

A determination of areas of biocorrosion development on the route of a main gas pipeline in the Western region of Ukraine

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Abstract: A determination of soil corrosivity on three sections of a main gas pipeline in the Western region of Ukraine has been carried out. We have distinguished areas of development of biocorrosive processes with the participation of sulphate-reducing bacteria (SRB). Soil evaluation by the degree of corrosivity has been conducted. We used weight, titrimetric, gravimetric methods, pH-metry, ten-fold limit dilution method and Postgate nutrient medium B for culturing SRB. It has been established that the soil in the first area selected along the lower generatrix of the main gas pipeline has a high degree of corrosivity. It was ascertained that corrosive metal damage increases in the "ferrozone" with the growth of metal exposition time from 24 to 72 hours. The soil selected in the middle part and from above the pipeline refers to soils with a normal degree of corrosive activity. Innovative biostable insulating coatings based on bituminous-polymerous mastic MBPID-1 have been developed, modified with organic inhibitors from the class of quaternary ammonium salts and amines. Regularities of influence of nature of nitrogen-containing corrosion inhibitors of industrial production M, N, L, H and K on the corrosion rate of 17G1S steel for 180 days have been established. It was found that the corrosion rate of steel specimens remained unchanged throughout the study in variants with the presence of inhibitors H and K in the test systems, which indicated their bioresistance to the effect of SRB bacteria.

Keywords: main gas pipeline, soil corrosion and biocorrosion, soil corrosivity, biostability, insulating coating

INTRODUCTION

One of the important elements of ensuring the ecological and energy security of Ukraine is the effective functioning of the gas transmission system, the main components of which are underground main gas pipelines (MG). The MG are potentially dangerous objects, with large energy potential which could cause negative environmental impacts in emergency situations due to

the pollution of the air basin, soil mass, and hydrosphere by products of energy transportation (Rozhonjuk et al. 2000, Shyshkivs'kyy & Havryl'tsev 2000, Kryvenko 2005). Both Ukrainian and international authors (Mironyuk & Pronina 2001, Serednyts'kyy et al. 2004, Banakhevych & Vil'chans'kyy 2007, Van den Brand & Kenter 2011) have analyzed the reasons for the failures of pipelines. The main reasons for failures are metal corrosion, intentional damage, factory and

construction defects that create environmental risk. Accident statistics on the linear part of the MG showed that the dominant factor among more than 90% of failures in the gas transportation system is the corrosion factor, which accounts for between 15 and 50% (Shyshkivs'kyy & Havryl'tsev 2000). Accidents and failures on the linear part of the main gas pipelines lead to significant gas losses, costs connected with the liquidation and damage control and gas supply shortfalls. Average gas losses during accidents total 2401 million m. The total annual downtime of gas pipelines due to accidents is 3,554 hours (Hrudz et al. 2009).

During the long-term operation of underground gas pipelines, the destruction of metal due to soil and microbial corrosion is intensified. The environmental impact caused by the destruction of pipelines following their long-term exploitation (more than 30 years) is very important. The degradation of insulating coating due to soil and microbe corrosion are formed in the process of the prolonged exploitation of underground gas pipelines laid in soils of various corrosivity in some regions of Ukraine. One of the environmental aspects providing effective and safe operation of underground pipelines is monitoring of the corrosivity of the soils in which they are laid. The processes of the corrosion of pipelines in the underground environment are influenced by a number of factors, namely: the chemical nature

of soils, their moisture, resistivity, redox potential, the presence of association of soil microorganisms. Due to the influence of these factors, the degradation of the insulating coating occurs (Fig. 1), which promotes the growth of plant roots, leading to further cracking that result in the intensification of the corrosion of the pipe metal in the soil electrolyte.

An important factor in ensuring the trouble-free operation of underground gas pipelines is the protection of their surface from soil corrosion by high-quality insulating coatings. Despite the presence of modern types of insulation (polyurethane, polyepoxide, three-layer polyethylene), the tendency in Ukraine has been to retain the much cheaper "traditional" mastic and mastic-tape coatings based on bitumen. The share of such mastic and mastic-tape coverings in the structure of insulating coatings in the gas sector of the economy exceeds 94% (94.6) (Polutrenko 2013). However, we cannot underestimate the destruction of the protective insulating coating and the pipeline metal under the influence of associates of soil microorganisms, among which sulfate-reducing bacteria (SRB) play a dominant role. The corrosivity of soils in Ukraine is presented in Figure 2, its analysis shows that the rate of soil corrosion in all regions is in the range of 0.1–0.36 mm/year, while the prevalence of biocorrosion is heterogeneous in different regions.



Fig. 1. Degradation of insulating coating (A) and metal damage (B) of main gas pipeline

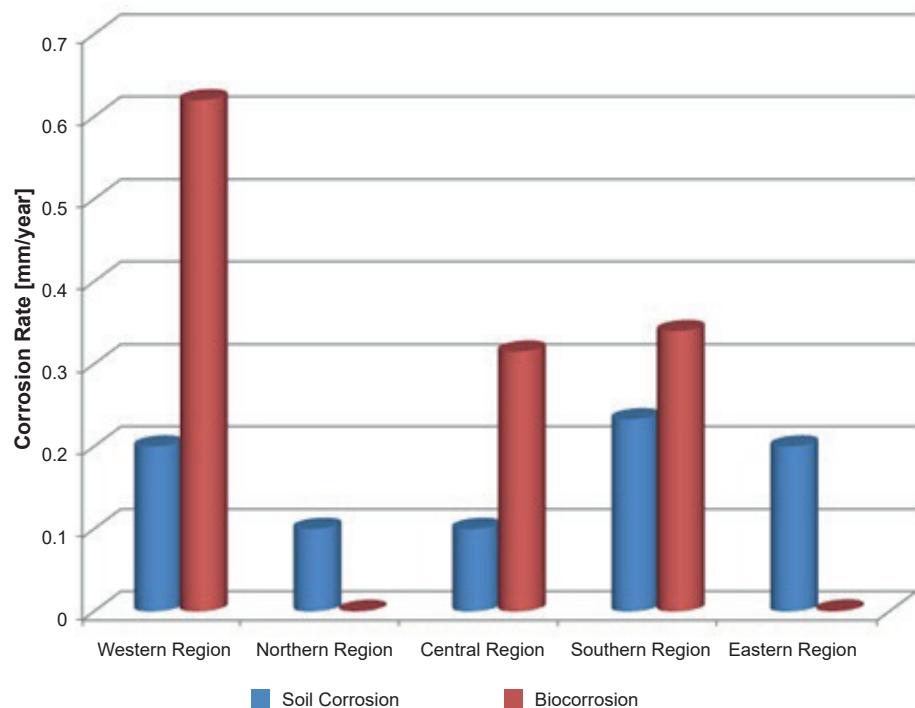


Fig. 2. Soil corrosivity in Ukraine

According to the available data (Stepachov & Lemeshyn's'kyy 2016), the biocorrosion rate is the highest in the Western region and reaches 0.62 mm/year, it is slightly lower in the Southern and Central regions and is at the level of 0.36–0.32 mm/year. Biocorrosion activity in the Northern and Eastern regions was not revealed. In view of this, the role of microbiological processes in the soil corrosion of underground oil and gas equipment is extremely important and must be taken into account. Due to the microbial destruction of the protective insulating coating as a result of the action of the heterotrophic unit of aerobic and anaerobic bacteria: HOB, FRB, DNB, and SRB, the physical-mechanical properties of protective materials change (Andreyuk et al. 2005, Kozlova et al. 2008), their solidity, elasticity, adhesive characteristics are reduced, whereupon the main function of the coating is lost – the protection of metal from corrosion. The degradation of insulating coatings in the process of exploitation is another aspect of the formation of the ecological danger of emergencies as a result of increased corrosive damage to the pipeline metal. One of the methods for improving

the anticorrosive protection of underground pipelines is the development of formulations of new compositions of anticorrosive coatings with the introduction of corrosion inhibitors and biocides (bactericidal additives) in order to improve anti-corrosive insulating characteristics and the elaboration of regimes for qualitative preparation of a coated surface. Thereon hangs the effectiveness of the protective coating, its durability and the strengthening of the metal surface. The purpose of this work is to conduct the research to determine the corrosive activity of soils with the allocation of areas of biocorrosion and the development of innovative bio-resistant insulation coatings.

METHODS

The object of the study were soil samples taken from different sections of the main gas pipeline in the Western region of Ukraine. Bituminous-insulating mastic brand MBPD-1 was chosen as the basis for the production of modified mastics and primers using corrosion inhibitors with biocidal properties. Despite a large number of field and

laboratory methods for determining the corrosivity of soils (Zhukov & Khramikhin 1964, Strizhevskiy 1986), a correct assessment can only be made after studying soils by means of different methods and comparing the results of these studies, taking into account local geographical and climatic conditions. The first stage of the research was devoted to the monitoring of corrosivity of soils, which covered the complex of determination of factors such as acidity of soils, mass fraction of moisture, presence of sulphate ions, and also loss of mass of the metal, determined by the gravimetric method,

characterizing the corrosive destruction of the metal. The sampling of soil was carried out according to the scheme shown in Figure 3.

Soil samples with a diameter of 529 mm were taken for analysis at three plots on the route of the MG. The first two sections were at a distance of about 1000 m apart, the soils in the third section were selected according to the following scheme in the direction of gas supply (Fig. 4).

The samples of soils taken from the areas under study differed greatly in terms of their granulometric composition (Tab. 1).

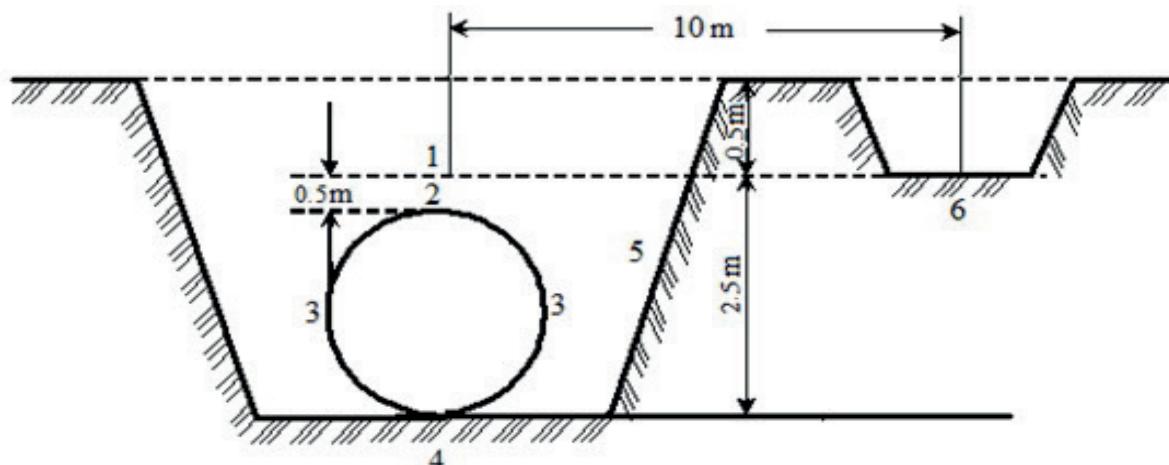


Fig. 3. Scheme of soil sampling when digging the pipelines: 1 – at the depth of 0.5 m, 2 – along the upper generatrix of the gas pipeline, 3 – along the side generatrix of the gas pipeline, 4 – along the lower generatrix of gas pipeline, 5 – along the wall of the test pit at point 3, 6 – at the distance of 10 m from the gas pipeline at the depth of 0.5 m (control)

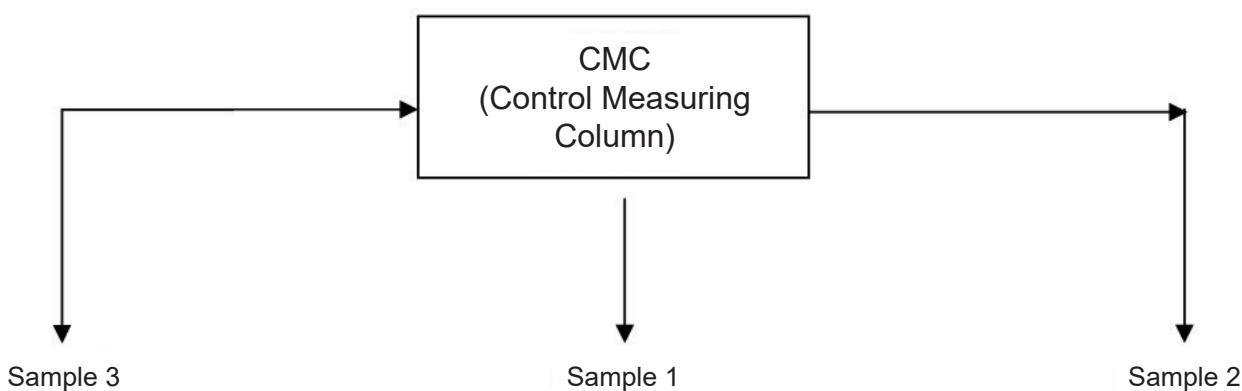


Fig. 4. Scheme of soil sampling on the route of MG (site 3)

Table 1
Granulometric composition of soil from the areas under study

Section No.	Place of sampling	Granulometric composition
1	sample No. 1 (lower part)	black and grey dense clay with high iron content
	sample No. 2 (upper part)	black and grey sandy soil of medium grained composition with residues of plant roots
	sample No. 3 (middle part)	grey and brown loam with the inclusion of crushed stone
2	sample No. 1 (lower part)	yellow and grey dense clay
	sample No. 2 (upper part)	loam with seams of sandy soil
	sample No. 3 (middle part)	loam with brown seams of iron oxides and manganese
3	sample No. 1	yellow and brown clay
	sample No. 2	black and gray clay
	sample No. 3	dark brown clay

Soil samples were dried out in a drying cabinet at 95–98°C, chopped in a porcelain mortar, sifted through metal sieves and soil fractions were selected (≤ 2 mm) for further research. Since one of the important factors characterizing the soil corrosivity is acidity, it is important to trace the change in the acidity of the soil, taken from above and from below of the pipeline array, and from the middle along the gas flow. Acidity of soils is due to the presence of hydrogen ions, the concentration of which is expressed in pH. The value of pH in the soil varies depending on the total mineralization of subsoil waters and the presence of carbonic and mineral acids, acid and basic salts in them (Krykunov et al. 2003).

According to the pH value, the soils were distinguished as follows: strongly acid (3–4.5), sour (4.5–5.5), weakly acid (5.5–6.5), neutral (6.5–7.0), weakly alkaline (7.0–7.5), alkaline (7.5–8.5), strongly alkaline (more than 8.5) (Strizhevskiy 1986). Actual acidity is caused by hydrogen ions located in the soil electrolyte. Its amount is determined by the results of the analysis of water extraction from the soil. Actual acidity characterizes the soil acidity at the time of its determination. The actual acidity of the soil samples taken was determined by the procedure (Krykunov et al. 2003) with the help of a multipurpose indicator and a pH-meter of the mark pH 150MI (Tab. 2).

Table 2
Determination of pH, presence of SO_4^{2-} and mass fraction of moisture in soil samples of MG

Index	Section No.	Place of sampling		
		lower part	upper part	middle part
pH	I	5.25	5.57	5.51
	II	5.07	5.72	5.44
	III	6.78	6.83	6.68
Presence of ions in subsoil waters (qualitative sample)	I	++	±	+
	II	+++	+	++
	III	-	-	-
Mass fraction of moisture [%]	I	18.21	4.74	10.48
	II	19.73	5.62	12.55
	III	11.64	4.14	8.22

"+" – formation of mud was observed; "±" – a weak mud; "-" – mud is missing.

RESULTS AND DISCUSSION

The obtained results showed that main gas pipeline on the route sections under study were laid in heterogeneous acid soils. The soils on the first two sites are weakly acid soils by the value of pH, which indicates to the soil corrosivity in relation to the steel, soils on the third section are neutral, which, by the value of pH, are corrosively inactive. To exclude the possibility of increase of corrosion processes by the influence of sulphate ions (SO_4^{2-}), water extracts of soils were analyzed for SO_4^{2-} by qualitative reaction with the aqueous solution of barium chloride (Tab. 2). The fall of white sediment (appearance of turbidity) points out to the presence of SO_4^{2-} ions in subsoil waters. The results of the qualitative analysis demonstrate that subsoil waters from below the pipeline are the richest in sulphate ions, the content of which is reduced in the direction towards the top of the pipeline. The content of sulphate ions points out the presence of metal sulphates in soils (possibly Na_2SO_4 , FeSO_4 , MgSO_4 , $\text{Al}_2(\text{SO}_4)_3$), which is a prerequisite for the development of biocorrosion with the participation of sulphate-reducing bacteria (SRB). Sulphate ions were not detected in the soil of the third area of the route under study. Another important factor for the correct estimation of soil corrosivity is soil moisture. The determination of soil moisture by gravimetric method from the sections of the

pipeline route under study display (Tab. 2) that the dampest soil is located below the pipeline under investigation, which contributes to the strengthening of corrosion processes.

To give a complete snapshot of the corrosivity of soils, we also analyzed the loss of metal mass, calculated by means of the gravimetric method. Therefore, the metal rods, weighed previously on the analytical balance of VLF-200, were placed in steel cylinders that served as a cathode. Each of the cylinders was filled with an appropriately prepared clay fraction (clay selected from above, from below, in the middle of the pipeline). The prepared cylinders were left for specified time under the action of constant electric current 6 V.

After the exposition, the metal rods were taken out of the soil, subjected to mechanical and chemical treatment for the removal of corrosion products from their surface. It should be noted that due to washing the rods with the solution of hydrochloric acid, there was a sharp smell of hydrogen sulphide, formed as a result of destruction of the black sediment of ferrous sulphide, which covered the metal rods. The formation of biogenic ferrous sulphide shows development of microbiological processes in the soil electrolyte with the participation of sulphate ions, which caused intensification of the metal corrosion. In Figure 5 you can see the dynamics of mass loss of metal on the first section of the route under study.

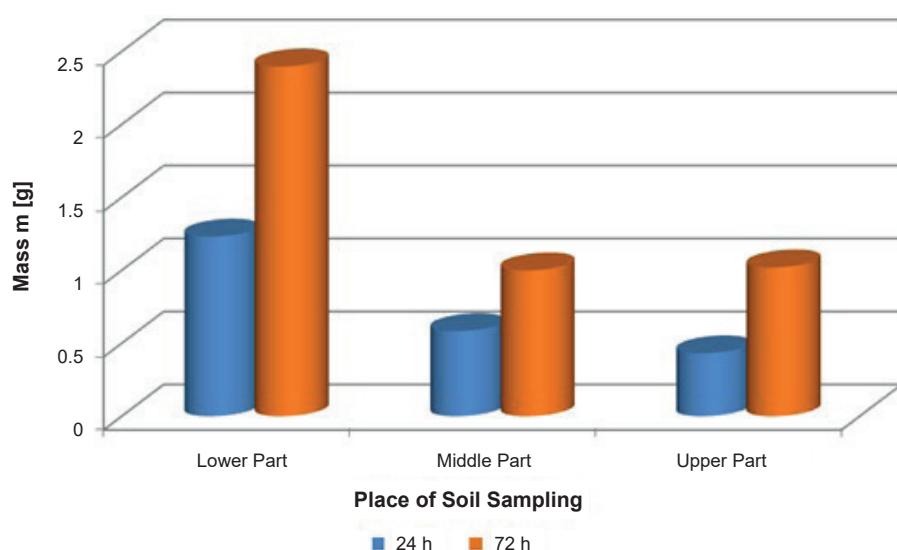


Fig. 5. Loss of the metal mass on section 1 of the route of MG

The analysis of the experimental results of mass loss of the metal shows that the soil with the largest corrosivity is located below the pipeline along so-called lower generatrix of the pipeline, which is characterized by the prevailing content of sulphate ions. This zone is in direct contact with the metal surface of the pipeline, the so-called "ferrosphere" (Andreyuk et al. 2005). We have ascertained that due to the corrosion damage in the metal of the pipeline in the soil occurring in this zone, there is accumulation of ions of ferrous iron and decrease of redox potential that intensifies the activity of SRB, which stimulate corrosion processes, the intensity of which is maximum in dense corrosively dangerous clay soils. The metal corrosion increases with growth of the exposition time up to 72 h in the zone adjacent to the bottom of the pipeline. According to the data obtained, the soil from below the gas pipeline has a high degree of corrosivity by the loss of metal mass, while the soil taken from the middle part and from above the pipeline, refers to soils with a normal degree of corrosivity. A somewhat different picture was observed for the loss of metal mass determined on the second section of the trace of the MG (Fig. 6).

The results of the research show that in 24 hours the most marked corrosive damage is also characteristic of the soil from below the pipeline on section 2 of the route of the MG. The

intensification of corrosive damage was observed with the growth of time of the metal exposition in the soils up to 72 hours, which was accompanied by an increase in the metal loss for all soil samples. However, a slight difference was established in the mass loss of the metal (up to 10%) in the soil, taken from below and from the middle part of the pipeline, and characterized by an increased degree of corrosivity. It should be noted that at the end of the exposition, all the metal rods were covered with black sediment of ferrumsulphide, when removing it a strong smell of hydrogen sulfide was felt. The formation of biogenous hydrogen sulfide is connected with the reduction of sulphate ions to sulfide ions by microorganisms. A biogenic ferrous sulfide is created as a result of their interaction with Fe(II) ions. It acts as a cathode in relation to the metal of the pipeline, which leads to the intensification of corrosion damage with the participation of SRB bacteria.

The most intensive corrosion occurs in soil sample No. 3 (Fig. 7) on the third section of the route. The corrosion processes "fade" in the direction of the gas flow, as evidenced by the lower values of Δm for samples No. 1 and No. 2 (which may be due to moisture oversaturation of the soil, which in turn blocked the access of oxygen to the metal and led to the reduction in the rate of corrosion in the soil).

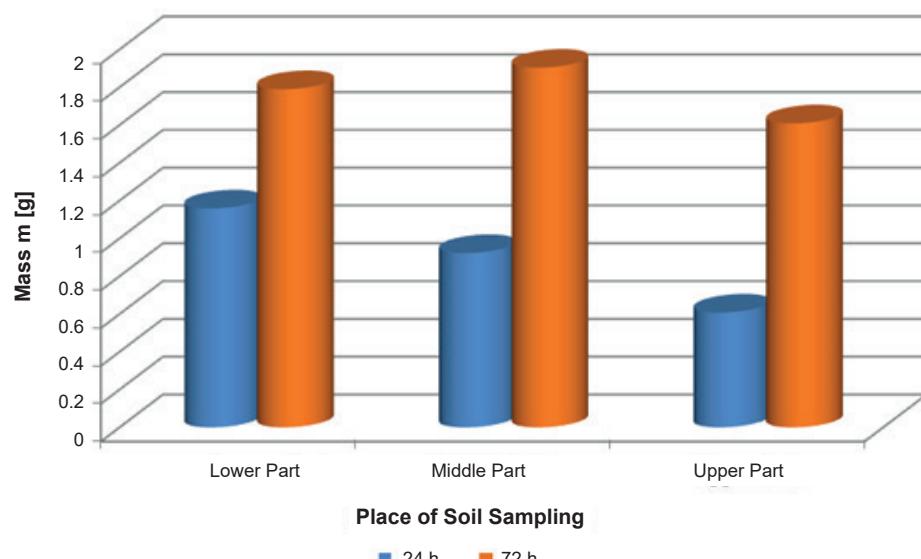


Fig. 6. Loss of the metal mass on section 2 of the route of MG

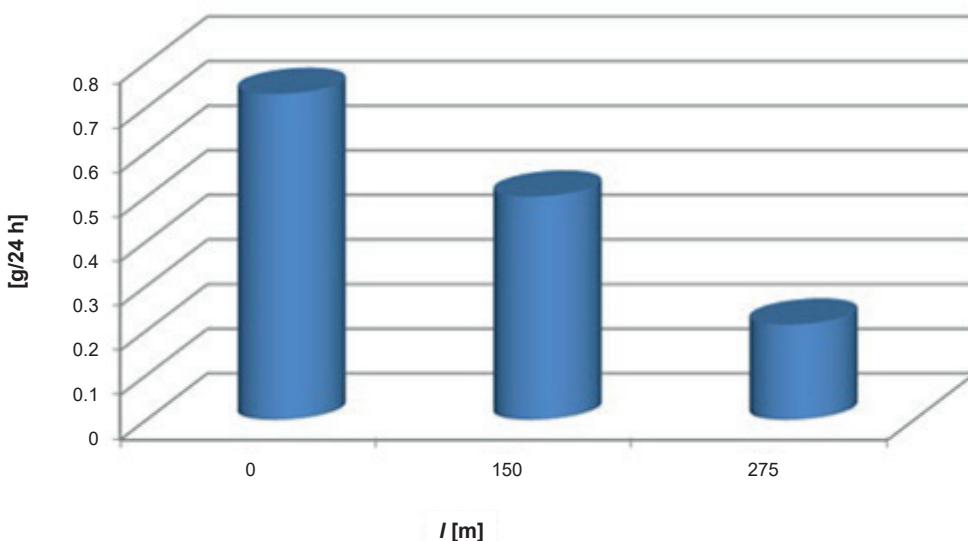


Fig. 7. Loss of the metal mass on section 3 of the route of MG

Thus, it was found that the degree of soil corrosivity is an important environmental aspect in the anticorrosive protection of underground pipelines. For reliable and efficient operation of underground pipelines in order to prevent soil corrosion, including microbiological corrosion, we need insulating coatings modified by corrosion inhibitors (biocides), capable of resisting the attacks of soil microorganisms and the germination of plant roots that lead to the premature degradation of the insulating coating and the intensification of corrosion damage of the metal.

The second stage of experiments was devoted to the development of new compositions of inhibitory systems with increased anti-corrosive and biostable properties by modification of the MBPID-1 bituminous-polymerous mastic. Previous studies have found (Kryzhanivs'kyy et al. 2009) that the modification of the model mastic with organic corrosion inhibitors (diamines, quaternary ammonium salts (QAS), salts of naphthenic acids, ethanolamine, unsaturated fatty acids) allows bituminous polymerous mastics with increased adhesive parameters and improved plasticizing and hydrophobic properties to be produced. As the quality of the insulating coating depends on its biostability to soil microorganisms,

the developed samples of bituminous-polymerous mastics modified by corrosion inhibitors from the class of amines and QAS were transferred to the Institute of Microbiology and Virology of the National Academy of Sciences of Ukraine, where the laboratory tests on the biostability of these materials showed that modified bituminous-polymerous insulating mastics are biostable to SRB bacteria. This fact pointed out that given corrosion inhibitors not only slow down the electrochemical corrosion of the metal, but also display bactericidal properties.

For the first time, innovative coatings protected by patents were obtained with the participation of organic inhibitors of different classes, introduced in the primer and in the mastic (patent 822775 2008, patent 84769 2008, patent 84769 2010). The indicated inhibitors are well combined with the bituminous-polymerous base, enhancing its protective effect. The inhibitor from the class of QAS for 17G1S-U steel impeded growth of SRB bacteria almost completely showing a high degree of metal protection (95.74%) from biocorrosion (Polutrenko & Pilyashenko-Novokhatnyi 2013, Polutrenko et al. 2018). Inhibitors on the basis of derivatives of dioxodecahydroacridines (Polutrenko 2012), which are known to have

antimicrobial and antifungal activity, were also used in the work. We considered these inhibitors as a promising object for the purposeful search and modeling of the new generation of corrosion inhibitors (biocides).

We conducted laboratory studies to determine the laws of the influence of the nature of nitrogen-containing corrosion inhibitors of industrial production M, N, L, H and K on the rate of steel corrosion 17G1S for 6 months. The rate of the corrosion of metal samples immersed in the test-systems was determined gravimetrically.

The results of the research, presented in Figure 8, demonstrate that the corrosion rate of steel samples remained unchanged throughout the study period in variants with the presence of G and K inhibitors in test-systems indicating their bioresistance to the action of SRB bacteria.

The results obtained are of practical value, since the bioresistance of inhibitors H and K allows them to be used as modifiers for obtaining biostable insulating materials and as components for the modeling of innovative inhibiting systems to protect underground pipelines from microbiological corrosion.

We conducted tests of coatings in route conditions on the main gas pipelines in the Western and

Southern regions of Ukraine in the conditions of "Prykarpattranshaz". The results of the tests show that when applying the bituminous-polymerous mastic of mark MBPID-1, modified with biocides from the class of amines, the regulated physical and mechanical characteristics of the coating completely meet the values of regulatory documents. The technical specifications TU U No. 26.8-02070855-001:2010 *Bituminous-polymerous isolating modified mastic MBPIM-D* have been developed, which opens up prospects for mass production.

As the developed modified mastics on the bituminous-polymerous basis are biostable to the action of corrosively active soil microorganisms, in order to reduce the environmental risk connected with the intensification of corrosion damage of the pipeline metal under the influence of aggressive soil microorganisms, they can be used to re-insulate pipelines on such problem areas of the route, like marshy soils, muddy soils, salt marshes, soils with high humidity.

The introduction of innovative biostable modified mastics on the domestic market in the current practice of the reinsulating of pipelines will allow reliability to increase and reduce financial and labor costs for the maintenance of underground pipeline systems.

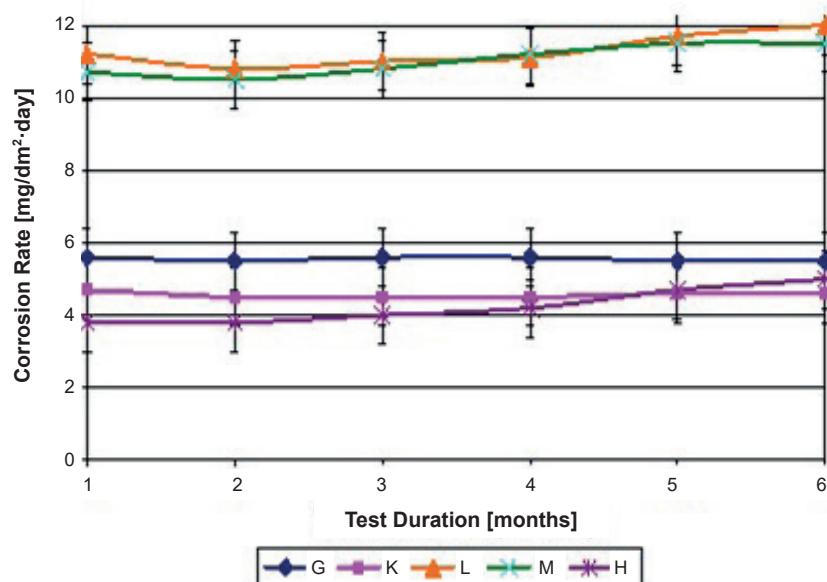


Fig. 8. Influence of nature of nitrogen-containing inhibitors on the steel corrosion rate 17G1S in the long-term experiment

CONCLUSIONS

The following results were obtained as a result of research.

1. A study was conducted to determine soil corrosivity in three areas of the MG in the Western region of Ukraine, which covered the determination of such factors as soil acidity, mass fraction of moisture, presence of sulphate ions, and also the loss of metal mass computed by the gravimetric method which characterized the corrosive destruction of the metal.
2. On the basis of the conducted research, the I and II sections of the pipeline development of biocorrosion with participation of SVB are allocated.
3. It has been established that the soil on the first section of the route, selected along the lower part of the MG, has a high degree of corrosivity and will lead to the acceleration of corrosion damage to the pipeline metal.
4. It has been experimentally established that the corrosion damage of the metal in the "ferro-zone", the soil zone adjacent from below the pipeline intensifies with an increase in metal exposition time in the soil from 24 to 72 h. The soil, taken from the middle part and from above the pipeline, refers to the soils with a normal degree of corrosivity.
5. Innovative biostable insulating coatings (patents Nos. 82775, 84769, 89709) have been developed on the basis of MBPID-1 bituminous-polymerous mastic, modified with the inhibitors from the class of organic quaternary ammonium salts and amines, to reduce environmental risks during prolonged exploitation of underground gas pipelines from soil and microbiological corrosion.

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