PROPERTIES OF COMPOSITE WATER-COAL FUEL STABILIZED WITH CARBON SUBMICRON MATERIAL AND AMINO ALCOHOLS

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Abstract

Composite Water-Coal Fuel (CWCF) 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 coal particles, which causes an increase in the viscosity of dispersed systems. To regulate the rheological properties and stabilize the CWCF, additives such as dispersants, plasticizers, and stabilizers are used. The objective of the study was to examine the possibility of stabilizing composite water-coal fuel by adding amino alcohols and a submicron carbon substance (microdisperced carbon MC), obtained by plasma-chemical conversion of wastewater containing organics. The study focused on systems based on G-grade coal with a solid phase concentration of 61 %. Amino alcohols, 2-amino-2-methyl-1-propanol (AMP), and 2-amino-2-ethyl-1,3-propanediol (AEPD) were added to the CWCF at concentrations of 0.05 %, 0.1 %, 0.15 %, and 0.2 % by weight of CWCF. Its apparent viscosity was measured using a Rheotest-2 rheometer at a temperature of 20 °C, and the shear-stress/shear-rate data for the CWCF covered a range of 0.1 to 470 s^{-1}. The sedimentation stability of the CWCF was assessed using a water-separating test and a storage period measured in days. The study found that the ξ-potential of G-grade coal particles in the presence of amino alcohols is 40–45 mV in absolute value. The obtained CWCFs demonstrate a pseudoplastic type of flow in the range of shear rates of 0-80 s^{-1}. Systems with AMP are more stable than those with AERD. The introduction of highly dispersed carbon leads to an increase in the apparent viscosity of the systems and can be recommended for controlling the fluidity of the CWCF. To sum up, the sedimentation stability in the presence of highly dispersed carbon and amino alcohol additives increases from 5–6 to 10–14 days, almost twice. The sedimentation stability of these systems in the presence of MC makes the CWCFs suitable for short-term storage.

Keywords: composite water-coal fuel; coal; amino alcohol; carbon; sedimentation; viscosity.

ВЛАСТИВОСТІ КОМПОЗИЦІЙНОГО ВОДОВУГІЛЬНОГО ПАЛИВА, СТАБІЛІЗОВАНОГО ЗА ДОПОМОГОЮ ВУГЛЕЦЕВОГО СУБМІКРОННОГО МАТЕРІАЛУ ТА АМІНОСПИРТІВ

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Анотація

Композиційне водовугільне паливо (КВП) є реальною альтернативою не тільки твердому вугіллю, але може слугувати замінником мазуту та дизельного палива. Проблемою комбінованих систем є низька стабільність та неоднорідність розподілу частинок вугілля, що призводить до зростання в'язкості дисперсних систем, тому для регулювання реологічних властивостей та стабілізації КВП застосовують добавки хімічних реагентів (диспергаторів, пластифікаторів, стабілізаторів). Мета роботи полягала у вивченні можливості стабілізації композиційного водовугільного палива за допомогою добавок вуглецевого субмікронного матеріалу, отриманого шляхом плазмохімічної конверсії органовмісних стічних вод, та аміноспиртів. Досліджено системи на основі вугілля марки G з концентрацією твердої фази 61 %. У якості аміноспиртів застосовували 2-аміно-2-метил-1-пропанол (АМР) та 2-аміно-2-етил-1,3-пропандіол (AEPD). Аміноспирти додавали до КВП у концентраціях 0.05; 0.1; 0.15 та 0.2 % на масу КВП. Встановлено, що ξ – потенціал частинок вугілля марки G у присутності аміноспиртів становить за абсолютною величиною 40–45 мВ.. Отримані КВП демонструють псевдопластичний тип течії у діапазоні швидкостей зсуву 0–80 с$^{-1}$. Введення високодисперсного карбону веде до зростання ефективної в'язкості систем і може бути рекомендоване для регулювання плинності КВП. Седиментаційна стійкість в присутності добавок високодисперсного карбону та аміноспиртів зростає з 5–6 до 10–14 діб, тобто практично вдвічі.

Ключові слова: композиційне водовугільне паливо; вугілля; аміноспирт; карбон; седиментація; ефективна в'язкість.

Introduction

Liquid organic waste, often toxic and flammable, is a common byproduct of production. However, this waste can be turned into a potential source for fuel dispersion systems based on natural coal [1–4]. Composite water-carbon fuel (CWCF) is an alternative to solid coal, fuel oil, and diesel fuel. Unfortunately, research on CWCF has mostly focused on the inorganic components of the dispersion medium [5–7]. Adding organic components to CWCF can improve its energy value and help solve the problem of technogenic waste.

The problem of combined systems is the low stability and heterogeneity of the distribution of coal particles in organic liquids, which leads to an increase in the viscosity of dispersed systems [1–3; 8]. It is widely known that chemical reagents (dispersants, plasticizers, stabilizers), in addition to coal particle size distribution and natural coal characteristics (including mineral composition, coal porosity, and surface properties), are an important factor in improving the rheological properties and stability of CWCF. The most widely used additives include Na, Ca, and Mg lignosulfonates, naphthalene sulfonic acids, polycarboxylates, etc., which effectively improve the properties of the CWCF [9–11]. However, this kind of reagents gives limited performance because of its weak adsorption and steric hindrance effect on the coal–water interface [8; 11–13]. Therefore, the question arises of finding substitutes for these products.

Amino alcohols, widely used as dispersants and stabilizers in water-based paints, can serve as an alternative. They are low-toxic, relatively cheap, do not cause excessive foaming, and serve as anticorrosive agents and pH stabilizers [14]. Another way to improve the spatial structure is by introducing reactive particles like micro-dispersed carbon (MC) into the CWCF. Unlike polyelectrolytes and surfactants, MC can have different numbers of active centers. Therefore, by varying the properties of MC, an increase in the number of contact centers can be achieved without increasing the concentration of surfactants and polyelectrolytes [15].

To stabilize CWCF, highly dispersed carbon can be introduced into its composition to enhance the formation of stable spatial structures. Due to the catalytic properties of MC, it is also possible to increase the degree of burnout and calorific value of the fuel [16; 17].

However, in the presence of organic components, mechanochemical activation can be ineffective due to the shielding of active centers by large organic molecules. Thus, identifying the most effective stabilizing reagents and figuring out how to introduce them into the dispersed system is a crucial and complex problem in preparing CWCF.

Statement of the problem. The aim of this study was to investigate the possibility of stabilizing the CWCF based on G grade coal using MC and amino alcohols as additives.

Experimental

Objects and methods of research. G-grade coal from a Ukrainian deposit was used to prepare CWCF. The coal was first ground in a roller mill and then separated into different fractions using sieves SLM 200, resulting in a homogeneous dispersion and particle size distribution: 37 % of fraction 250-100 μm, 29 % of 100–40 μm, and 33 % of 40–1 μm. Table 1 provides the technical and elemental analysis of G grade coal and microdispersed carbon (MC) for dry ash free state (daf).

Table 1

Technical and elemental analysis of G grade coal and microdispersed carbon (MC)

Coal grade	Technical analysis, %			Elemental analysis (wt. % *daf*)				
	moisture	ash	volatile matter	C	H	N	O	S
MC	2.34	0.11	8.60	96.2	1.9	0.1	1.8	0.0
G	9.70	22.3	43.8	76.2	4.9	1.1	13.7	4.1

Table 2 illustrates structural and sorption characteristics of coal and microdispersed carbon.

Table 2

Structural and sorption characteristics of coal and microdispersed carbon obtained by DFT from the analysis of N_2 adsorption: specific gravity (ρ), specific surface area (*SSA*), pore volume (V_{pore}), and pore diameter (d_{pore})

Coal grade	ρ, g/sm³	SSA, м²/g	V_{pore}, sm³/g	d_{pore}, nm
MC	1.10	81.68	0.064	22.61
G	1.39	2.15	0.037	8.60

The number of surface functional groups of G-grade coal: (COOH – 0.088 mg-eq/g; OH – 0.012 mg-eq/g; >C=O – 0.057 mg-eq/g) was determined by titration with 0.05 M NaOH, Na_2CO_3, and $NaHCO_3$ according to Boehm [18; 19]. According to X-ray diffraction analysis (DRON 2.0 diffractometer with filtered Co-radiation), the inorganic component of G-grade coal is represented mainly by quartz, feldspar, and kaolinite.

The amino alcohols used were 2-amino-2-methyl-1-propanol (AMP) CAS 124-68-5 (DOW Chemical) and 2-amino-2-ethyl-1,3-propanediol (AEPD) CAS 115-69-5 (DOW Chemical). Amino alcohols were added to the CWCF at concentrations of 0.05 %, 0.1 %, 0.15 %, and 0.2 % by weight of CWCF.

Plasma chemical treatment of wastewater for the preparation of MC was carried out in a reactor, the scheme of which is given in [20]. The medium was a sample of organ-containing wastewater, after washing the technical parts, the composition of which is given in Table 3. In addition to the above components, the samples contain < 5 mg\dm³ of non-ionic surfactant from the group of ethoxylated fatty alcohols (Rokanol NL-6).

MC is formed as a result of the destruction of organic components of wastewater by condensation of a new phase [20; 21].

Table 3

Composition of organics-containing wastewater for the preparation of the CWCF

Composition of components,	C, mg\dm³
SO_4^{2-}	34.8
Cl^-	74.4
$HCO_3^- + CO_3^{2-}$	262.1
diesel fuel (hydrocarbons C_{10}-C_{14})	13.1
xylene + toluene	0.75

To produce the CWCF, coal powder was mixed with a dispersion medium (see Table 3) and then homogenized by wet grinding in a ball mill for 20–25 minutes. Wet grinding continued until the coal particles became smaller than 0.25 mm. The coal particles were then gradually mixed in a beaker filled with dispersant solutions and continuously agitated at 900 rpm for 5 minutes to ensure homogeneity. The resulting CWCF had a 61 % concentration of the solid phase. Its apparent viscosity was measured using a Rheotest-2 rheometer at a temperature of 20 °C, and the shear-stress/shear-rate data for the CWCF covered a range of 0.1 to 470 s^{-1}.

The particle size distribution of the CWCF was determined using the SOPAT method. It was found that particles between 1 and 5 μm made up 67 % of the sample, while submicron particles accounted for the remaining 33 %. A detailed description of the methodology is available in [22]. The ξ-potential was determined using the method presented in [23].

The sedimentation stability of the CWCF was assessed using a water-separating test and a storage period measured in days. After the CWCF was prepared, it was stored in a glass cylinder (25 ml in volume; CWCF layer 10 cm in height) at room temperature. The time it took for the sample to stratify in a measuring cylinder was used to measure the sedimentation stability of the CWCF.

Results and discussion

The key factor in using concentrated suspensions of the CWCF type is their compliance with the necessary operational requirements, especially their viscosity and sedimentation stability. The stability of CWCF is ensured by ionic-electrostatic and adsorption-solvation factors. To better understand the electrostatic interactions, they use the electrokinetic potential (ζ-potential) that occurs at the sliding boundary between the adsorption and diffusion layers. These processes determine the stability and rheological properties of the dispersed system.

Coal surfaces consist of an organic hydrophobic base, O, N, and S surface groups, and mineral inclusions with varying degrees of hydrophilicity [24]. The amount and composition of these inclusions depend on the coal's nature. In G-grade coal, there are a significant number of oxygen-containing groups (-OH, -O-R, >C=O) and mineral inclusions (Ad=22.3 %). Figure 1 shows the dependence of the ξ-potential of G-grade coal in the presence of amino alcohols and submicron carbon at different pH. At pH > 5, the ξ-potential becomes negative and reaches 25–30 mV in absolute value.

In the presence of amino alcohols, the ξ-potential shifts slightly to the negative region due to the adsorption of amino alcohols by the coal surface. The presence of submicron carbon shifts the isoelectric point of the surface of the dispersed

phase particles. The reason for this is a much smaller number of H+ ion donor groups on the surface of the MC due to the much lower ash content and oxidation of the latter. Since the MC particles are much smaller and have a more branched surface, they provide a significant contribution to the surface charge of the dispersed phase. Sufficiently high absolute values of the ξ-potential are provided only in highly acidic and highly alkaline environments. However, these environments do not meet the operational requirements for fuel injection equipment, as they will inevitably cause increased corrosion of fuel equipment, pipelines, and liquid fuel storage tanks.
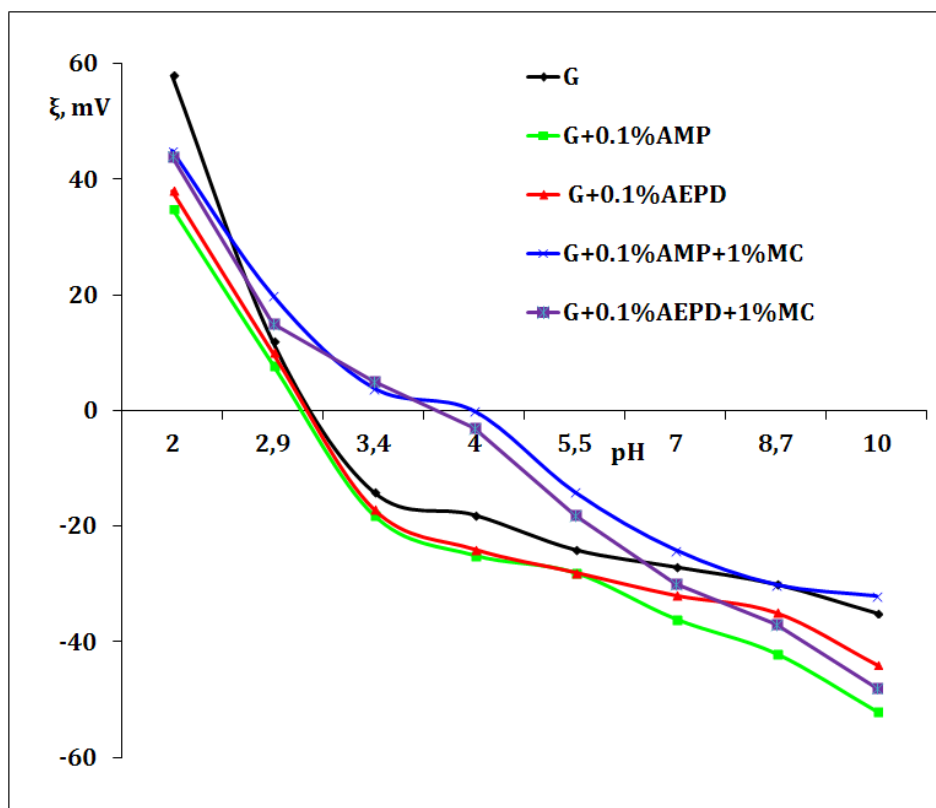

Fig. 1. Diagram of ξ-potential values of CWCF at different conditions

In the pH range of 4–9, the potential (ξ) does not exceed 35–35 mV in absolute value. Such values provide the desired stability of submicron-sized CWCF particles, but to obtain concentrated CWCF, particles of different sizes must be present in the suspension. This is a condition for achieving the maximum concentration of particles in the dispersed phase. However, particles of tens and hundreds of microns in size are prone to sedimentation, so additional stabilization factors should be applied.

In a slightly alkaline media, G-grade coal particles have a negative charge due to the dissociation of acidic surface groups. The addition of amino alcohols slightly increases the ξ-potential of coal particles by intensifying the binding of H+ ions desorbed by the coal surface. The effect of AMP is somewhat more noticeable than that of AEPD. The latter molecule has an additional OH group as a substituent, so the alkalinity of AEPD is slightly reduced. In addition, the somewhat

greater branching of the AEPD molecule causes greater spatial obstacles when interacting with the surface of the carbon particle. While the found values of ξ-potential of 40–45 mV may be sufficient for stabilization for particles smaller than 10 μm, the particles > 50 μm in size present in the CWCF suspension are not stabilized and may precipitate.

The amino alcohols adsorbed on the surface of coal particles will change the charge density on the surface of coal particles and enhance the electrostatic repulsion among the particles. In addition, MC particles will create steric repulsion and prevent particle adhesion. Electrostatic repulsion and/or steric effect are beneficial to improving CWCF stability.

Since electrostatic repulsion is not sufficient to stabilize the obtained CWCF samples at low concentrations of amino alcohols (Table 3), the effect of additives of MC was investigated. Amino alcohols form a weakly alkaline dispersion

medium, and an increase in the dissociation of H^+ donor groups of coal surface ions and an increase in the value of the ξ-potential can be expected. They can be considered cationic surfactants, and thus they are adsorbed by the negatively charged surface of the coal. In this case, the NH_2 group is protonated by the H^+ group of the coal surface, and the amino alcohol molecule is oriented by the OH alcohol group into the solution volume. The coal particle is then stabilized by the resulting adsorption-solvent layer of amino alcohols and H_2O dipoles. Additionally, since amino alcohols form a weakly alkaline environment, the dissociation of OH^- groups is enhanced, this increases the ξ potential of coal particles. This, in turn, enhances the ionic-electrostatic factor of suspension stabilization.

Table 4 shows that the sedimentation stability of both G-AMR and G-AERD coal systems is similar and increases as the concentration of amino alcohols increases. For CWCF in storage conditions, a stability of at least 5–7 days is acceptable. This is only possible for samples with a high concentration of AMP and AERD. It is recommended to use CWCF with a lower content of these additives immediately after preparation.

The sedimentation stability of G-grade coal systems – AMP and AEPD – significantly increases (by about two times) when 1 % MC is added to the system. This makes it possible to achieve the desired stability with less consumption of amino alcohols.

Table 4

Sedimentation stability St of CWCF for systems of G-grade coal - AMP, G-grade coal - AEPD without and with the addition of MS.

| aminoalcohol content, %	St, days			
	without MC		addition of 1% MC	
	AMP	AEPD	AMP	AEPD
0	0.2	0.2	1	1
0.05	3	2	5	4
0.10	5	4	8	7
0.15	7	5	12	12
0.20	8	7	>14	>14

The reason for this is that surfactants based on amino alcohols mainly form a 3-D mesh through hydrogen bonds, while particles of CWCF interact through intermolecular forces (Van der Waals forces). The energy of hydrogen bonds is greater than that of Van der Waals forces (6–130 kJ/mol vs. 10–20 kJ/mol), so although the spatial network in the case of surfactants with a branched structure is stronger, it also greatly increases viscosity. Additionally, surfactants based on amino alcohols with active hydrophilic groups (-OH, -NH_2) work well in highly polar environments (H_2O) due to the easy dissociation of these groups.

Dispersion (London interaction) between MC particles and coal is mostly achieved through the Hamaker constant, and in low-polar environments, it can provide the desired stability of CWCF while maintaining fluidity by reducing viscosity [25]. Since dispersion (and induction) interactions are inversely proportional to the sixth power of the distance between surfaces, their realization in systems with MC is due to the small size and the ability of MC particles to approach the surface of the dispersed phase. In this case, a spatial structural network is formed, which can keep the particles of the dispersed phase from aggregating and settling even if the particles are large (> 100 μm) (Fig. 2).

In addition, the effect of amino alcohols on improvement in CWCF concentration has mainly relied on hydrophilic modification and electrostatic force instead of the steric hindrance effect and reduction in interfacial tension. In contrast, 2-D and 3-D factors in the presence of MC give better dispersion performance due to a superior steric hindrance effect [12; 26].

The rheological properties of CWCFs are essential physical, chemical, and technological indicators. They are determined by the type, particle size distribution, and concentration of solid phase particles. These parameters were unchanged in the experiments. The type and concentration of amino alcohols in the systems without and with the addition of MC were varied.
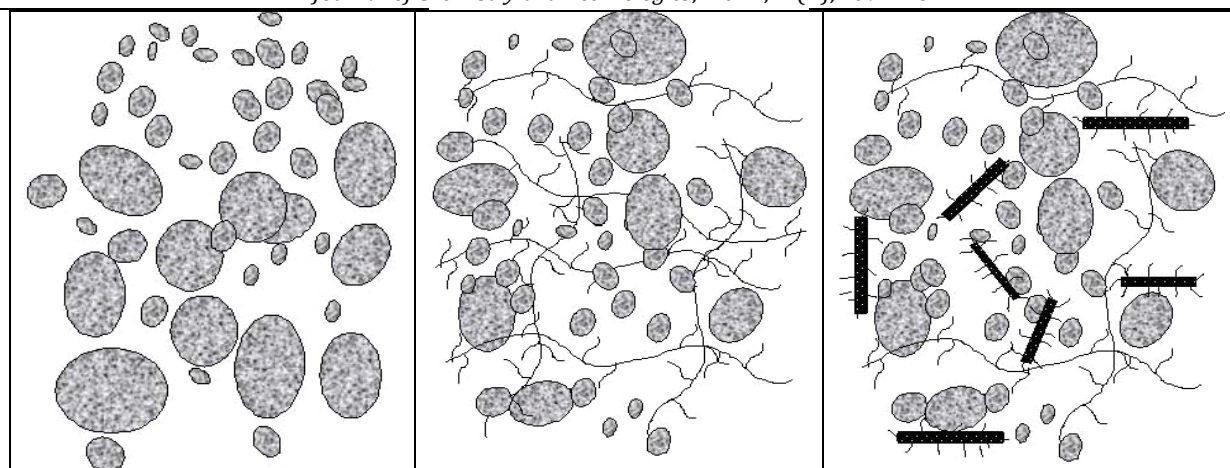
Fig. 2. Sedimentation of polydisperse systems without stabilizer additives, stabilized by amino alcohol-based surfactant, stabilized by amino alcohol-based surfactant, and MC
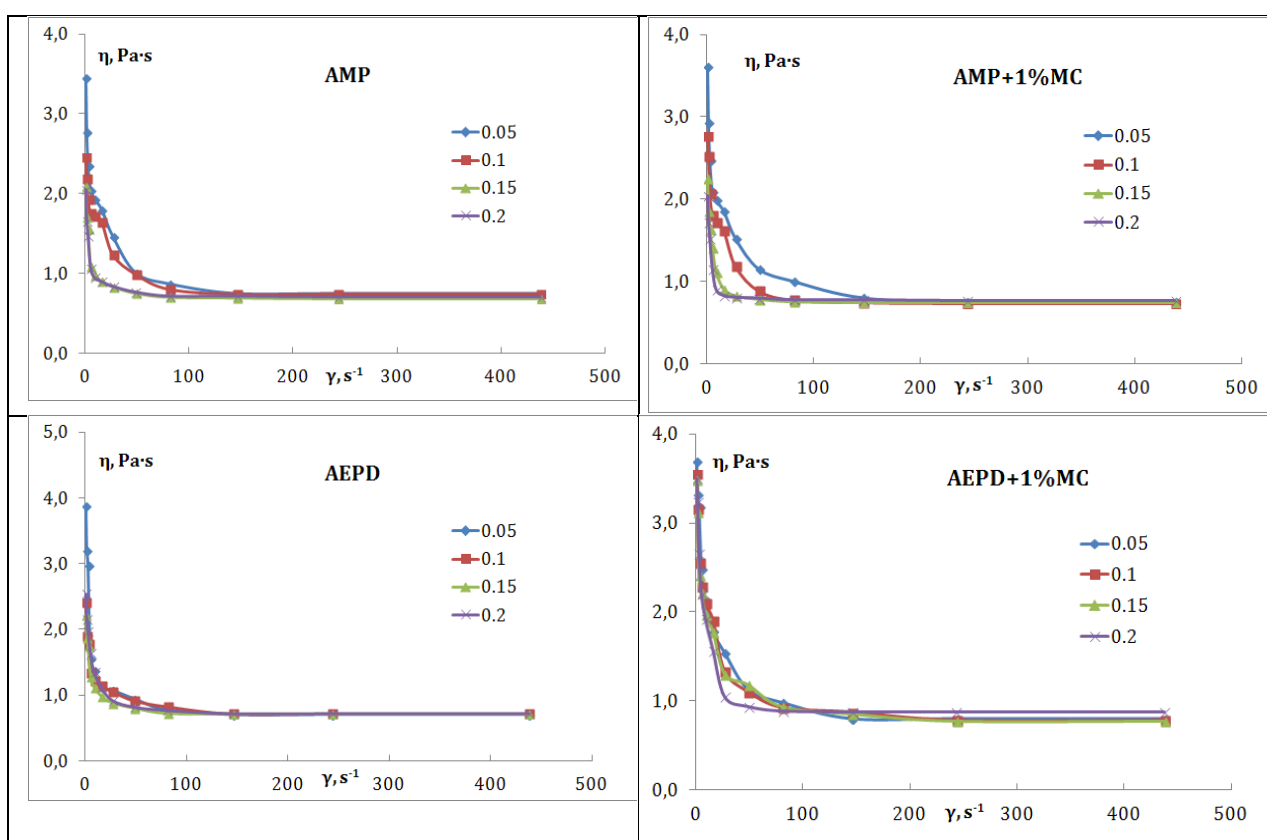
Fig. 3. Effect of amino alcohols and MC on the apparent viscosity η of CWCF at different shear rates γ

Fig. 3 illustrates the rheological behavior of the studied systems in the range of shear rates γ 0-80 s^{-1}, showing that the systems have a pseudoplastic behavior. When the shear rate γ increases, the apparent viscosity η decreases. When the shear rate γ exceeds 100 s^{-1}, the flow changes from pseudoplastic to Newtonian, indicating the destruction of the system's structure [27; 28]. Systems with AMP are more stable than those with AERD. Modifying the CWCF with MC can increase both η and the critical shear rate at which structure destruction is observed. However, to avoid the destruction of the CWCF structure, it is advisable to avoid high dynamic loads during pumping and mixing.

The apparent viscosity η of water-coal suspensions for fuel purposes is usually compared at a shear rate of γ = 9 s^{-1}. In order to achieve the desired properties, the fuel slurry should have η in the range of 1.5–1.65 Pa·s. By varying the content of amino alcohols and MS, the properties of the CWCF can be selected and adjusted to obtain the desired performance characteristics of the suspension fuel, as shown in Fig. 3. The sedimentation stability of these systems in the

presence of MC ensures that the CWCFs can be used for short-term storage.

Conclusions

The study showed that composite water-coal fuel can be stabilized by adding carbon submicron materials obtained by plasma-chemical conversion of organics-containing wastewater and amino alcohols. The ξ-potential of G-grade coal particles without surface modifiers only reaches high absolute values in highly acidic and highly alkaline media. However, these environments are not recommended for CWCF operation because they can lead to increased corrosion of fuel equipment, pipelines, and liquid fuel storage tanks.

It was found that the ξ-potential of G-grade coal particles in the presence of amino alcohols is 40–45 mV in absolute value. The introduction of highly dispersed carbon shifts the isoelectric point to the alkaline region. These systems demonstrate a pseudo-plastic type of flow in the range of shear rates of 0–80 s^{-1}. The introduction of highly dispersed carbon leads to an increase in the effective viscosity of the systems and can be recommended for controlling the fluidity of the CWCF. Sedimentation stability in the presence of additives of highly dispersed carbon and amino alcohols increases from 5–6 to 10–14 days, almost doubling the original stability. The sedimentation stability of these systems in the presence of MC makes the CWCFs suitable for short-term storage.

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