

# INVESTIGATIONS OF THE STRUCTURE OF WELDED JOINTS OF POLYMERS USING THE REHBINDER EFFECT

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## ABSTRACT

Welds of polymer materials, butt welded with a heated tool, are distinguished by a variety of structural components. Investigations of such welds allow studying the general fundamental processes of structure formation in the formation of welded joints of polymers. In this work, mechanical tests of welded joints of polymer pipes with a local load in the environment of surface active substances and with the use of the Rehbinder effect were carried out. Experimental specimens of pipe joints with a diameter of 63, 90, 110 and 160 mm from PE80 polyethylene were produced in the standard equipment for butt welding with a heated tool on the modes recommended by DBN V.2.5-41:2009. The tests were carried out in accordance with the requirements of the international standard ISO 22088 "Cracking under the influence of the environment" (ESC). The characteristic features of cracks propagation in the tests of the base material and the fusion zone of the weld were investigated. Using optical microscopy, the fracture surfaces of the specimens were investigated immediately after tests and after chemical etching in sulfuric acid solution. It was shown that in the fusion zone, a weakened "blurry" structure of polymer material is formed, which is fractured during a local load.

**KEYWORDS:** butt welding with a heated tool, supramolecular structure, mechanical tests, fracture surfaces

## INTRODUCTION

Supramolecular structure of polymer is very complex and divides for several levels depending on size of the structural elements. The spherulites of size at a level of 10  $\mu\text{m}$  are the main component for crystalline polymers. At the next level there are domains of crystalline-lamellaes of 10–100 nm size. A size of folded lamellaes is evaluated in 10 nm. An elementary folded link forming lamellaes are of 0.1–1.0 nm size limit.

Most of the engineering plastics have in their content various admixtures and fillers, therefore, examination of a material microstructure using light microscope is usually impossible [1]. Visual evaluation of the details of complex supramolecular structure is possible by means of investigation of the typical features of weld fracture under the effect of local mechanical loads [2, 3].

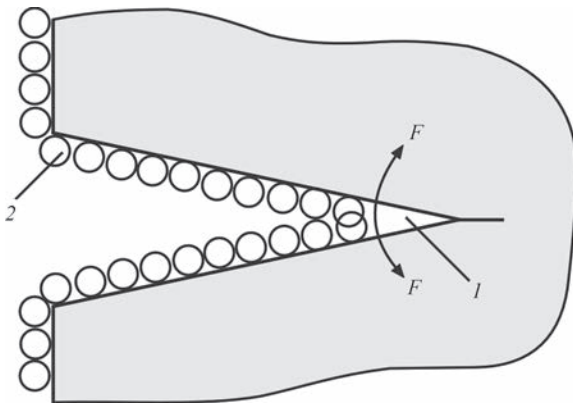
Since the polymers consist of the long flexible molecular chains, they have typical features of the material in a solid state, namely inhomogeneity, non-linearity and time dependence of mechanical properties [4]. Loss of strength by a polymer usually takes place in several steps after sufficient plastic deformations. The following terms are used in the theory of polymer fracture, namely failure, i.e. accumulation of structure defects, fracture — formation of cracks with development of new surfaces and rupture — division of body on separate parts. In other words, gradual loss of strength by polymer material in loading takes place due to expansion of existing and formation of new defects of supramolecular structure. The investiga-

tions of crack propagation are used for determination of different types of strength of polymer materials [5]. In this work there were carried out the investigations of strength of base material and welded joints of polymers using mechanical loading with additional stimulation of fracture with the help of surface-active substances (SAS).

The processes of fracture taking place in a solid polymer significantly depend on its physico-chemical interaction with environment. Such organic substances as alcohols, fatty acids, rosins, soaps contain in their molecule composition at the same time hydrophilic and hydrophobic groups and being called surface-active. SAS molecules in contact are intensively adsorbed on a surface of solid body that significantly decreases its surface energy and proportionally reduces body strength [6, 7].

Adsorption decrease of strength of solid bodies was titled as Rehbinder effect [8]. The effect has a versatile nature and observed for any solid materials.

In polymers development of cracks in material takes place through successive rupture of separate macromolecular chains. Crack propagation promotes formation of the new material surfaces which adsorb SAS. The SAS molecules covering the newly formed crack planes neutralize their high energy potential and try to move as close as possible to the very beginning of the crack. Even at absence of surface loads these molecules colliding between themselves create a fracture force  $F$  in a crack basis (Figure 1). It can reach significant value in the minimum gaps.



**Figure 1.** Scheme of SAS effect on crack development in solid body: 1 — crack start; 2 — SAS molecules on surface

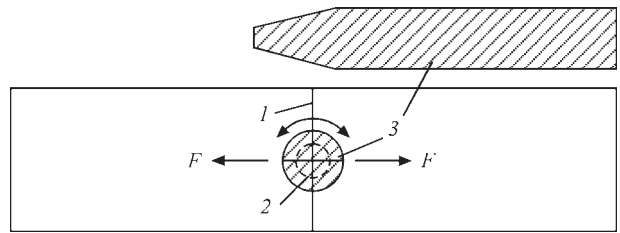
There are different procedures being used for generation of local loads and stress concentrators in a polymer specimen. International standard ISO 22088-1 [9] provides the definition of the process of crack formation in polymer material (plastic) which is located in a certain chemical environment and being under the effect of constant stress less than a value of yield strength. This process has a standard name — environmental stress cracking, ESC.

One of the most widespread methods of evaluation of strength to ESC is a bend strip method [10]. A cut is made on a plate from polymer material. It serves as a concentrator for crack initiation. Then the plate is bent in half in the place of cut developing constant mechanical stresses. Bent plates are located in SAS environment and after a while evaluate the number and size of the formed cracks. However, the load in this method is applied asymmetrically and at once for all width of the plate-specimen, therefore, this method is used for testing of base material and for polymer welded joints it is not applicable. The task of this work lies in performance of the comparative tests of strength of base material and weld material as well as evaluation of supramolecular structure of a transition fusion zone in the welded joints of polyethylene pipes.

The segments of the standard polyethylene pipes of 63, 90, 110 and 160 mm diameter designed for construction of gas pipelines made of polyethylene of PE80 grade according to the requirements of DSTU B V.2.7-73-98 were used in work. The pipes of black and yellow colors were used. The section of polyethylene pipes were joined using butt welds on standard equipment for butt welding with a heated tool applying the modes recommended by the standard [13].

#### METHOD FOR DETERMINATION OF CRACK FORMATION RESISTANCE

Resistance of base material and weld material to crack formation was determined according to a procedure of ISO 22088-4 [11] by means of formation of local



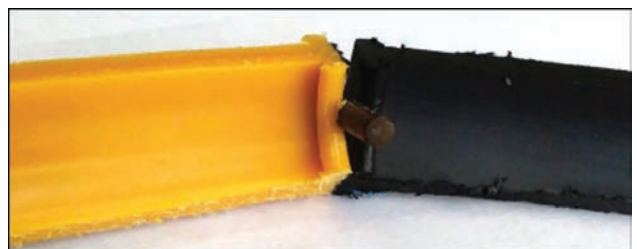
**Figure 2.** Scheme of initiation of fracture of welded joint using hole and cylinder rod: 1 — weld fusion line; 2 — through hole in specimen; 3 — cylinder rod

loading in a necessary area of a polymer specimen. For this a pin impression method was used, the essence of which is schematically shown in Figure 2. A lamellar specimen with a weld in the middle is cut out from a welded joint. A hole of 3–4 mm diameter depending on specimen size is drilled on a fusion line. A cylinder rod of somewhat larger diameter with a cone tip is impressed in the hole that creates in the specimen a total tensile load  $F$  and tangential loads on a hole perimeter  $f$ . The works using this method have already been carried out at the E.O. Paton Electric Welding Institute (PWI) for comparison of strength of a butt welded joint of the polyethylene pipes in the different points of butt orbit [12].

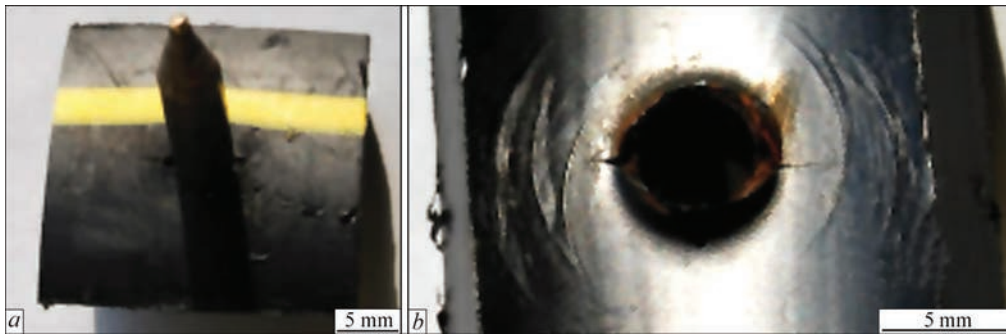
For all the tests by ISO 22088-4 procedure there were used a rod with a cone tip varying the relationship of diameters of hole and rod to change of a value of stresses in the specimen (Figure 2). After immersion of the rod in the hole and appearance of local load the specimens with welds were placed in a SAS water solution (grade OP-7) at up to 80 °C temperature and hold there from several minutes to hours till crack appearance. The joints from black and yellow pipes were made in order to ease the study of weld fusion line. In most of the cases at local initiation of stresses using a rod the welded joints fracture close to a fusion line (Figure 3). The difference in pipe colors provides the possibility for good evaluation of belonging of polymer material to one or another pipe part on the fracture surfaces.

The photos of specimens were made using digital camera Nikon D350 (NIKON Corp, Tokyo, Japan).

The magnified images of the surfaces of specimens were made by the same camera imbedded in a



**Figure 3.** Specimen of butt welded joint of polyethylene pipes fractured after testing



**Figure 4.** Cracks in specimens from base material of polyethylene pipes of gas grades PE80 with wall thickness 10 mm on outer (a) and inner pipe surface (b)

light polarizing microscope Versamet-2 (UNITRON, New-York, NY, USA).

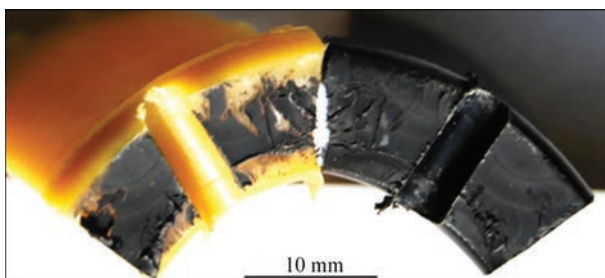
### INVESTIGATION RESULTS

Polyethylenes of pipe grades PE80 and PE100 are sufficiently ductile materials with high crack resistance. Therefore, the small cracks in testing of base metal of polyethylene pipes appear only after holding under stress in SAS environment for several hours (Figure 4). The cracks in the base material are of small length and propagate on both sides of the stress source along a straight line. A direction of crack on the outer surface of a pipe exactly matches with a longitudinal axis of the pipe that indicates some structural organization of material. Cracks on the inner pipe surface appear in a transverse direction relatively to the axis direction, they do not have through nature and form on the surfaces of specimens in the places where stresses and effect of SAS are the biggest.

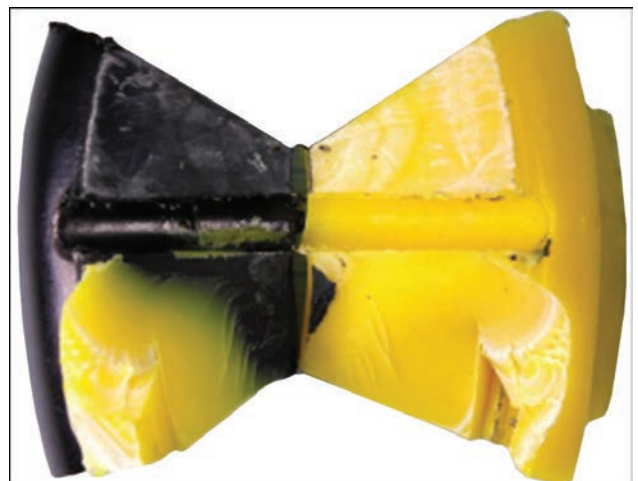
The fracture surfaces of pipe specimen of PE80 polyethylene with outer diameter 110 mm and wall thickness 10 mm (Figure 5) demonstrate complete cohesion nature of the welded joint, i.e. the fragments of material of one element of the joint are present on the fracture surface of another one. The specimen of welded joint of pipes of 160 mm diameter and wall thickness 14.6 mm fractured inhomogeneously during testing (Figure 6). The part of specimen fractures exactly along the fusion surface, another part demonstrates a joint nature of crack propagation along the weld and base material of the pipe. At significant pipe

wall thickness the direction of crack propagation can change and deviate from a failed fusion zone.

Analyzing a magnified image of the crack formed in fracture of the welded joint (Figure 7) it is possible to judge a nature of a transition structure being formed after cooling of molten metal inside the weld. A solid spherulitic structure along a delimitation line is not observed. Obviously, the monoliths of two parts are joined through the finer arranged structures of fibrils, folded domains or micelles. Therefore, a line of joining in a transition structure appears to be the weakest place. Tension of the supramolecular structures (Figure 7, below) that hold joint elements starts at local loading. It takes place slowly under standard conditions and sufficiently quick in SAS environment. Tensile structures reach a strength limit and tear provoking further crack propagation. If such failed structures in the weld have local nature, the crack ruining them stops or changes the direction meeting stronger structural areas. In other case, the transition structures are uniformly distributed along the whole area of butt orbit, therefore, fracture takes place in this plane or its vicinity. Only the remnants of torn transition structures are observed on the fracture surface (Figure 7, upper part).

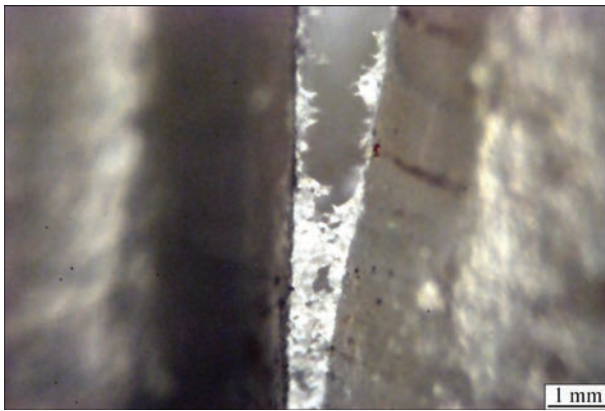


**Figure 5.** Fracture surfaces of welded joint of pipes from PE80 of 110 mm diameter and wall thickness 10 mm



**Figure 6.** Fracture surfaces of welded joint of pipes from PE80 of 160 mm diameter and wall thickness 14.6 mm



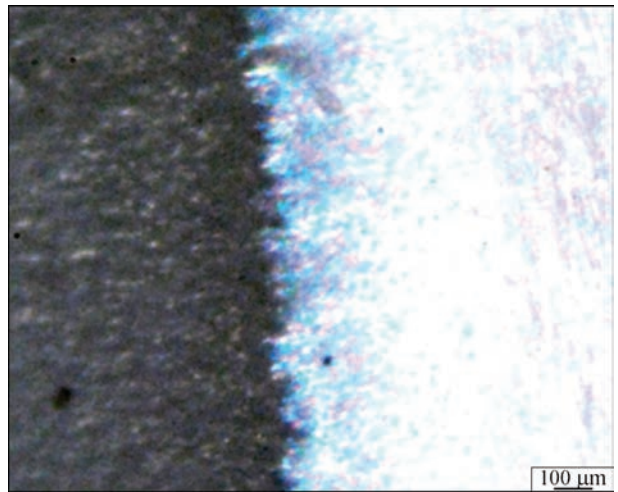


**Figure 7.** Crack propagation at fracture of welded joint of polyethylene pipes,  $\times 10$

The fusion line of two pipe specimens of different color polyethylene (Figure 8) demonstrates partial interpenetration of the supramolecular domains of black and yellow polyethylene (white on the photo). An interface profile has a non-uniform jagged nature, probably formed under effect of random factors. Such a profile apparently can not be a consequence of a joint flow of melts of two parts being joined under effect of upset force. A transition zone demonstrates the sharp elongated elements of black and yellow material which mutually pass through the fusion line “diluting” it. It is logical to assume that the deeper such interpenetration of the materials in weld formation, the stronger formed welded joint will be. The single driving force of such interpenetration is a heat movement of the segments of macromolecular chains at elevated temperature. The velocity of such movement is limited and dramatically drops with temperature decrease. Therefore, formation of sound welded joint requires staying of the material in a fusion zone for some time at elevated temperature.

Information on nature of microstructure of a polymer can be obtained by means of examination of their fracture surfaces. Fracture of any material is formation of the new surfaces. In given case a fracture of the specimens of welded joints of polyethylene pipes is a process taking place relative to slow propagation of a crack plane from the place of its initiation and nucleation. A polyethylene of high density is a semi-crystalline material. In it the areas of ordered and amorphous material border each other on a supramolecular level. The amorphous areas and intermolecular cavities also accumulate different admixtures and oligomers. Crack propagation takes place directly on failed, amorphous areas, thus allows revealing material structure.

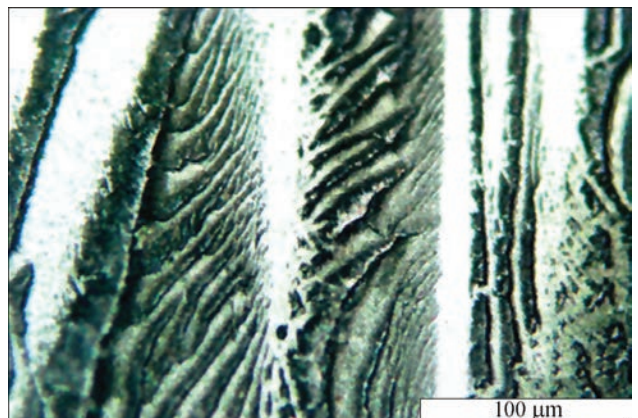
The fracture surfaces were examined using the light microscope on the black specimens since dark material allow obtaining more contrast image. A typical example of the directed structural formations in a



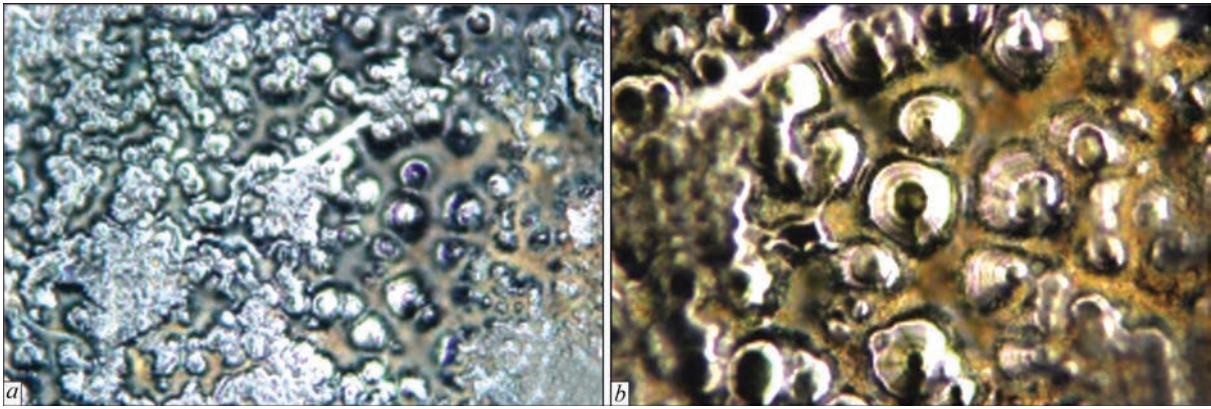
**Figure 8.** Line of welded joint of polyethylene pipes of different colors at  $\times 20$  magnification

bulk of polymer material is given on Figure 9. A series of objective and random factors is superimposed on the process of crack propagation. Therefore, on the fracture surface we can observe the elongated curvilinear structures, which are demonstration of a regular spherulitic structure and a randomly located area, formed due to crack “wandering” in the sections with relatively isotropic structure.

A spherulitic structure of a crystalline polymer is formed non-uniformly depending on heat conditions at a level of micron sizes and presence of necessary amount of crystallization nuclei. During formation of the ordered structures, low-molecular substances are displaced in the areas between them. Randomly formed spherulites in turn form the supracrystalline structures, which in fracture form a surface similar to double-sided lamellae only of larger scale. It is well observed on Figure 9 that these lines are the double-sided or semi-round formations with deep, sharpened to bottom cavities between them. It is necessary to take into account that such a surface was formed not as a result of some special treatment or etching, but due to free process of material fracture. Such a surface indicates significant physical inhomogeneity.



**Figure 9.** Fragment of fracture surface with  $\times 10$  magnification



**Figure 10.** Area of ordered spherulite structures, magnification: *a* —  $\times 10$ ; *b* —  $\times 20$

generality of the material, in which areas of strong and failed material alternate in orderly manner.

The double-sided ordered formations border with more extended elongated linear areas on the surface. Obviously, in these areas the material on a microlevel created less resistance to crack propagation and fracture took place linearly remaining on the surface the traces made as “under the ruler”. There are also areas with disordered structure of the surface, which, obviously, correspond similar areas of structure in a bulk of material. Depressions in the surface between double-sided structures actually have uniform homogeneous structure. Obviously, this is a consequence of the fact that the failed areas between the crystalline areas have own certain minimum size, which appear on the surface in a form of filamentary formations of almost similar width. It can be seen that such nature of a surface is present only in some certain places, therefore, it appear only at match of several necessary factors in the process of crack propagation.

Etching of the specimen in a sulfuric acid solution helps clearly reveal crystalline and amorphous areas on the surface of polymer. The amorphous failed areas dissolve significantly quicker than the crystalline ones, therefore appear on the surface in form of depressions at a background of higher ordered areas. Fracture surfaces of the specimens were subjected to etching and examined using light microscope (Figure 10).

The areas of surface with linear ordered structure almost do not undergo the changes after etching. Obviously, these are areas with the highest level of crystallinity and percent of amorphous phase here is small. Therefore, after etching the surface contains only narrow longitudinal bands that were formed after removal of small amount of failed material. The zones of spherulite structure are characterized with great non-uniformity of surface and presence of round-shaped formations. Since crystallization of spherulites is non-uniform, they accumulate in some places forming supracrystalline associations that con-

sist of rounded objects. Admixtures, oligomers and other amorphous material in small quantity remain in spherulites content, but they are mainly displaced out of the borders of ordered zones, forming, in turn, amorphous associations, sometimes of sufficiently large size.

Figure 10, *a* shows that structure of the surface is non-uniform and changes from one area to another. Crystalline areas of irregular shape are obviously a demonstration of the associations of spherulites which united in one large formation. Not by chance, one of the most realistic theories of polymer structure, namely paracrystalline Hosemann [14] model describes its structure as a moving matrix with crystalline folded formations floating in an amorphous phase located between them. Separate convex rounded objects can be the remains of torn molecular domains — fibrils which were formed in the process of crack formation. Amorphous material occupies in some regions sufficiently large areas, therefore, in these zones the polymer material has decreased strength that can serve a direction for crack propagation. The photo of the largest scale (Figure 10, *b*) demonstrates the ordered formations of rounded shape, which after etching got a typical cone-like shape, inside which a finer layerwise structure similar to spherulite is observed. Chemical etching agent removed from these objects all amorphous loops of chains and through molecules remaining only crystalline phase of spherical shape.

It is necessary to note that the examined welded joints are mechanically full-strength according to tensile test under [12]. In such a way, regardless the fact that the microstructure of polymer material is non-uniform enough, a failure zone is formed along the fusion surface, but on macrolevel it is possible to assume that the material of welded joint is statistically isotropic. A semi-crystalline transition zone is formed in a zone of fusion of butt weld. It obviously consists of fragments of macromolecules and clusters that by a thin layer join a monolithic spherulite structure of base material.



## CONCLUSIONS

1. There was carried out a crack resistance testing of base material and welded joints of polyethylene pipes according to the requirements of International Standard ISO 220088-4 using the Rehbinder effect.

2. A nature of crack propagation in a base material indicates some structural organization of polyethylene in pipe longitudinal direction. On external pipe surface the direction of crack matches exactly with its longitudinal axis. In the welded joints of polyethylene pipes the cracks are formed along the fusion surface or in vicinity to it.

3. A fracture nature shows that the solid spherulite structure is absent along a fusion line, and failed “diluted” structure of polymer material is formed in this zone by mutual penetration of fragments of molecules. The magnified images of the fracture surface of welded joints demonstrate the disordered randomly located areas as well as fragments of regular spherulite structure that indicate “wandering” of a crack in the region of failed fusion zone.

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## CONFLICT OF INTEREST

The Authors declare no conflict of interest

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## SUGGESTED CITATION

G.M. Korab, M.V. Iurzhenko, M.O. Kovalchuk, L.S. Parshutina, O.M. Chukashkin (2023) Investigations of the structure of welded joints of polymers using the Rehbinder effect. *The Paton Welding J.*, **2**, 18–23.

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Received: 02.01.2023

Accepted: 30.03.2023