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NEW BORON-NITROGEN CONTAINING MODIFIED MULTIFUNCTIONAL ALKYLPHENOLATE ADDITIVES TO MOTOR OILS

NUEVOS ADITIVOS MULTIFUNCIONALES PARA ACEITE DE MOTOR BASADOS EN ALQUILOFENATO MODIFICADOS QUE CONTENIEN BORO Y NITRÓGENO

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ABSTRACT

By carrying out condensation reactions for alkylphenols and methylene-bis-alkylphenols with amines, alkanolamines and formaldehyde, working with boric acid and the neutralization of the obtained product with calcium hydroxide, new boron-nitrogen-containing alkylphenolate, additives have been synthesized (AKI-114B, AKI-212B, AKI-115B, AKI-210B, AKI-214B, AKI-219B).
The structure of the obtained - boron esters (derivatives of phenolate) has been studied through the use of the IR spectroscopy method. Simultaneously with studying the functional properties of the synthesized additives in motor oil, it has been established that their detergent, water, corrosion, oxidation and wear resistance properties greatly overbalance the same properties of their single nitrogen-containing and industrial analogues. Through thermoanalytic research conducted on OD-102T typed derivatograph, the high stability (through to 360ºC) of additives containing boron-nitrogen has been determined.
On the basis of new boron, nitrogen-containing modifications AKI-210B and AKI-115B different motor oil compositions such as M-8B, M-10G2 and etc. have been prepared and tested. The creation of high-synergy efficiency upon interaction of synthesized additives with other ones (sulfonates, dithiophosphates) has been observed, particularly in the improvement of tribological properties, and the increase in the oxidation induction period.

RESUMEN

Nuevos aditivos para aceite de motor basados en alquilofenatos modificados que contienen Boro y Nitrógeno (AKI-114B, AKI-212B, AKI-115B, AKI-210B, AKI-214B, AKI-219B), se sintetizan mediante reacciones de condensación de alquilofenoles y metil-bis, alquilofenoles con aminas, alcanolaminas y formaldehído. Los productos de reacción se neutralizan con Hidróxido de Cálcio.

La estructura de los ésteres de Boro (derivados del fenolato) ha sido analizada a través del uso del método de espectroscopía IR. El estudio de las propiedades funcionales de los aditivos sintetizados para el aceite de motor, ha establecido que sus propiedades detergentes de agua, de corrosión, de oxidación y de resistencia al desgaste superan en gran medida en las mismas propiedades a los aditivos que contienen nitrógeno y aditivos industriales análogos. La investigación termo analítica realizada en un derivatógrafo OD-102T permitió determinar que los aditivos conteniendo Boro-Nitrógeno tienen una alta estabilidad (hasta 360°C).

Sobre la base de los nuevos aditivos Boro- Nitrógeno se prepararon las modificaciones AKI-210B y AKI-115B y diferentes composiciones de aceite de motor tales como M-8B y M-10G las cuales fueron igualmente evaluadas. Se ha observado la creación de una alta eficiencia y sinergia tras la interacción de aditivos sintetizados con otros (sulfonatos, ditiofosfatos), particularmente en la mejora de las propiedades tribológicas y el aumento en el período de inducción de la oxidación.

KEYWORDS / PALABRAS CLAVE

Alkylphenolate | Amine | Alkanolamine | Boric Acid | Additives | Motor oil.
Alquilofenato | Amina | Alcanolamina | Ácido Bórico | Aditivos | Aceite de motor.

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1. INTRODUCTION

The coordination bond that boron atoms form with nitrogen atoms, especially cyclic ethers formed by alkanolamines, increase the water and temperature resistance of the boron containing organic compounds.

Boron atoms adsorbed on the surface of the metal form an isolated layer with it, while also overcoming the catalytic effects of metal in the oxidation process. With an increase in temperature B_2O_3, Fe_3B_2 and BN layers are formed. It improved the tribiological properties of the additives and improved its oxidation and corrosion properties.

Existence of O=B–O–B=O...,HN<, >N→B- OH fragments in nitrogen-containing com¬pounds, for example: in borated succinimides and amines, increasing the temperature resistance and at the same time detergent-dispersive properties. In order to increase the dissolving of boron compounds in oil and effectiveness of a complex of boron atoms with nitrogen-containing organic compounds, boron ethers are used at the same time. [1]–[6].
Due to this,it is possible that their exploitation features would be increased when embedding the boron atom in the nitrogen molecule containing alkylphenolate additives.

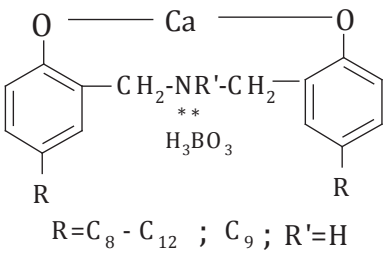
2. EXPERIMENTAL DEVELOPMENT

Obtaining the synthesized alkylphenolate typed boron-containing additives was carried out in the following steps:

1. Condensation of alkylphenol or methylene-bis-alkylphenol with formaldehyde or para-form and amine compounds (ammonia, octadecylamines, mono-and diethanolamines, alkenyl-succinimids). Synthesized on Mannich base to be controlled for ester variant by refraction ratio of mixture.

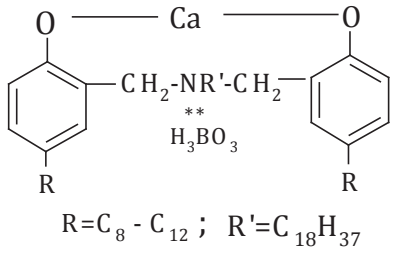
2. Processing of the condensation product with boric acid; (6.6-8.0% due to alkylphenol upon provided additives)

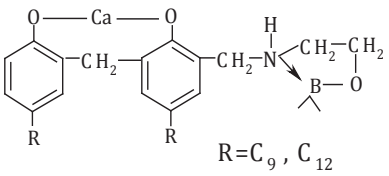
3. Neutralization of the obtained product with calcium hydroxide. (18-20% due to alkylphenol (weight part) upon provided additives).Diluent oil is diluent I-12A. Each additive is released from the water, getting dried, centrifuged for cleaning from impurities and thinners (benzole, heptane). Within the framework of the aforementioned steps, we have obtained boron-nitrogen-containing alkylphenolate additives (not known to literature) with the formulas indicated below [7]–[9].



AKI-114B
Calcium salt from the product of condensation of alkylphenolate with formaldehyde and ammonia and processing with boric acid;

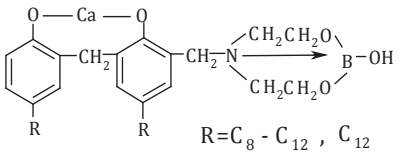
AKI-212B
Calcium salt from the product of condensation of alkylphenolate with formaldehyde and octadecyl amines and processing with boric acid;

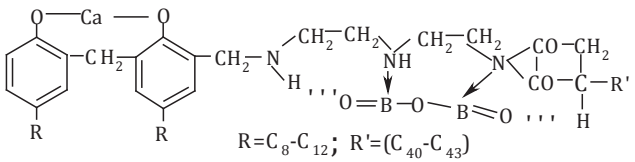




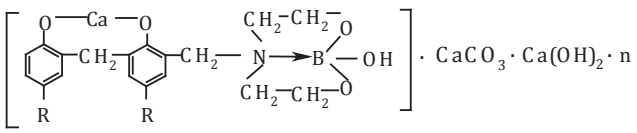
AKI-115B
Calcium salt of the product of condensation of methylene-bis-alkylphenol with formaldehyde and mono ethanolamine and processing with boric acid;

AKI-210B
Calcium salt from the product of condensation of methylene-bis-alkylphenol with formaldehyde and diethanolamine and processing with boric acid;





AKI-214B
Calcium salt from the product of condensation of methylene-bis-alkylphenol with alkenyl- succinimide and paraform and processing with boric acid.



AKI-219B
Carbonated calcium salt from the product of condensation of methylene-bis-alkylphenol with diethanolamine and formaldehyde and processing with boric acid.

This additive is a carbonated variant of AKI-210B additive. The feed rate of CO_2 gas during carbonatation is 15-16 ml/min at the temperature 70-80^{0}C.

The characterization of physicochemical and functional properties of boron nitrogen containing additives is provided in **Table 1.** IR-spectrums were recorded using a Fourier-Nicolet İS-10 IR Spectrometer, made in the United States of America.

Thermooxidation properties of synthesized alkylphenolate additives have been studied by derivatographic analysis at ОД-102Т Paulik (Hungary) typed derivatograph with dynamic working mode at air conditions and a temperature of 10ºC. As a sample, 0.1g of standard aluminum oxide is used.

3. RESULTS AND ANALYSIS

It is clear from **Table 1** that boron-nitrogen-containing additives are significantly better (0.03-0.08% against 1.0-1.2%, 0.38-0.40 mm against 0.5-0.55 mm) than their single nitrogen-containing analogues regarding antioxidation and antiwear properties, as well as regarding detergent-dispersive and anticorrosion properties. For example: a succinimide fragment containing AKI-214"B" additive has better detergent properties than its analogue without boron (0 point).

The multifunctionality of boron-nitrogen-containing alkylphenolates makes it possible to create high-quality motor oils based on them. Creation of ether groups in modifications obtained on the basis of alkanolamine was determined by IR spectrum analysis of the products received from first condensation through to last neutralization.

Absence of a peak belonging to NH-group in the IR spectrum of aminomethylation product of alkhanolamines with methylen-bis-alkylphenol and presence of 1049.54 cm^{-1} zone characterizing –C–O– bond in alcohols, and diffuse line overlapping by OH groups of phenol at 3362,29 cm^{-1} confirms the structure of the aminomethylized product (**Figure 1**). In the IR spectrum of the intermediate product obtained after condensation of aminomethylized product with $B(OH)_3$ we observe the –C–OH– group, the absence of 1049.54 cm^{-1} zone, and instead of it formation of 1160.40 cm^{-1} zone, which can be said belongs to the –C–O– bond of ether groups (**Figure 2**).

Appearance (for sliding) of simple ether groups at 1131.56 cm^{-1} or 1129.46 cm^{-1} in the final product – phenolates proves the creation and maintenance of ether-covalent >B–O–C–group bonds until the end.

Coordination bonds can be easily created at low temperatures (80-85ºC). Virtual absence of observed zone in phenolic OH after condensation of phenolate with boric acid in the IR spectrum of phenolates confirms that the phenolate was obtained (**Figures 3 and 4**).

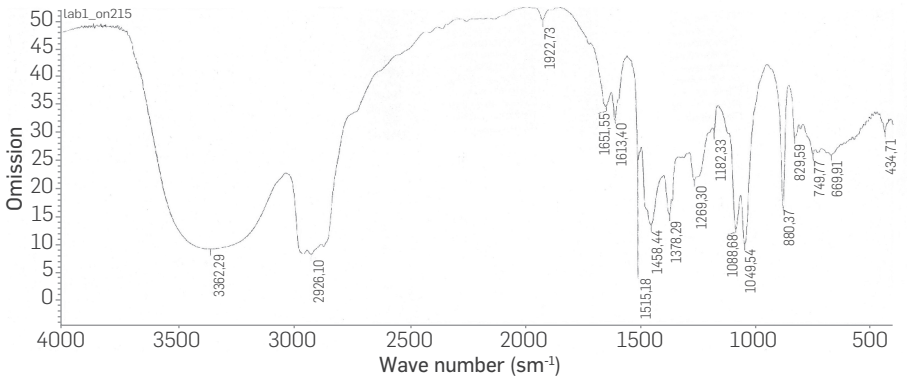


Figure 1. IR spectrum of aminomethylation product of alkylphenol with diethanolamine

Table 1. Physicochemical and functional properties of boron-nitrogen-containing alkylphenolate additives.

Additive samples with heteroatoms	Alkalinity number, mgKOH/g	Sulphate ash, %	Kinematic viscosity, mm^2/s	Composite, %			M-8 oil with 5% additive			
				B	N	Ca	Corrosion (lead plates) g/m^2	Stability on induction period of sediment formation % (by SFIP 30 hours) sediment	Detergent property, (point)	Wear resistance, (wear spot diameter, mm)
AKİ-114 (N)	95-100.8	9.7-9.8	73.5-76.9	–	0.9	2.2	2.8-5.0	0.45-0.50	0.5-1.0	0.5-0.55
AKİ-114 “B”(N)	106.4	9.95	68.5	0.5	0.8	2.3	2.5-3.7	0.1-0.25	0.5	0.42
AKİ-115 (N)	110-115	10.5	75-90	–	1.1	2.7	2.5-3.9	1.0-1.2	0.5	0.55
AKİ-115“B”(N)	125.7-130	11.01	67.1-89.1	0.56	1.1	2.9	0.7-1.2	0.03-0.08	0-0.5	0.38-0.40
AKİ-212“B”(N)	76.4	7.4	56.0	0.39	0.75	2.1	1.8	0.15	0.5	0.40
AKİ-210“B” (N)	120-128	10.5-11.5	75.0-85.0	0.52	0.71	2.9	0.1-1.0	0.01-0.09	0-0.5	0.38-0.40
AKİ-214"B" (N)	78.0-82.0	6.5-7.0	79.0-86.0	0.39	1.25	2.25	0.5-1.0	0.01-0.03	0	0.38-0.40
AKİ-219(B, N)	150-160	17.5	89.0	–	–	4.9	M-8 oil +4%			
							0.2-1.5	0.01-0.05	0-0.5	0.40

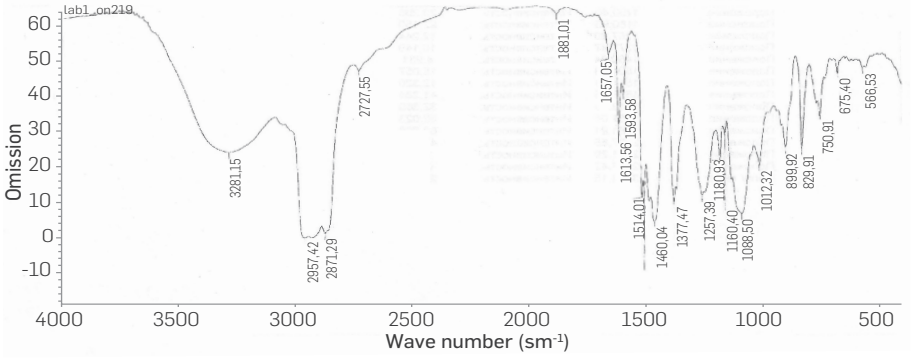


Figure 2. IR spectrum of the product of aminomethylation of alkylphenol with diethanolamine and condensation with boric acid

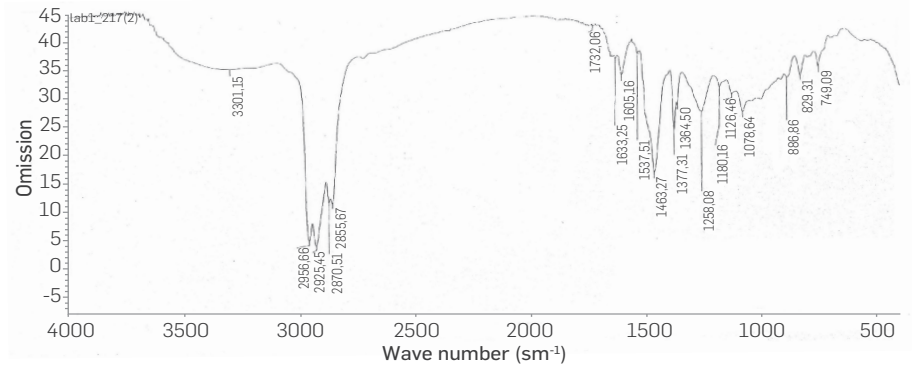


Figure 3. IR spectrum of calcium salt from the product of aminomethylation of alkylphenol with diethanolamine and condensation with boric acid

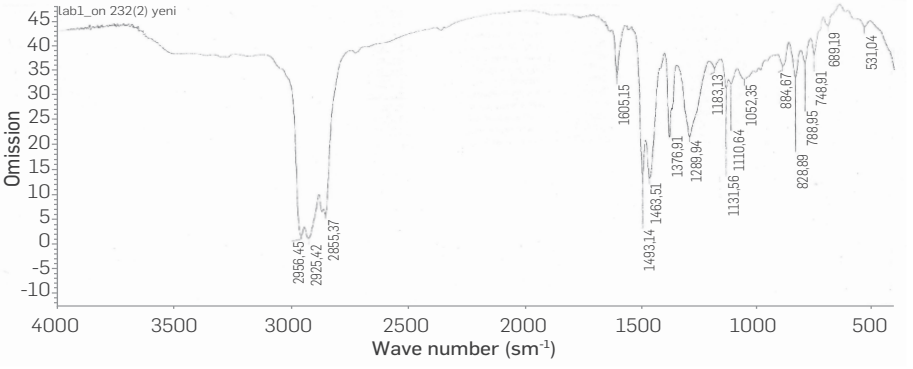


Figure 4. IR spectrum of calcium salt from the condensation product of alkylphenol with monoethanolamine and boric acid

examples are defined by exothermic effects and this is shown in differential-thermal analysis curves (DTA).

The thermooxidation stability of AKİ-114"B" is 320^{0}C and difference between it and AKİ-114 is just 10^{0}C. At the same time, as can be seen from the DTA of AKİ-114"B", the exo-effect is considerably less than the same indicator for the additive without boron. In this part of the curve, borders have a spread shape. But in the condensation products of additives (CP) the beginning of the second stage of destruction for CP of AKİ-114 and AKİ-114"B" additives is 285^{0}C and 310^{0}C, respectively. The maximum level for thermodestruction without boron is 610^{0}C, and with boron it is 700^{0}C (**Figure 5**).

The first stage for thermodestruction of the condensation products of AKİ-115 and AKİ-115B additives is 300^{0}C and 320^{0}C respectively, but in the additives it is 345^{0}C and 360^{0}C, respectively. But half-life period $T_{50\%}$ makes 360^{0}C for the AKİ-115 additive, and 400^{0}C for AKİ-115"B". The thermooxidation efficiency (exo-effect) of AKİ-115B in comparison with analogues is observed at temperatures of more than 200C (**Table 2, Figure 6**).

As can be also seen from **Table 2**, the difference between thermostability indicators of AKİ-114B and AKİ-115B additives is 40^{0}C (320-360^{0}C). It shows moving thermo-chemical transformations (thermooxidation stability) of solvent oil contained in the additives, which usually takes place at 280-290^{0}C to high-temperature zones [10].

At the same time according to **Table 2**, the half-life period for such industrial samples as ACK and MACK, which are part of the carboxylate group containing additives, makes 340-345^{0}C. The thermooxidation stability of nitrogen-containing carboxylate can be compared with boron-nitrogen-containing ones. Both of them are a little higher than sulfur-containing ones.

COMPARISON OF THERMOOXIDATION PROPERTIES OF BORON-NITROGEN-CONTAINING ALKYLPHENOLATE ADDITIVES WITH INDUSTRIAL AND SYNTHESIZED ANALOGUES

The thermostability of additives is determined by the ending temperature of the first stage and beginning temperature of the second stage of destruction, thermogravimetric (TQ) curves by weight loss ($T_{10\%}$, $T_{20\%}$, $T_{50\%}$, $T_{70\%}$), and the thermooxidation degrees of

AKI-114 - Calcium salt of condensation product of alkylphenol with ammonia and formaldehyde [11];

AKI-115 - Calcium salt of condensation product of alkylphenol with monoethanolamine and formaldehyde [7]

VNIINP-714–Carbonated calcium salt of thio-bis-alkylphenol [12];

OLOA-218A–Carbonated calcium salt of thio-bis-alkylphenol [13];
ASK–Calcium alkylsalicylate;

Table 2. Comparative thermooxidation stability of alkylphenol additives

Additives Cont. heteroatomsan func. groups.	$T_{10\%}$,°C	$T_{20\%}$,°C	$T_{50\%}$,°C	$T_{70\%}$,°C	The end of the 1st stage of thermodestruction and the beginning of the 2nd stage	
					temperature, °C	weight loss, %
AKI-114 (N)	225	285	370	495	310	42
AKI-114"B" (N)	235	275	390	495	320	34
AKI-115(N)	250	285	360	465	345	47
AKI-115"B" (N)	270	310	400	475	360	40
VNIINP -714 (S)	230	270	355	440	335	44
OLOA-218A (S)	250	295	370	460	320	40
ACK (COOH)	265	295	345	385	330	45
MACK (COOH)	265	280	340	400	340	50

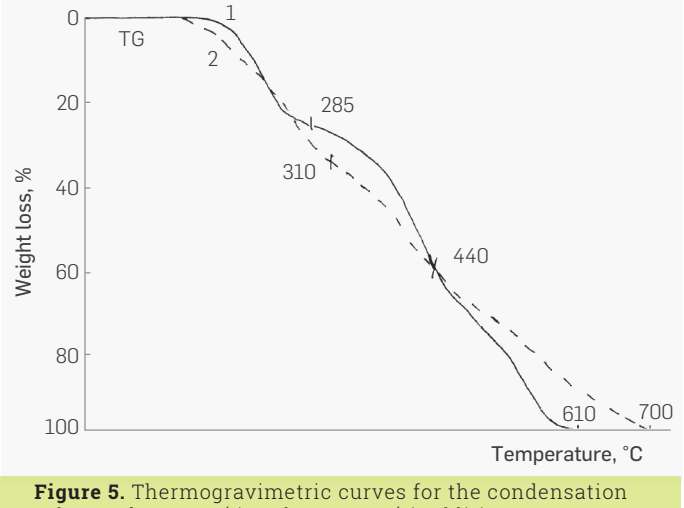


Figure 5. Thermogravimetric curves for the condensation products of AKI-114 (1) and AKI-114B (2) additives 1.AKI-115; 2. AKİ-115B, 3. AKI-114B

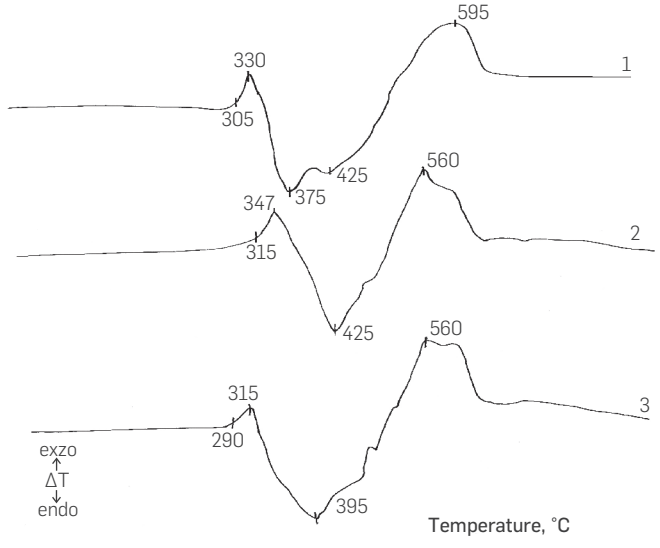


Figure 6. DTA additive curves 1. AKI-115; 2. AKİ-115B, 3. AKI-114B

MASK–High-alkalinity alkylsalicylate [14].

It is clear from the table that the modified alkylphenolate additive AKI-115"B" has the highest thermostability indicators (360°C).

It has been established that thermostability of:

- Boron-containing additives is higher (10-20°C) than analogues without boron;
- Carbonated high alkalinity additives is much higher (10-15°C) than average alkalinity ones;
- Additives obtained on the basis of alkanolamines is higher (30-40°C) than additives containing primary amine.

Through use of the above-mentioned heteroatom-containing additives, several motor oil compositions for gasoline and diesel engines have been developed. For example, M-10G_2 oil is an industrial diesel, especially heavy-loaded auto-tractor diesel oil produced to GOST 8581. This M-10G_2 oil has been prepared by use of VNIINP-360 additive with insignificant multifunctionalty tribological properties and average alkalinity.

Through use of the boron-nitrogen-containing AKI-115B, an additive composition meeting the requirements of the oil has been created.

On the basis of the oil and a small portion of AKI-210B and AKI-115B additive and other components, oil compositions have been developed and tested M-10G_2k, M-10V_2 [15], [16].

In Tables 3 and 4 there are ASTM numbers and GOST methods according to which physicochemical and functional properties of oils are determined. Both compositions of oils M-8B and M-10G_2. meet modern requirements. [17]–[19].

So, it has been shown that the multi-functionality and high efficiency of boron nitrogen-containing alkylphenolate additives make possible their use as part of motor oil for gasoline and diesel engines that meet modern requirements.

CONCLUSIONS

For the first time, on the basis of the condensation of alkylphenoles with formaldehyde, amines, alkanol amines and boric acid step by step, boron-nitrogen-containing alkylphenolate additives have been synthesized, with several advantages having been determined for boron-containing additives (AKİ-114B, AKİ-115B, AKİ-210B, AKİ-212B, AKİ-214B, AKI-219B) in comparison with their analogues without boron:

- detergent properties (0 points against 0.5-1.0 points);

Table 3. Physicochemical and functional properties of M-8B oil composition with AKI-210B additive

Indicators	M-8B oil GOST 10541-78 norm	Experimental motor oil with AKİ-210"B" additive	The oil firm Shell Rimula C20W-20
Kinematic viscosity, 100^{0}C,mm^2/s	8±0.5	8.2	8.8
Viscosity index, not less than	93	94	102
Base number, mgKOH/g not less than	4.2	5.1	4.5
Sulfate ash, %, not more than	0.95	0.88	0.78
Weight of mechanical mixture, %, not more than	0.015	0.010	-
Flash Point determined at the open pot ^{0}C, not less than	207	220	236
Freezing temperature ^{0}C, not more than	Minus 25	Minus 25	Minus 21
Stability on induction period of precipitation formation (SFIP), 30 hours, %	-	0.1	-
Corrosion (on lead plates) g/m^2, not more than	10	stable (0.3-1.0)	-
Detergent properties (due to PZV) point, nor more than	0.5	0.5	0.5

Table 4. Physicochemical and functional properties of M-10G$_2$ oil composition with AKI-115B additive

Indicators	M-10G$_2$ oil GOST 8581-78 norm	M-10G$_2$ oil AKİ-115B additive	The oil firm Shell Rotella TX-30	Test method	
				GOST	ASTM
Kinematic viscosity, 100^{0}C, mm^2/s	11±0.5	10.9	10.8	33	D 445
Viscosity index, not less than	85	94	102	25371	D 2270
Base number, mgKOH/g not less than	6.0	8.5	9.4	11362	D 4739
Sulfate ash, %, not more than	1.65	1.1	1.3	12417	D 874
Flash point determined at the open pot ^{0}C, not less than	205	215	202	4333	D 92
Freezing temperature ^{0}C, not more than	Minus 15	Minus16	Minus 18	20287	D 97
Weight of mechanical mixture, %, not more than	0.015	0.008	0.010	6370	–
Density, 20^{0}C, kg/m^3, not more than	905	905	896	3900	D 4052
Stability on induction period of precipitation formation (SFIP), 40 hours, %	40 (0.5)	40 (0.08)	40 –	11063	–
Corrosion (on lead plates) g/m^2, not more than	20	Stable	Stable	20502	–
Detergent properties (due to PZV) point, not more than	1.0	0.5	0.5	5726	D 892

- antiwater stability: alkalinity reduction (13-15% against 18-20%);
- along with slightly improved corrosion, higher oxidation stability (the amount of precipitation is reduced from 1.2% to 0.03-0.05%);
- prevailing thermooxidation properties: according to results of the thermogravimetric analysis, the beginning of the second phase of thermodestruction makes 320-3600C;
- AKI-115"B"additive obtained from alkanolamine has higher thermal oxidation stability (3600C) than additives obtained from primary amines;
- the antiwear properties of boron-containing additives is higher than the same properties for nitrogen-containing analogues (wear scar diameter 0.40 mm against 0,55 mm).

Taking into account everything mentioned above, it is possible to create modern motor oil compositions through the application of additives containing boron-nitrogen.

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