC and OP-10 in the amount of 0.1% and 1%, respectively, without a plasticiser was ineffective due to the high viscosity values of CSF. As can be seen from Table 2, sodium humate in combination with OP-10 provides sufficient stability of CSF and can be recommended as stabilisers for obtaining composite suspension fuel based on technical pyrocarbon.

As shown (in Fig. 4), the flow curves of CSF based on pyrocarbon in the presence of various reagents have a significant deflection in the region of Dr – 0 - 15 sec^{-1}, which indicates that all systems have a pseudo-plastic nature of the flow. The deflection on these curves proves the formation of a structured system, and the greater the deflection, the more stress must be applied to destroy such a structure. The most structured system is CSF with 2% NF and 0.1% CMC.

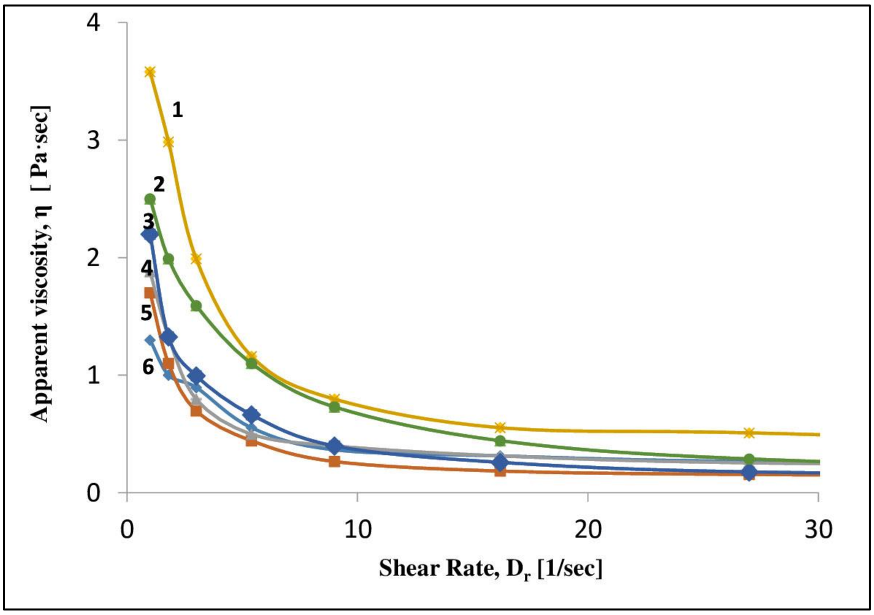

Fig. 4 *Curves of the dependence of the apparent viscosity (ή) of CSF on the shear rate (D_r) with different dispersants with a concentration of: 1-2% NF; 0.1% CMC, 2-2% HA; 1% OP, 3 – 2% HA, 4- 2% NF, 5- 2% LSTNa, 6- 2% C3*

In Fig. 4 it can be seen that the viscosity of CSF decreases with an increase in shear stress. For all studied samples, when Dr reaches 15-20 sec^{-1}, the structure collapses, and when Dr > 20 sec^{-1} increases, it acquires a Newtonian fluidity. The difference between the effectiveness of additives is not very noticeable, but it can be noted that the NF additive reduces the apparent viscosity the most. Therefore, further research has focused on the study of this dispersant.

Most pseudoplastic structures are characterised by the loss of structure under the influence of high shear rates and the restoration of the initial high viscosity when high loads are removed and at rest. This phenomenon of thixotropy is characterised by the hysteresis of yield curves and is characteristic of CSF, the dispersed phase of which is particles of the solid phase. Fig. 5 shows the flow curves with a gradual increase and subsequent decrease in the shear rate for pyrocarbon-based CSF with various additives.

As can be seen from the data (Fig. 5, curve a), the pseudoplastic nature of the flow is observed for additives based on sulfonated naphthalene formaldehyde (NF and C3). Pseudoplastic flow can also include a sample of CSF with LSTNa. For the sample with HA (Fig. 5, curve b), the curve is close to the Newtonian flow adding HA to the pyrocarbon-based system does not lead to noticeable structuring of the system. The behaviour of the systems, when the shear rate decreases, is characterised by an almost constant angle of inclination of the rheological curve and the Newtonian nature of the flow.

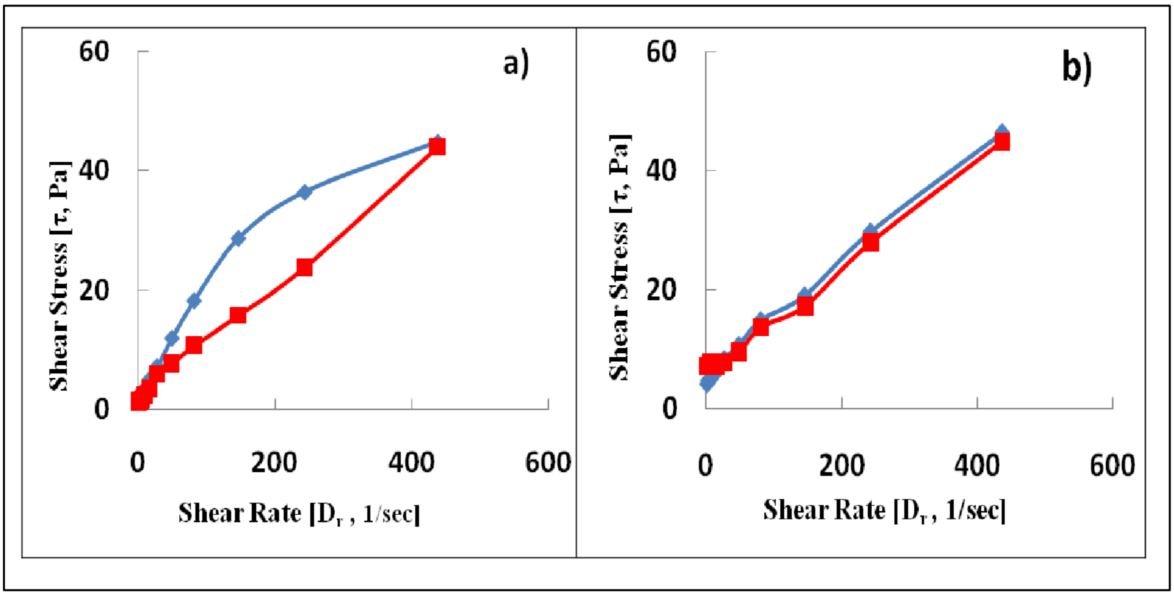

Fig. 5 *Fluidity curves of CSF based on pyrocarbon with various additives (blue curve – increase in Dr, red curve - decrease in Dr) with a concentration of 2% by mass: a) NF, b) HA (humic acids)*

The rheological curves for systems with the addition of NF, C3 and LSTNa are satisfactorily described by the Oswald de Wiel equation:

$$\tau = KD^n \quad (1)$$

Where: *τ* - shear stress; *K*- coefficient of structuring; *D* - shear speed; *n* is the flow index (*n<1* for pseudoplastic structures).

For the curves with C3 and NF additives, n = 0.43 and 0.68, respectively, which characterise the CSF data as pseudoplastic. In contrast to coal-based systems, with a fairly quick recovery of the structure when the shear load is removed, CSF with pyrocarbon recovers much more slowly and only in a state of rest, or at low (Dr < 20 sec^{-1}).

From a practical point of view, this means that CSFs with pyrocarbon are significantly more sensitive to mechanical loads than systems with coal. Much more time is needed to restore their destroyed structure. Fig. 6, shows the dependence of the apparent viscosity of CSF as a function of the concentration of the NF additive, chosen as the most effective, and it decreases with increasing concentration of the NF dispersant.

All curves indicate the pseudoplastic nature of the flow of systems with NF additives. After reaching Dr ~ 20 sec^{-1}, the pseudoplastic nature of the flow changes to the Newtonian one, and the structure of the CSF is destroyed. Therefore, in the practical application of CSF based on pyrocarbon, it is necessary to avoid the effect of the suspension under too high mechanical loads (Table 3).

As can be seen from Table 3, all of the above systems demonstrate the pseudoplastic nature of the flow under moderate loads and a rapid decrease in η as the shear rate increases. At Dr = 9 sec^{-1}, which is usually used to compare the operational characteristics of water-coal fuel [5], η does not exceed 0.8 Pa·s. This is a satisfactory indicator for liquid suspension fuel and has room for improvement until the concentration of pyrocarbon in CSF increases to approximately 60% by mass.

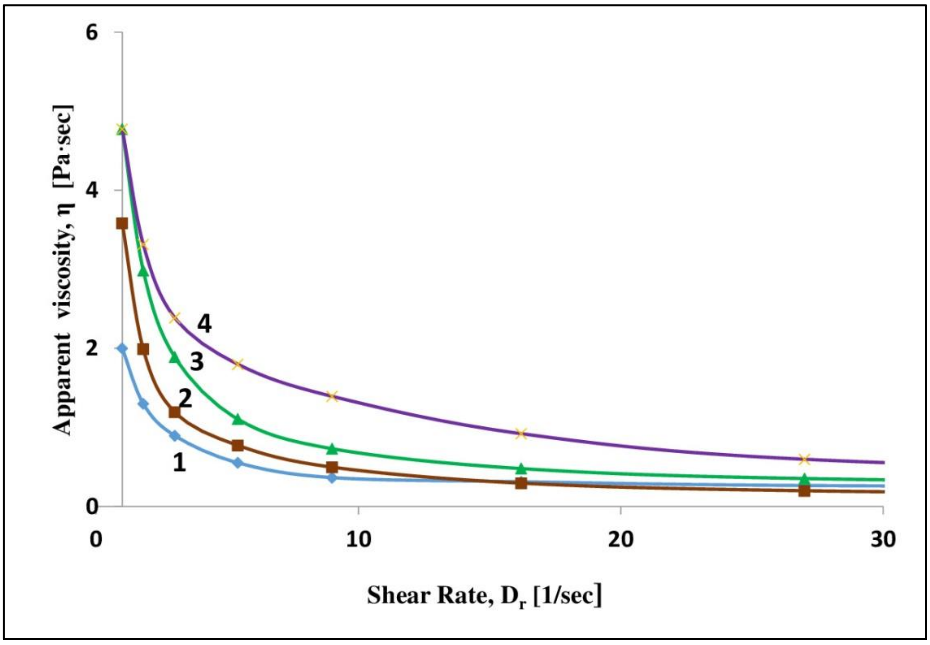

Fig. 6 *Curves of the apparent viscosity (ή) of CSF at different values of the shear rate Dr for different concentrations of NF dispersant: 1- 2%, 2- 1%, 3- 0.5%, 4- 0.1% (pyrocarbon content in the suspension 52%)*

Table 3 *Apparent viscosity (ή) of CSF based on pyrocarbon, has been obtained by adding rheological additives with different concentrations in % by mass, at different shear rates (Dr) (pyrocarbon content 52%)*

Shear rate, Dr (sec^{-1})	Apparent viscosity (ή) when various rheological additives are added, Pa·					
	Concentration of rheological additive, % by mass					
	2% C3	2% LSTNa	2% NF	2%NF; 0.1% CMC	2% HA	2% HA; 1% OP
1.0	1.194	1.791	1.791	3.582	2.388	2.388
1.8	0.829	1.161	1.327	2.985	1.327	1.990
3.0	0.896	0.697	0.796	1.990	0.995	1.592
5.4	0.553	0.442	0.498	1.161	0.663	1.106
9.0	0.365	0.265	0.398	0.796	0.398	0.730
16.2	0.313	0.184	0.313	0.553	0.258	0.442
27.0	0.265	0.155	0.254	0.509	0.177	0.287
48.6	0.246	0.135	0.221	0.405	0.147	0.184
81.0	0.225	0.118	0.155	0.369	0.125	0.170
145.8	0.197	0.106	0.127	0.246	0.102	0.139
243.0	0.150	0.068	0.095	0.177	0.088	0.111
437.4	0.102	0.051	0.063	0.138	0.056	0.074

4. Conclusion

The study shows the possibility of using pyrocarbon, which has been obtained as a result of the pyrolysis of used car tyres, in composite suspension fuel (CSF). Since it has been established that the systems based on pyrocarbon without additives are not suitable for use as fuel due to thickening, the effect of additives of stabilisers, plasticizers and dispersants on the physicochemical properties of CSF is evaluated in this study. Carboxymethylcellulose, lignosulfonate Na, OP-10, NF and C3 - condensation products of β-naphthalene sulfonic acid and formaldehyde, and humic acids have been used as additives. The surface of unmodified pyrocarbon has a positive surface charge in the pH range of 2-10, as has been established. The addition of reagents causes a change in the surface charge of pyrocarbon and surface recharging. The best results when used in CSF formulations have been shown with the NF additive at a concentration of 2% by mass. The apparent viscosity of the resulting suspensions at a shear rate of 9 sec^{-1} does not exceed 0.8 Pa·s. This makes it possible to use the obtained CSF as a liquid suspension fuel with a solid phase concentration of up to 60% by mass.

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Conflict of Interest

The authors declare that there is no conflict of interests regarding the publication of the paper.

Author Contribution

The authors confirm contribution to the paper as follows: ***study conception and design:*** *Olena V. Shkutkova, Anatolii S. Makarov;* ***data collection:*** *Olena V. Shkutkova, Roman Evh. Klishchenko;* ***analysis and interpretation of results:*** *Olena V. Shkutkova, Roman Evh. Klishchenko, Oleksandr A. Konoval;* ***draft manuscript preparation:*** *Olena V. Shkutkova, Iryna M. Kosygina. All authors reviewed the results and approved the final version of the manuscript.*

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