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Nature Environment & Pollution Technology

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Original Research

Aqueous phase of hydrothermal liquefaction as a concrete modifier

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Abstract: The processes of liquid fuel production from organic wastes by hydrothermal liquefaction lead to the formation of wastewater characterized by a high content of organic compounds, including lignosulfonates and their derivatives. The paper presents the results of a study designed to assess the possibility and feasibility of using such wastewater as concrete modifiers. The study confirmed the hypothesis that the use of HTL-AP slows down concrete curing processes (curing time increased 1.9 times compared to the control sample). It was found that the modifying properties of HTL-AP are higher than those of the commercial concrete modifier because even at higher curing retardation rates (curing time is 12.2% higher compared to concrete modified with the commercial solution), the use of HTL-AP results in minimal reduction in the strength properties of the concrete. The application of HTL-AP and commercial modifier results in a 7.1% and 14.5% reduction in compressive strength, respectively, and a 6.2% and 12.2% reduction in tensile strength, respectively. Based on the results of the study, it is concluded that the use of HTL-AP as a retarder in concrete curing processes is well justified, as the positive effect has been experimentally confirmed. Using of HTL-AP as concrete modifier will improve the environmental efficiency of HTL processes and reduce the cost of frost-resistant concrete by eliminating the use of traditional expensive modifiers.

Key Words	Concrete modifier; aqueous phase of hydrothermal liquefaction; recycling; sustainable construction materials; green chemistry
DOI	https://doi.org/10.46488/NEPT.2025.v24i03.D1749 (DOI will be active only after the final publication of the paper)
Citation of the Paper	Yuliya Kulkova, Ibragim Garifullin and Maria Dmitrieva, Olga Babich 2025. Aqueous phase of hydrothermal liquefaction as a concrete modifier. *Nature Environment and Pollution Technology*, 24(3), D1749. https://doi.org/10.46488/NEPT.2025.v24i03.D1749

1. INTRODUCTION

The issues of waste recycling and their beneficial use for solving technological problems in the construction industry and for solving energy saving problems are quite actively being considered by scientists from various countries (Achi et al. 2024, Gupta et al. 2023).

Hydrothermal liquefaction technology is very effective for processing organic wastes with high moisture content (up to 75-95%) at temperatures of 200-400 °C and pressures of 4-22 MPa, resulting in valuable energy sources (bio-coal, bio-oil, and biogas), the ratio and yield of which depend on the conditions of the technological process (Mahima et al. 2021, Yu et al. 2022).

There is considerable interest in this technology worldwide, as it offers a number of significant advantages, including the elimination of pre-drying of raw materials, resulting in higher energy efficiency (Mishra & Mohanty, 2020). One of the key problems of hydrothermal liquefaction processes is the formation of wastewater (HTL-AP) up to 80% by mass of the processed feedstock (Watson et al. 2020, Lenget et al. 2020, SundarRajan et al. 2021).

HTL-AP is characterized by a high content of organic compounds and nutrients (organic carbon content up to 25 g/L) (Thomsen et al. 2024, Cheikhwafa et al. 2024, Shen et al. 2018, Yang et al. 2018). According to some studies (Watson et al. 2020, Shen et al. 2018, Kulikova et al. 2023), heavy metals can be detected in the composition of HTL-AP, which is characteristic of wastewater from the processing of excess activated sludge. Thus, ensuring the environmental and economic efficiency of HTL-AP treatment, utilization, or beneficial use is a critical task.

Based on literature data, HTL-AP was found to potentially contain polyaromatic compounds such as lignosulfonates or their analogs (Kulikova et al. 2023, He et al. 2020, Si et al. 2019, Chen et al. 2019, Jokinen et al. 2023). A number of studies aimed at finding technical and technological solutions for the beneficial use of HTL-AP are known, for example, as a nutrient medium for the cultivation of microalgae (Cordovaet et al. 2020, Shende et al. 2017), for the production of biogas (Venkiteshwaran & Zitomer 2021), biohydrogen (Si et al. 2019, Torri et al. 2021), and as a fertilizer for plant cultivation (Jesse et al. 2019). The literature does not describe the use of HTL-AP in the construction industry as a source of lignosulfonates for concrete modification.

At the same time, lignosulfonate preparations are widely used as modifying additives for concrete, providing improvement of concrete technical properties such as strength, water, and frost resistance (Breilly et al. 2021). Due to the disruption of global supply chains, the Russian construction materials market is currently experiencing the need to seek out and develop innovative technologies for the production of such modifiers.

An important aspect of using fine-grained concrete is not only its strength and durability but also its environmental safety. Lignosulfonate-based modifiers have low toxicity and no harmful effects on the environment, making them a preferred choice for use in construction (Liu et al. 2018).

Considering the relevance of the search for innovative, ecologically and economically effective ways of HTL-AP utilization, a hypothesis was made about the possibility of using the above mentioned wastewater for concrete modification. The paper presents the results of testing the hypothesis that the addition of HTL-AP to concrete mixtures will increase its mobility, increase the curing time while maintaining the strength properties of products.

Currently, there are no studies in the literature showing the feasibility of using HTL-AP for concrete modification. This work will open a new direction for processing this type of wastewater, which will give impetus to the development of HTL technologies and will reduce the cost of certain types of building materials and products.

2. MATERIALS AND METHODS

2.1. Conditions for obtaining the water aqueous phase of hydrothermal liquefaction (HTL-AP)

Excess activated sludge from the municipal biological wastewater treatment plant in Kaliningrad (Russia) was used as raw material. The raw materials were delivered to the laboratory in their natural wet state.

The process was conducted at 280 °C in an autoclave reactor under a pressure of 3.6 MPa, with a total processing time of 30 minutes. Hydromodule 1:10. After HTL, the reactor was discharged to a filter system where the solid phase was separated from the liquid phase. The liquid phase was then mixed with dichloromethane in a ratio of 1:2 and the extraction of the oil fraction (biofuel) was performed. The mixture of dichloromethane and water was separated on a separating funnel. The aqueous phase (HTL-AP) was stored in a refrigerator until use.

2.2 Methods for studying HTL-AP composition

The density of the additive was determined by the weight method. A photometric method was used to determine the concentration of lignosulfonates based on their interaction with nitric acid and the formation of yellow nitro derivatives in alkaline medium. The interfering influence of phenols and aromatic amines was eliminated by steam predistillation without changing the pH of the medium. Then, 2 cm^3 of sodium nitrite solution and 2 cm^3 of acetic acid solution were added to the 100 cm^3 of diluted sample and stirred. After 15 minutes, 4 cm^3 of ammonia solution was added. The optical density was measured at a wavelength of $\lambda = 430$ nm and then subtracted from this value the optical density of the blank, which was treated in the same way as the test sample, but with the addition of 2 cm^3 of distilled water instead of the sodium nitrite solution. The content of the substances was determined using the calibration curve. The graduation curve was plotted against tannin (Khabarov 2004).

2.3 Composition of mortar mixture

The composition of the tested concretes is presented in Table 1. Portland cement (PC 300-D0-B), quick-setting cement with compressive strength at 28 days (M-300), without additives (produced by Nargit, Russia, Mytishchi) was used. Sand was purchased from the supplier NerudStroy-M, Moscow (the origin of the sand is not indicated on the package). Composition of mortar mixture were selected in accordance with the state standard of the Russian Federation GOST 27006- 2019 “Concretes. Composition selection rules” (GOST 2019).

Table 1: Compositions of tested fine-grained concretes

Component	Control sample	Sample with HTL- AP	Sample with commercial lignosulfonate
HTL-AP	-	92 mL	-
SAVEMIX4000 lignosulfonate, mL	-	-	8 mL
Portland cement (PC 300-D0-B)	1400 g	1400 g	1400 g
Quartz sand fr. 1.0-1.5 mm, quartz sand with a size modulus	3360 g	3360 g	3360 g
Water	700	608	692

2.4 Determination of rheological characteristics

The consistency of the mortar mixture was determined according to GOST 5802-86 using the device presented in Fig. 1.

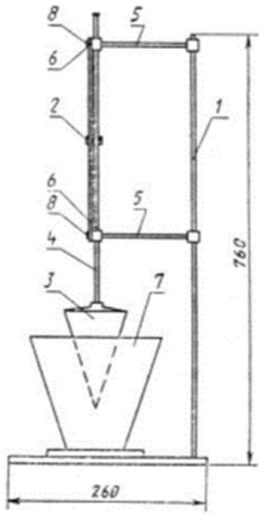

Fig. 1: Device for determining the mobility of the mortar mixture: 1 – stand rod, 2 – scale, 3 – reference cone, 4 – stand rod, 5 – holders, 6 – guides, 7 – vessel of the mortar mixture, 8 – locking screw.

The device is positioned on a horizontal surface and the degree of freedom of sliding of the rod 4 within the guides 6 is evaluated. The vessel 7 is filled with the mortar mixture to 1 cm below the edges, the mortar mixture is then rammed by poking with a steel rod 25 times and by taping 5-6 times lightly on the table, then the vessel was placed on the site of the device. The tip of the cone 3 is brought into contact with the surface of the solution in the vessel, the cone rod is fixed with the locking screw 8 and the first reading on the scale is made. Then the locking screw is released. The cone must sink freely into the mortar mixture. The second reading is taken on the scale 1 minute after the beginning of the cone immersion. The depth of cone immersion, measured with an error of up to 1 mm, is determined as the difference between the first and second readings. The normal consistency of the mortar mixture was determined according to ASTM C191-08 using a Vika OGC-1 device mark (Laborkomplekt, Moscow, Russia).

When the mixing is finished, the ring is quickly filled with cement paste in a single step and shaken 5-6 times, tapping the plate against a hard surface. The surface of the paste is leveled with the edges of the ring by cutting off the excess paste with a knife wiped with a damp cloth. Subsequently, the instrument pestle is brought into contact with the surface of the paste situated in the center of the ring, and the rod is secured with a locking device. The rod is then released rapidly, allowing the pestle to plunge into the paste. Following the release of the rod, the immersion is measured on the scale after a period of 30 seconds. The ring with the paste must not be jolted when measuring. In the event that the consistency of the cement paste is deemed inappropriate, the quantity of water should be adjusted and the paste re-mixed, ensuring that the pestle is immersed to a greater depth. The quantity of water required to create a paste of standard consistency is calculated with a precision of 0.25%.

The setting time of the concrete mortar was determined in accordance with the ASTM C191-08 standard using the Vika OGC-1 device. To determine the onset of setting, the needle is lowered until it touches the surface of the cement paste and the rod is secured in this position with a locking device. After 1-2 seconds, the rod is released, and metal needle sinked without barriers into the cement paste. The depth of needle penetration was measured after 30-45 sec. Measurements were repeated in the same way every 10 minutes, but ring were shifted on 10-15 mm after each measurement. Care should be taken to ensure that the needle does not hit the site of the previous needle injection. After each immersion, the needle was cleaned carefully with the tissue. Between the measurements cement paste on the plate was covered with the textile material, but it without contact with cement paste. The temperature in the room should be 20-25^{0}C and humidity not less than 65. The setting time is defined as the time from the moment of mixing water with cement powder to the moment when the needle does not penetrate the cement paste or penetrate by less than (4±1) mm.

2.5 Determination of mechanical characteristics of concrete

In order to evaluate the influence of the modification on the quality of the products, 40 x 40 x 160 mm prisms were produced and cured under normal conditions at a temperature of 20±2 C and humidity of 95-95%. The deflection and compressive strength of the specified samples were investigated. The deflection and compressive strengths were determined using a ToniPRAX unit (Berlin, Germany) according to DIN EN 196-1-2016 (Academia 2024). The deflection tests were performed at 1 day, 7 days and 28 days intervals.

3. RESULTS AND DISCUSSIONS

3.1. Analysis of physicochemical properties and composition of HTL-AP

Composition analysis using NMR spectroscopy was previously performed for the above wastewaters (Kulikova et al. 2023). Carbon in aliphatic chains (34.05-41.82%) was found to be pre-dominant in the wastewater composition, followed by carbon in bonds with carboxyl groups and aromatic rings (26.42-34.44%). Based on these data, it was concluded that the predominant components are oxygenated aromatic compounds, including lignosulfonates (Fig. 2).

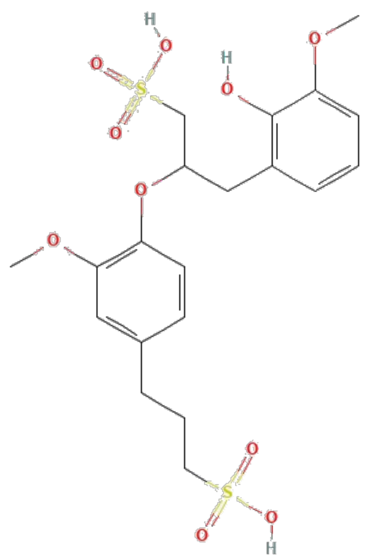

Fig. 2: Structure of lignosulfonates (PubChem, 2024)

Since there was no clear confirmation of the presence of lignosulfonates in previous works, it was decided to quantitatively analyze their content using the photometric method and tannin as a standard. The results of the evaluation of HTL-AP properties are summarized in Table 2. Since the content of lignosulfonates in HTL-AP is about 30 times lower than in the commercial modifier solution, this fact must be taken into account when formulating the test concrete mixtures.

Analysis of published data, showed that the HTL-AP in most cases is characterized by high COD values (in some cases over 100 g/L), and contains large amounts of organic carbon (SundarRajan et al. 2021). Obviously, the quality of wastewater significantly depends on the type of raw materials used. According to the literature data, the content of organic compounds in HTL-AP from algae biomass processing is 20-65 g COD/l, from manure processing 10-55 g COD/l, sludge processing 5-15 g COD/l and lignocellulose biomass procrssing 10-22 g COD/l (Fan et al. 2022, Gu et al. 2019, Leng et al. 2021, Usman et al. 2019).

This variability in the composition of wastewater, depending on the raw materials, creates certain difficulties in ensuring the consistency of concrete modifier composition planned for production. However, this problem can be overcome by averaging, adjusting, and constantly monitoring key parameters of manufactured products. In case than composition of the raw materials is constant (if the HTL equipment is aimed at processing a specific type of waste), the consistency of the HTL-AP composition will also be achieved.

Table 2: Main physicochemical parameters of HTL-AP

Parameter	HTL-AP	SAVEMIX4000
Density	1.0112	1.1488
Lignosulfonate concentration	2.89±0.14 g/L	300 g/L
pH	6.5	4.5-5.5

3.2. Evaluation of rheological properties of concrete mixtures

As mentioned above, 3 types of concrete mixtures were tested: unmodified concrete (sample 1), modified with HTL-AP (sample 2), and modified with the commercial modifier SAVEMIX4000 (sample 3). The results of the evaluation of rheological properties are presented in Table 3.

From the experimental results it can be concluded that the mixture modified with HTL-AP is 31.6% more mobile than the unmodified control sample. At the same time, the mobility of the concrete mix modified with commercial composition is inferior to the sample with HTL-AP modification (mobility is lower by 6.3%).

The initial density of the concrete paste, determined by analyzing the immersion depth of the needle of the Vika device, with the same amount of liquid for sample 2 was the maximum of 40 mm, which is 8 times higher than that of the control sample. At the same time, the setting time of sample 2 is 1.9 times higher than that of the control sample and 12.2% higher than that of the sample modified with commercial solution. It can be clearly concluded that HTL-AP has modifying properties that significantly increase the mobility of the concrete paste and the setting time.

Table 3 Results of evaluating the rheological properties of concrete

Parameter	Sample 1	Sample 2	Sample 3
Concrete consistency, ΔP, см	6.5	9.5	8.9
Depth of immersion of Vika device when evaluating the density of concrete paste, mm	5	40	34
Setting time	2 h 10 min	4 h 5 min	3 h 35 min

An important parameter in this case is the preservation of the mechanical properties of the modified concrete at the increase of the setting time, in this connection, in the second stage of the research the mechanical properties of the products from the studied concrete samples were evaluated.

3.3. Mechanical properties of concrete

The results of the evaluation of the tensile strength of the products made from the concrete samples tested are presented in Table 4 and Fig. 3. Based on the data, the following conclusions can be drawn: on the first day, the tensile strength of the modified concrete is 20% lower than that of the control, which indicates that lignosulfonates acts as a retarder in the hardening of the concrete mix. Further testing showed that the rated tensile strength of the modified concretes (at 28 days) exceeded that of the control mix by 6.2% for the HTL-AP modified mix and 12.3% for the mix modified with commercial lignosulfonates (SAVEMIX4000).

Table 4 Tensile strength test data

Sample	Period of testing	Sample N	Weight, g	V, cm2	p, g/cm2	G, tensile, kN	R, tensile, MPa
Control	24 hours	1	560	256	2.19	1	3.88
		2	558	256	2.18	1.1	
		3	555	256	2.17	1	
	7 days	4	550	256	2.15	2.1	7.88
		5	540	256	2.11	2.1	
	28 days	6	551	256	2.15	2.6	10.13
		7	575	256	2.25	2.6	
		8	**584**	**256**	**2.28**	**2.9**	
Modification with HTL-AP	24 hours	1	560	256	2.19	0.8	3.13
		2	560	256	2.19	0.9	
		3	547	256	2.14	0.8	
	7 days	4	530	256	2.07	2	8.00
		5	535	256	2.09	2.2	
		6	525	256	2.05	2.2	
	28 days	7	529	256	2.07	2.6	9.50
		8	541	256	2.11	2.6	
		9	538	256	2.10	2.4	
Modification with SAVE-MIX4000	24 hours	1	569	256	2.22	0.8	3.25
		2	571	256	2.23	0.9	
		3	565	256	2.21	0.9	
	7 days	4	572	256	2.23	2.1	8.06
		5	564	256	2.20	2.2	
	28 days	6	552	256	2.16	2.5	8.88
		7	541	256	2.11	2.4	
		8	545	256	2.13	2.2	

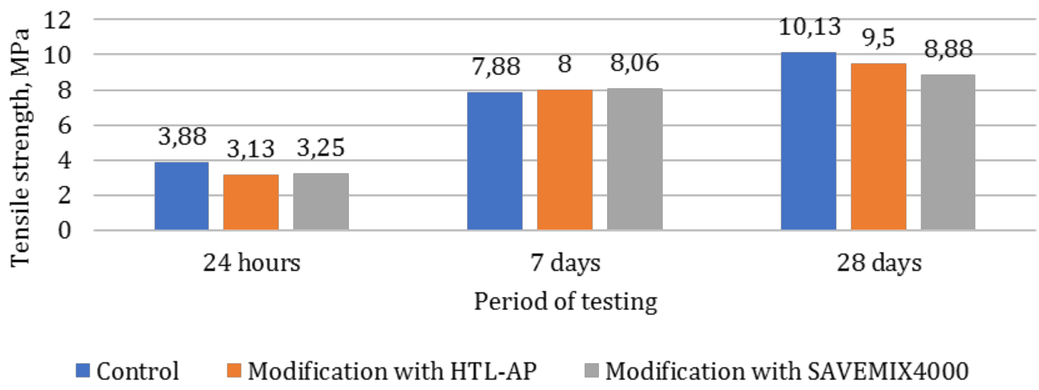

Fig. 3. Tensile strength

The results of the compressive strength measurements are shown in Table 5 and Fig. 4. Similar results were obtained in the compressive strength tests. It can be seen that on the first day, the strength of the modified concretes is 20% lower than that of the control concrete, indicating that lignosulfonates acts as a retarder in the hardening of the concrete mix. Further tests showed that the ultimate strength of the modified concretes (at 28 days) was lower than that of the control mix by 7.1% for the mix modified with HTL-AP and by 14.5% for the mix modified with commercial lignosulfonates (SAVEMIX4000).

Table 5 Compressive strength test data

Sample	Period of testing	Sample N	Weight, g	V, cm^2	p, g/cm^2	P, compr., kN	N, compr., MPa
Control	24 hours	1	560	256	2.19	34.53	21.58
		2	558	256	2.18		
		3	555	256	2.17		
	7 days	4	550	256	2.15	95.03	59.39
		5	540	256	2.11		
	28 days	6	551	256	2.15	135.70	84.81
		7	575	256	2.25		
		8	584	256	2.28		
Modification with HTL-AP	24 hours	1	560	256	2.19	25.05	15.66
		2	560	256	2.19		
		3	547	256	2.14		
	7 days	4	530	256	2.07	89.57	55.98
		5	535	256	2.09		
		6	525	256	2.05		
	28 days	7	529	256	2.07	126.00	78.75
		8	541	256	2.11		
		9	538	256	2.10		
Modification with SAVE-MIX4000	24 hours	1	569	256	2.22	24.13	15.08
		2	571	256	2.23		
		3	565	256	2.21		
	7 days	4	572	256	2.23	97.15	60.72
		5	564	256	2.20		
	28 days	6	552	256	2.16	116.07	72.54
		7	541	256	2.11		
		8	545	256	2.13		

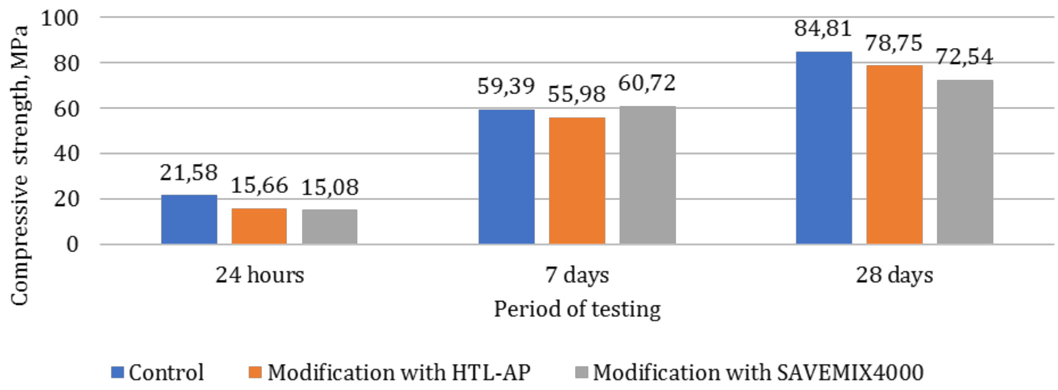

Fig. 4. Compressive strength, MPa

The data presented above confirm the technological feasibility of using HTL-AP as a modifier. Besides that, the beneficial use of HTL-AP will reduce the environmental burden created by wastewater from the hydrothermal liquefaction process. In addition, the use of these wastewater will reduce the resource intensity of the construction industry, as primary resources will not be consumed for the production of concrete modifiers. Ultimately, the introduction of this technology will ensure a reduction in the resource intensity of the ecological and climatic footprint of human industrial activity.

Table 6 Comparison of economic parameters of various concrete modifiers

Name of modifier	Origin	Price	Consumption, % from the mass of cement
SikaPlast-520N	Sika, Russia	1.9 USD/l	0.3
Sodium Lignosulfonate	SIDLEYCHEM LTD., China	2 USD/kg	0.1
Concrete Plasticizer	A.S.Chemical, India	0.9 USD/l	0.2-0.4
HTL-AP	Depends on the place where the process is implemented	0	6.6

Table 6 shows a comparison of the economic parameters of commercial concrete modifiers with HTL-AP. As we can see, replacing traditional modifiers with HTL-AP will not only reduce the environmental impact on the environment, but will also reduce the cost of concrete by 1.46 USD per ton of mortar mixture.

Thus, a number of important conclusions can be drawn from the research conducted:

- Lignosulfonates and their derivatives are present in significant amounts in the composition of the aqueous phase of hydrothermal liquefaction processes.
- The aqueous phase from the hydrothermal liquefaction of excess sludge has significant resource potential and can potentially be used as a modifier in fine-grained concrete.
- The results of the tests on the use of HTL-AP as a concrete modifier show an increase in the mobility of the mixture by 40% in comparison with the control sample. At the same time, the admixture has a retarding effect on the concrete hardening process, which is confirmed by the results of tests on the strength of samples after 1 day.
- The application of HTL-AP leads to a slight decrease in the strength properties of the concrete (compressive strength by 7.1%, fracture strength by 6.2%), while the decrease in the strength properties when using commercial solutions was much more significant at 14.5 and 12.2%, respectively.
- The comparison of the effectiveness of the commercial solution with the studied solution showed that HTL-AP has a higher effectiveness in retarding the setting, as the setting rate of the HTL-AP modified concrete was 6.3% lower.

4. CONCLUSIONS

It can be concluded that the use of HTL-AP as a curing retarder is well justified, as the positive effect on the concrete curing process has been confirmed. This technological solution will allow solving a whole complex of problems, including ensuring the use and maximum utilization of the resource potential of HTL-AP; it will provide valuable and at the same time cheap concrete modifiers.

Author Contributions: Conceptualization: Y.K. and M.D., O.B.; methodology: M.D.; formal analysis: M.D.; investigation: I.G.; resources: M.D., O.B. and Y.K.; data curation: M.D.; writing—original draft preparation: Y.K.; writing—review and editing: Y.K. and M. D.; visualization: I.G.; supervision: M.D.; funding acquisition: O.B., Y.K. All authors have read and agreed to the published version of the manuscript. Authorship should be restricted to individuals who have made significant contributions to the research.

Funding: This research was funded by RUSSIAN SCIENCE FOUNDATION, grant number 23-24-00064, https://rscf.ru/project/23-24-00064/.

Conflicts of Interest: The authors declare no conflicts of interest

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