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Aging of drinking water transmission pipes during long-term operation as a potential source of nano- and microplastics

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ABSTRACT

Microplastics (MPs) and nanoplastics (NPs) released into drinking water from transmission pipes can pose a potential health risk to consumers. This paper presents the results of a comprehensive study of PE and PVC pipes after long-term operation in drinking water distribution networks, which confirmed that degradable polymers can be a significant source of MPs. Both plastics age relatively quickly, and the degree of damage to the pipe surface depends on the time and operating conditions. During aging, polymer chains deteriorate, leading to a weakening of the structure and increased amorphousness of the plastics. As a result, the surfaces of PE and PVC crack and peel, resulting in the formation of particles with sizes corresponding to NP and MP with high potential for release into water. The magnitude of the phenomenon increases as the diameter of the pipes decreases, indicating that the most vulnerable customers are those at the ends of the network to which drinking water is supplied through small-diameter pipes. Aging PE and PVC pipes should be considered a real and very important source of MPs and NPs in drinking water, and water quality in this aspect should be monitored by manufacturers.

1. Introduction

The quality requirements applicable to drinking water impose on water producers the need for advanced treatment and disinfection processes, as well as the obligation to monitor the condition of materials used in the construction of water transmission networks, due to the possibility of secondary contamination of transported water (Moel et al., 2006). While unit treatment processes and the quality parameters of water leaving treatment plants are easy to control, controlling changes in the quality of water flowing through complex drinking water supply networks (DWSN) is very difficult. DWSNs, growing over the years, have undergone significant changes in material structure over the past few decades. Commonly used, but subject to corrosion processes, metal alloy pipes (e.g., steel, cast iron) are gradually being replaced by increasingly improved plastics, such as polyvinyl chloride (PVC), polyethylene (PE), polypropylene (PP) or glass fiber reinforced polyester (GRP) resins. Since the 1950s, the expansion of systems based initially on PVC and later on PVC and PE began. In highly urbanized areas, PE-based water mains dominate, while PVC is more commonly used in less urbanized areas (Kowalski et al., 2013; Nawrocki and Świetlik, 2011; Świetlik et al., 2012). Nowadays, the most commonly used materials for pipelines are high-density polyethylene (HDPE) and polyvinyl chloride (PVC).

In DWSNs polymer-based water pipes are installed in both buried distribution systems and indoor networks in buildings (Brocca et al., 2002). Their widespread use and popularity is due to numerous advantages. Compared to metal alloys and concrete, plastic pipes have greater flexibility and resistance to mechanical damage, are easier to install and, most importantly, are not subject to corrosion (Frank et al., 2009). Moreover, the composition of HDPE is subject to continuous modification in order to obtain the best possible mechanical properties (e.g., resistance to cracking, restriction of migration of ions and low-molecular-weight organic compounds from the external environment, etc.) (Eng et al., 2011; Hassinen et al., 2004). In turn, PVC is characterized by high mechanical strength and thermoplasticity, achieved by the addition of plasticizers, lubricants, fillers, impact modifiers, pigments, antioxidants and thermal stabilizers (Hassinen et al., 2004; Andersson and Ifwarson, 2001). Among the most popular antioxidants used in polymer processing are bisphenol A, bound phenols (BHT, BHA), Irganoxes of various structures and quinones (Andersson and Ifwarson, 2001). In turn, thermal and UV degradation of polymers is prevented by the addition of stabilizers such as lead and tin. It has been anticipated that the use of PVC and PE in networks transporting drinking water to consumers would reduce pipeline failure rates, as well as minimize the risk of reduced biological and chemical stability of transported water

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caused by corrosion of metal alloy pipes. However, the results of studies published to date (unfortunately mostly model-based), indicate that during long-term operation these materials may also degrade under the influence of both water admixtures, such as disinfectants used (Eerkes et al., 2015), and other factors, e.g. water flow velocity (Su et al., 2016; Koelmans et al., 2015), hydraulic conditions (Su et al., 2016) and temperature (Kirstein et al., 2012; Anderson et al., 2017). As a result of degradation, networks made of PE and PVC are also susceptible to the development of biofilms nourished by compounds released into the plastic water and extracted by microbial/enzymatic decomposition of the overgrown polymers (Świetlik and Magnucka, 2024).

Degradative aging of plastics is understood as a series of physical, chemical or mechanical changes occurring in the structure of a given material, which contribute to changes in the material both during processing and use. These can lead to irreversible changes in the properties and structure of the plastic, and thus negatively affect its functionality, reliability and durability. These changes are often irreversible. Among the negative environmental factors affecting the deterioration of the properties of materials are light and ultraviolet radiation, heat, oxygen, humidity, precipitation, gases, microorganisms, bacteria, fungi or mechanical interactions (Gnatowski et al., 2014). In the available literature, the influence of water disinfectants is pointed to as the main reason for the deterioration of the mechanical parameters of PE pipes working in the distribution network. Numerous studies (Eng et al., 2011; Hassinen et al., 2004; Andersson and Ifwarson, 2001; Świetlik and Magnucka, 2024; Devilliers et al., 2018; Whelton and Dietrich, 2009; Whelton et al., 2011) have shown that despite good corrosion resistance, thermoplastic pipes made of polyethylene are sensitive to the effects of chlorine-based disinfectants, i.e. free chlorine, chloramines, and sodium hypochlorite or chlorine dioxide. Long-term exposure to these substances, causes deterioration of the chemical and mechanical stability of PE pipes, resulting in accelerated aging of the material in the distribution network. As a result, the material becomes less resistant to mechanical damage, which manifests itself in the formation of various types of cracks, peeling, etc., making it a source of micro/nanoplastic release into the transported water. The processes of mechanical aging of distribution materials are observed to a more significant degree for polyethylene pipes, since PVC, due to its properties, is considered a material much more resistant to chemical degradation and mechanical damage.

Studies conducted by Zhang et al. (Zhang et al., 2014, 2018; Zhang and Liu, 2014), Song et al. (2021), and Whelton et al. (2011) have shown that degradation of the polymer structure increases its permeability. This is associated with the possibility of diffusion through the pipe material, and compounds from the external environment, which can deteriorate the quality of transported water. It has been observed that undesirable compounds, e.g., petroleum derivatives, organic solvents and pesticides, but also ions of inorganic nature can easily migrate through both PE and PVC (Andersson and Ifwarson, 2001; Su et al., 2016; Di and Wang, 2018). Diffusion processes occur with varying intensity, which depends primarily on the structure and composition of the material building the distribution network. The pipe material itself can also be a source of compounds that contaminate drinking water (Świetlik and Magnucka, 2024; Sixsmith and Hanselka, 1997; Al- and Malack, 2001; Davis et al., 2007; Dietrich and Heim, 2007; Makris et al., 2020; Tang et al., 2013). Both compounds added to the polymer at the manufacturing stage and those released by material degradation can diffuse into the water. In the case of pipes made of polyethylene, several compounds released from this material into the water have been identified, among which the most numerous groups were by-products of oxidative degradation of antioxidant coatings, belonging to phenols and quinones, including 4-ethylphenol or 2,6-di-t-butyl-p-benzoquinone (Brocca et al., 2002; Świetlik and Magnucka, 2024; Flemming et al., 2002). Moreover, compounds released due to damage to the polymer building chain have been recognized, which are organic compounds such as aldehydes, ketones, esters, and aromatic hydrocarbons (Świetlik and Magnucka, 2024; Plastic pipes, 2023; Davis et al., 2007; Dietrich

and Heim, 2007; Makris et al., 2020). Their presence in drinking water can cause deterioration of its organoleptic properties, especially odor (Zhang et al., 2014, 2018; Zhang and Liu, 2014). In contrast, pipes made of PVC are a potential source of tin and lead, whose presence in drinking water is dangerous, especially for young children (Sixsmith and Hanselka, 1997). In addition, in water in contact with PVC, the presence of aldehydes, i.e., hexanal, and octanal have also been noted (Biedroń et al., 2017; Skjevraak et al., 2003, 2005). Studies by Sadiki et al. (1997) confirmed the presence of organotin compounds in water in contact with PVC, and Ye et al. (2020) showed the problem of continuous release of dibutyl phthalate, used as a plasticizer in the production of these pipes, into water.

As a consequence of the migration of various organic compounds, including biodegradable aldehydes and ketones, into tap water, microorganisms develop on the walls of PVC and PE pipes (Świetlik et al., 2012; Simões and Simões, 2013). When nutrients are limited, biofilm-forming bacteria release into the water organic compounds, obtained directly from the pipe through enzymatic decomposition of the polymer, which intensifies and accelerates the plastic aging process (Świetlik and Magnucka, 2024). The vital activity of biofilm-forming bacteria also leads to secondary water contamination by the products of their metabolism (Zhang et al., 2018; Ye et al., 2020; Denberg et al., 2007; Kalweit et al., 2019; Lutzhöft et al., 2012; Ryssel et al., 2015). Modeling studies have shown that networks made of PE and PVC are prone to biofilm formation, and the rate of biofilm growth depends on several factors, such as pipe material, nutrient availability, disinfectant concentration, hydraulic conditions and temperature. However, comparative studies have shown that despite the susceptibility to biofilm growth, significantly less biological material is deposited on PE and PVC pipes than on conventional gray iron pipes (Whelton et al., 2011; Inkinen et al., 2013; Almond et al., 2020).

Aging of thermoplastic materials primarily causes damage to their surface layer. Consequently, as a result of the coexistence of chemical and biological degradation processes on the internal surfaces of water pipes, numerous micro-damages, cracks and exfoliation can be expected, which can generate and release micro- and nanoplastic particles into the transmitted drinking water (Świetlik and Magnucka, 2024). These particles, less than 5 mm in diameter, are classified as microplastics (Eerkes et al., 2015; Su et al., 2016; Koelmans et al., 2015; Anderson et al., 2017; Di and Wang, 2018). In drinking water, they are most often found in the form of particles with sizes in the tens and hundreds of micrometers (Eerkes et al., 2015; Sixsmith and Hanselka, 1997; Andrady and Neal, 2009; Gambino et al., 2022; Mintenig et al., 2019; Novotna et al., 2019; Pivokonsky et al., 2018; Wessel et al., 2016). Recently, however, there has been growing interest in the finest fraction of microplastics - the so-called nanoplastics, which model studies have shown can penetrate cell membranes (Lehner et al., 2019; Leslie et al., 2022). According to recent studies in mice, nanoplastic particles in the order of 0.1 and 2 µm can reduce the viability of their cells, migrate with body fluids throughout the body, accumulate in tissues and in the brain and even modify behavior (Gaspar et al., 2023). Their important source in drinking water may be aging water pipes. In addition, micro- and nanoplastic particles have the ability to bind on their surface and transport numerous contaminants (Zhao et al., 2023), including compounds that, when released, can negatively affect living organisms and humans. Studies comparing the amount of microplastics in water taken directly from the treatment plant with the number of particles present in tap water indicate that the amount of microplastic particles increases as a result of water transport through pipes made of plastic (Eerkes et al., 2015; Świetlik and Magnucka, 2024; Mintenig et al., 2019; Pivokonsky et al., 2018). On this basis, two types of microplastic were distinguished according to their origin. The first group consists of particles not removed by the treatment process occurring as an admixture of raw water, so-called primary microplastics, which can settle and accumulate in the sediment present on the inner surfaces of the transmission pipes during the transport of water in the water supply network (Chu et al.,

Table 1

Technical data of water supply pipes taken from the distribution network in 2019–2022.

NAME	MATERIAL	SERVICE LIFE [YEARS]	DIAMETER [mm]
WYC_1_22	PE	13	180
WYC_2_22	PE	4	180
WYC_3_22	PE	16	125
WYC_4_22	PVC	31	160
WYC_1_21	PVC	45	100
WYC_2_21	PVC	41	80
WYC_3_21	PE	14	225
WYC_4_21	PVC	36	200
WYC_1_20	PVC	26	90
WYC_2_20	PVC	29	110
WYC_3_20	PE	20	100
WYC_4_20	PVC	30	160
WYC_5_20	PVC	27	160
WYC_6_20	PE100RC	6	315
WYC_7_20	PE	4	180
WYC_8_20	PVC	29	160
WYC_1_19	PE	18	90
WYC_2_19	PE	15	125
WYC_3_19	PVC	28	90
WYC_4_19	PVC	16	110

2022). The second type of microplastics are particles resulting from the degradation of the pipe material, so-called secondary microplastics, associated with the formation of microdamage and exfoliation on the polymer surface, which can be detached and transported with the water to consumers' taps under the flow and pressure conditions prevailing in the distribution network (Świetlik and Magnucka, 2024; Mintenig et al., 2019; Pivokonsky et al., 2018; Boucher and Friot, 2017; Shen et al., 2021; Tong et al., 2020). In addition, the flow rate and hydraulic pressure in water supply networks can also contribute to the detachment of primary particles from sediments that have accumulated over the years on the inner surfaces of metal alloy pipes (Chu et al., 2022). These contaminants can exhibit high toxicity and bioaccumulation both in small living organisms and in humans, i.e. consumers of drinking water (Al- and Malack, 2001).

Despite a relatively large number of studies signaling the occurrence of both chemical and microbiological degradation processes in plastic drinking water transmission pipes, the actual state of their internal surface after long-term operation in operating distribution networks has not yet been comprehensively analyzed. Available studies are based either on model systems or on the analysis of individual fragments of operating transmission networks. The purpose of this study was to comprehensively characterize numerous sections of actual PE and PVC water pipes taken from an operating DWSN characterized by widely varying service life (from 4 to 45 years). The evaluation of the degree of degradation of the analyzed plastics and the characterization of their surfaces made it possible to determine the course of the aging process of pipes and to identify operational factors affecting the speed and intensity of the phenomena occurring. In addition, the results showed how the process of generating micro- and nanoplastic particles on the surface of PE and PVC takes place, and proved that deteriorating plastic pipes are an important source of secondary water contamination with micro- and nanoplastics, which has a negative impact on the quality and safety of drinking water.

2. Materials and methods

2.1. Characteristics of the tested materials

The study was conducted in 2019–2022 on sections of pipes of various diameters, made of PVC and PE, taken from a metropolitan distribution network supplied with water disinfected with chlorine compounds (chlorine gas, sodium hypochlorite). In the course of the

study, 21 pipe fragments were taken: 10 made of PE and 11 made of PVC. The fragments made of PVC came from pipes with long service lives (16–45 years), while the PE fragments worked in the network for a much shorter period of time (4–20 years). The differences in the age of pipes made of each material were related to their availability - in the 1980s, only water pipes made of PVC were available in Poland, while the widespread use of PE has been recorded only since the late 1990s. Technical data of the analyzed real sections are shown in Table 1.

2.2. Pipe sampling

PE and PVC pipe cuttings were taken during the repair of water supply faults at various locations in the distribution network. For the purpose of the study, sections of pipe much longer than the damaged area (typically 2–4 m in length) were unearthed during repairs. After thorough cleaning of the soil residue, the damaged sections were removed together with a large surplus, which was an undamaged section of pipe. The surplus to be tested was always above the damage, ensuring that the sections were taken free of environmental contamination. Undamaged surplus pipes of approximately 1 m in length were then cut off, protected with aluminum foil and transferred to the laboratory.

2.3. Preparation of the fragments for further analysis

In order to carry out a complete characterization of the surfaces of the examined pipes, the taken sections were cut with a special knife into fragments of 2 × 6 cm. The cutting procedure was carried out eliminating completely the possibility of scratching/damaging the analyzed surfaces. First, samples of materials covered with natural wet deposits present on the surface at the time of collection were prepared. In this case, 2 × 6 cm sections with intact sediment/debris were secured immediately after cutting in Petri dishes and subjected to microscopic imaging (SEM). The remaining sections intended for studies showing plastic surface changes and the degree of polymer degradation required thorough cleaning from mineral and biological deposits. For this purpose, a several-step procedure was applied using high-purity HCl (POCH Gliwice, POLAND), H₂O₂ (POCH Gliwice, POLAND) and MeOH (Aldrich, POLAND). In the first step, inorganic deposits were dissolved by immersing the fragments for 15–30 min in HCl (1:1 v/v). The samples were then washed with high-purity water (Millipore, USA). In the next step, in order to mineralize the biofilms, the plastic fragments were immersed in beakers containing concentrated hydrogen peroxide for 48 h. The cleaned plastics were again immersed in HCl (1:1 v/v) for about 15 min to dissolve inorganic residues trapped in and under the biofilm layer. Rinsed several times with deionized water, the samples were left to dry. In the final step, the cleaned and dried sections were immersed in beakers of methanol to degrease the surface. After the methanol evaporated, the samples, cleaned of any sediment, were secured in Petri dishes. An analogous cleaning process was carried out on fragments of brand-new materials, which allowed us to conclude that this procedure had no significant effect on the results obtained in the presented study.

2.4. Analytical methodologies

2.4.1. Scanning electron microscopy (SEM)

Microscopic images of the surfaces of the materials studied were taken with a Quanta FEG 250 scanning electron microscope (FEI) under low vacuum at 70 Pa. Imaging of the samples was performed at a beam accelerating voltage of 10 and 30 kV. All analyses were performed without sputtering the samples.

2.4.2. Energy dispersive spectrometry (EDS)

Chemical characterization and elemental analysis of analyzed materials was performed using an Octane SDD EDS detector (EDAX). Acquisition of EDS spectra was performed at a beam accelerating voltage of 30 kV. All analyses were performed without sputtering the samples.

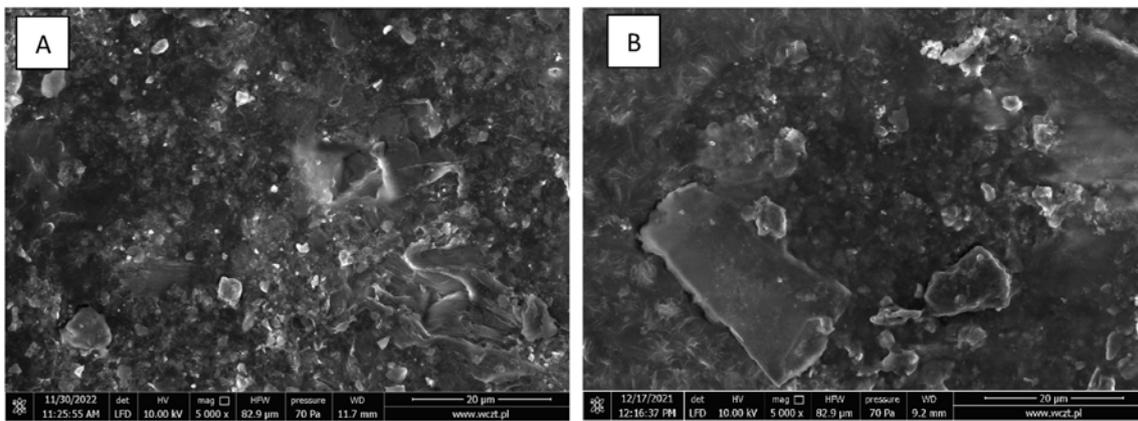


Fig. 1. Flaking and torn PE particles visible on the inner surface of the analyzed materials; A) 4 years of operation, B) 14 years of operation.

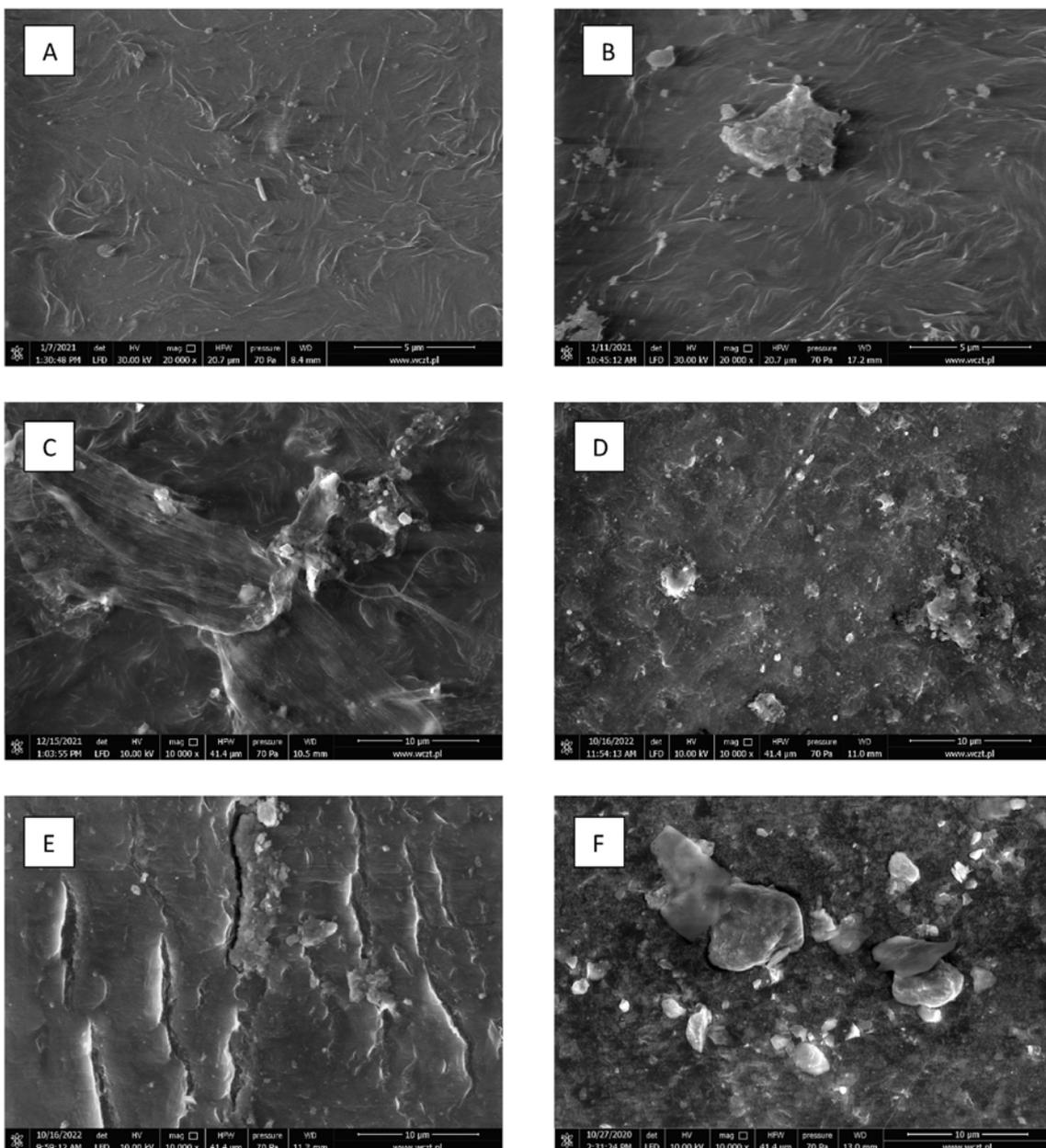


Fig. 2. Changes in the appearance of the inner wall surface of PE pipes during operation after A) 4 years, B) 6 years, C) 14 years, D) 15 years, E) 18 years and F) 20 years of operation.

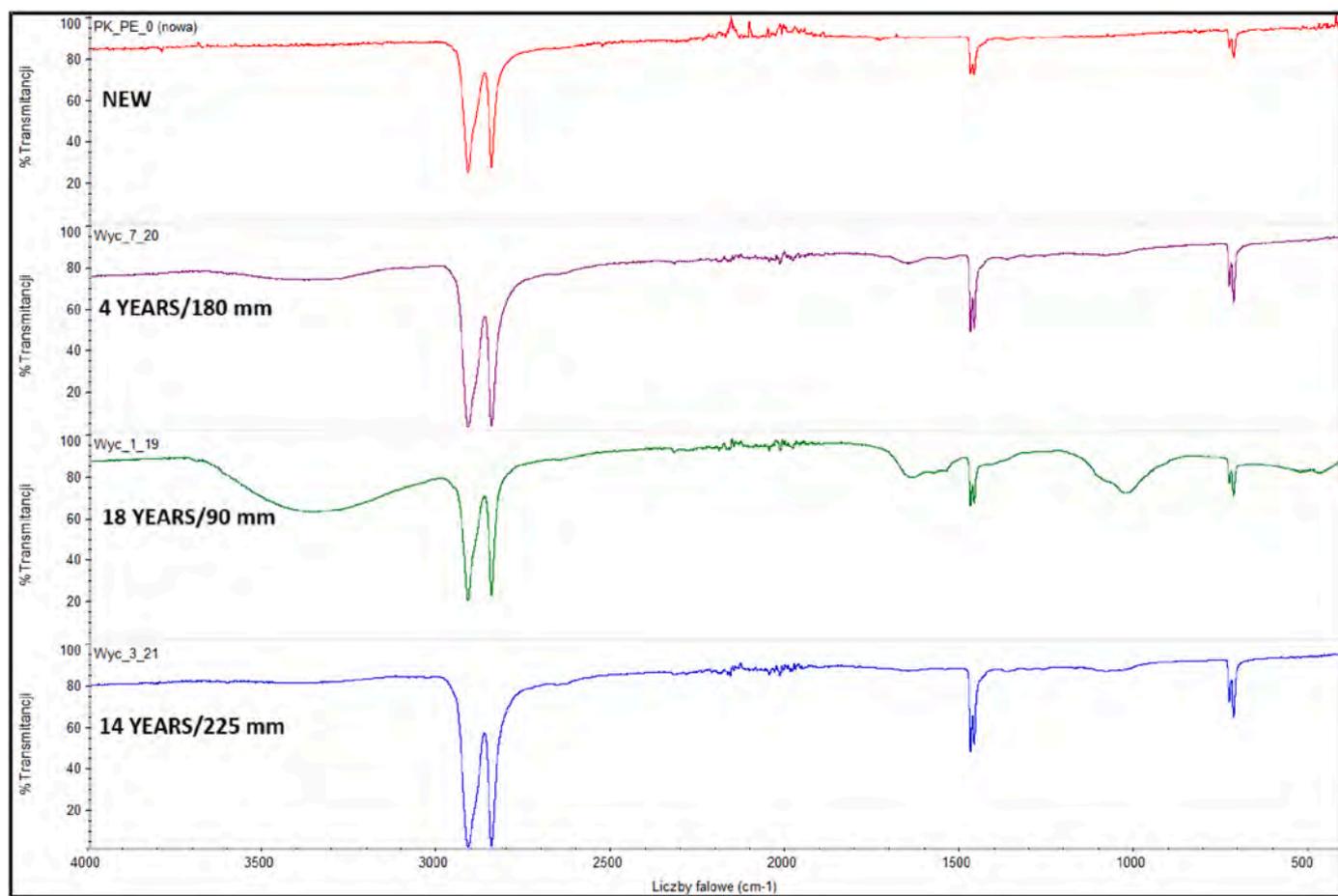


Fig. 3. ATR-FTIR spectra recorded for PE pipes.

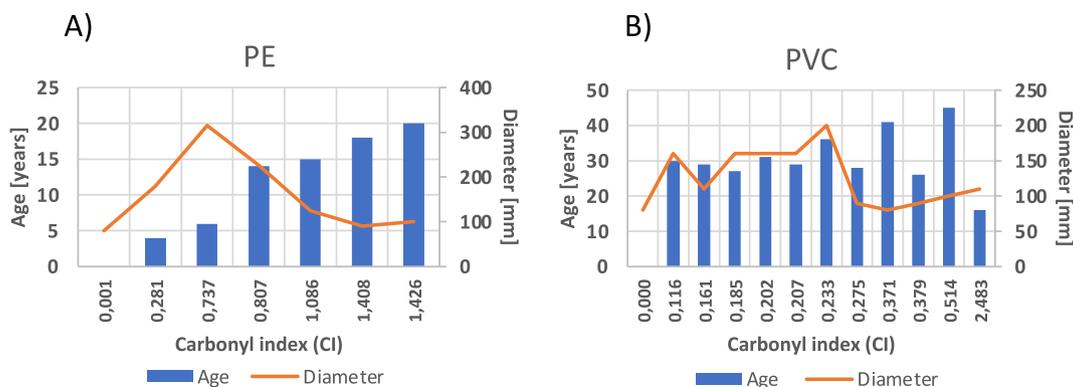


Fig. 4. Carbonyl index (CI) values as a function of pipe diameter and service life: A) PE and B) PVC.

2.4.3. Attenuated total reflectance fourier transform infrared spectroscopy (ATR-FTIR)

The study of structural changes resulting from aging of PE and PVC water pipes was carried out using Nicolet iS50 Fourier transform spectrophotometer (Thermo Fisher Scientific, Waltham, MA, USA) equipped with a diamond window ATR (Attenuated Total Reflectance) unit with a resolution of 4 cm⁻¹. For each measurement, a total of 16 sample scans were taken in a 4000-400 cm⁻¹ range. Eight scans of background were taken prior to each sample measurements session.

2.4.4. Determination of carbonyl index (CI)

CI was determined using ATR-FTIR with the specific area under the

band (SAUB) method, based on the ratio of the integrated absorbance of the carbonyl peak band (C=O) from 1850 to 1650 cm⁻¹ to the absorbance of the methylene peak (CH₂) from 1500 to 1420 cm⁻¹ (Almond et al., 2020).

2.4.5. Contact angle (CA)

Measurements of the contact angle (CA) were performed at room temperature (23°C) using sessile drop method on a Krüss DSA100 goniometer in an automatic mode. For each sample, three individual measurements with a 5 µl water drop were taken. The results were averaged to reduce the impact of surface non-uniformity. Krüss DSA 4 software was used for image analysis.

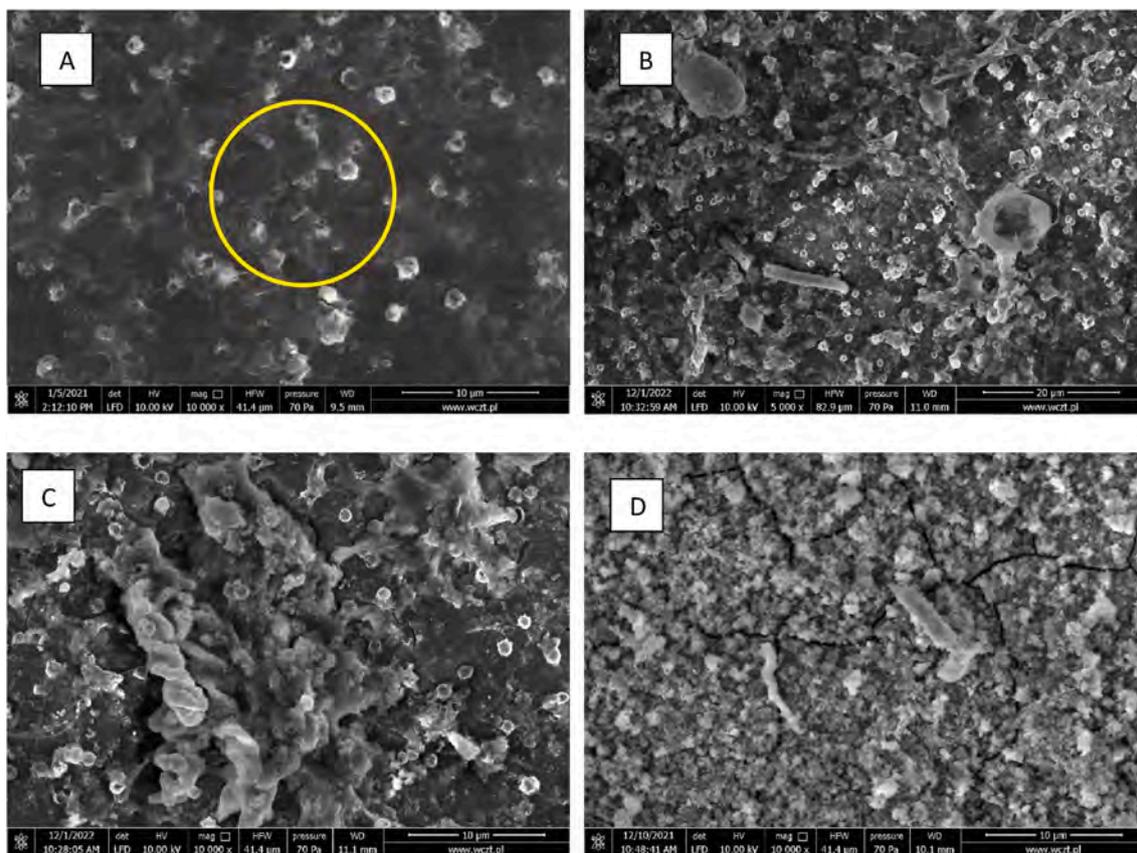


Fig. 5. Mineral and microbial deposits visible on the surface of PE pipes (untreated fragments); A) 4 years, B) 13 years, C) 13 years (iron oxidizing bacteria (Garrison et al., 2019)), D) 14 years of operation.

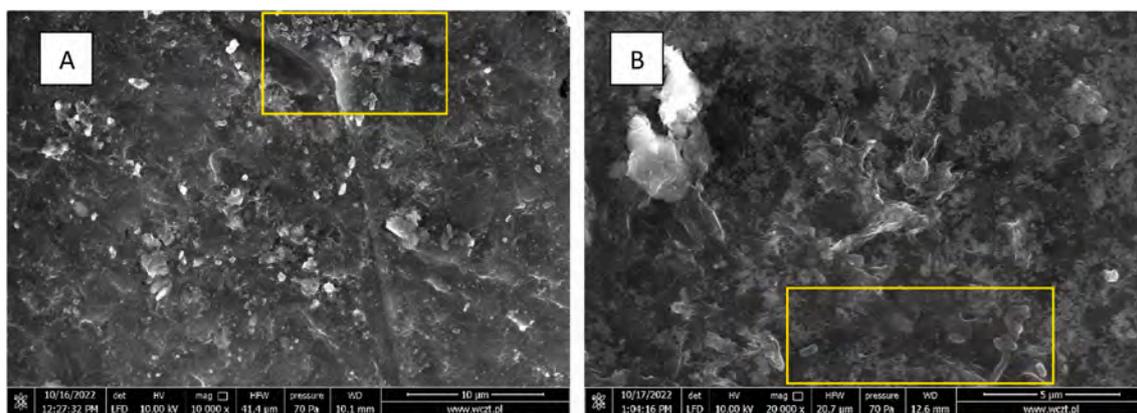


Fig. 6. Bacterial cell residues present on PE surface after cleaning procedure, A) 15 years and B) 20 years of operation.

3. Result and discussion

3.1. Ageing and degradation of internal surfaces of PE and PVC pipes associated with their long-term operation in the drinking water distribution system (DWDS)

Studies on the purity and safety of drinking water transmitted through extensive water supply networks indicate that even in treated water, characterized in its original purity, microplastics are determined after the distribution stage (Świetlik and Magnucka, 2024). This indicates that secondary contamination of water with MPs occurs during its contact with pipes made of plastics. In order to determine the course and rate of degradation processes occurring in plastic pipes, a detailed

characterization was carried out in the presented study cycle of HDPE and PVC water pipe fragments taken from real distribution systems after a lifetime of between 4 and 45 years. The sections were taken consecutively between 2019 and 2022. The internal surfaces of the analyzed pipes cleaned of mineral and organic deposits were characterized using scanning electron microscopy (SEM). Changes in the structure of the plastics were analyzed using ATR-FTIR and carbonyl index (CI), while contact angle (CA) measurements were used to assess the change in surface roughness during aging. Untreated, sediment-covered sections were also evaluated microscopically to assess the extent to which the inner walls of the analyzed pipes were covered with mineral and/or bacterial deposits.

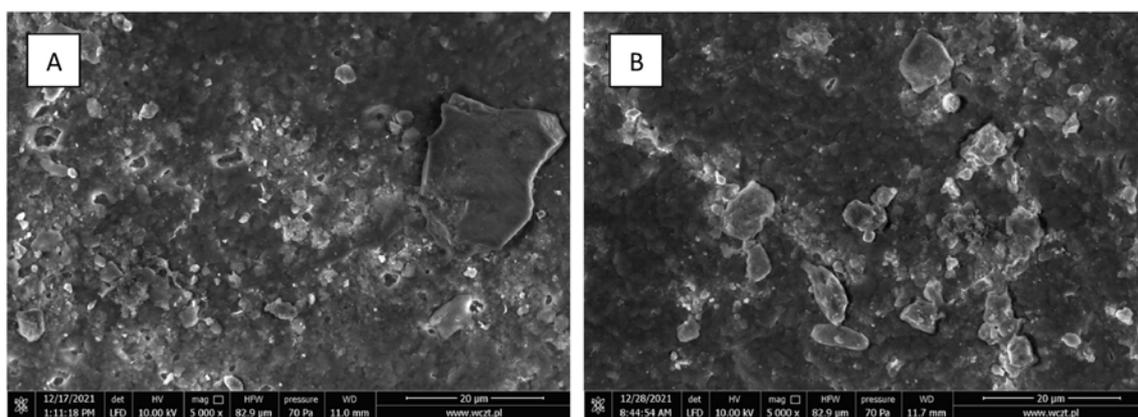


Fig. 7. Flaking and torn PVC particles visible on the inner surface of the analyzed materials; A) 36 years and B) 45 years of operation.

3.1.1. PE pipes

Pipes made of PE are increasingly used in water distribution systems due to their flexibility, durability, corrosion resistance, thermal stability, and high tensile strength. However, the material gradually degrades during operation, resulting in numerous network failures caused by burst pipes. Water producers also note numerous consumer complaints about the quality of water supplied through PE pipes, related to unacceptable odor and turbidity of water. As part of the study, 10 sections of pipes made of PE were obtained, which had been in operation in the working distribution system of a large city for 4–20 years. At the time of collection, small amounts of mineral-iron deposits were present on the materials, and their thickness increased with the age of the pipe. This is indicative of increasing surface porosity allowing sediment applied from other areas of the network made of metal alloys to be retained. Increasing damage to the pipe surface resulting in loss of uniformity and smoothness, as well as a decrease in the hydrophobicity of the tested materials, was confirmed by measurements of the wetting angle (CA), which in the case of PE pipes decreased successively with pipe age from 98.3° for new material to 69.5° for pipe after 20 years of service. The degree of material degradation was greatest for pipes with small diameters (<100 mm).

Detailed microscopic analysis of the surface of the extracted materials confirmed the occurrence of aging processes, manifested by the formation of numerous exfoliations, cracks and cavities on the PE surface. Microscopic images recorded for all sections, regardless of their age and diameter, showed numerous microfragments, which were associated with the polymer surface to varying degrees. These particles were characterized by different shapes - from irregular plates with sharp edges to threads/fibers. The number, distribution and shapes of microfragments are shown in the photos (Figs. 1 and 2). The SEM images also show microdamage to the PE surface, which may be the source of particles attached to the surface of pipes transporting tap water (Fig. 2C–E). These fragments, once fully detached from the surface, will be classified as nano- and microplastics. The defects, depressions and irregularities observed in the images may also provide excellent sites for the growth of microorganisms and, consequently, biodegradation of the polymer.

In-depth analysis of the microscopic images recorded during the study also made it possible to assess the rate and course of degradation processes occurring on the inner surfaces of PE pipes. The wrinkles visible in the images (Fig. 2A and B) are a consequence of the progressive degradation of antioxidant coatings, a layer of which is applied by manufacturers to the internal surfaces of pipes to increase the resistance of the plastic to oxidizing agents. These coatings, as a result of the action of oxidants constantly present in water (dissolved oxygen, disinfectant) during the first few years of operation degrade resulting in wrinkling of the surface layer on the walls, which then flakes off exposing the deeper layers of the polymer (Fig. 2C). In subsequent years of operation, such changes on the PE surface are less pronounced than in pipes with a

shorter service life, but are still visible. Further operation of pipes lacking the antioxidant coating leads to their increasing cracking and peeling, resulting in the release of plastic particles into the water (Fig. 2D–F).

The observed changes occurring on the surface of the pipes during actual operation are in agreement with the results of model tests published to date. In the available literature, the influence of disinfectants is indicated as the main reason for the deterioration of the mechanical parameters of pipes operating in the distribution network. The most commonly used disinfectants for drinking water are chlorine gas, chloramines, sodium hypochlorite and chlorine dioxide. They all work by producing so-called free chlorine (HOCl and OCl⁻). Under the influence of prolonged exposure to chlorinated water, plastic pipes undergo severe degradation stresses, and their physical properties change (Eng et al., 2011). In model studies, it has been shown that contact of PE with chlorine-containing water causes a significant reduction in the level of antioxidants, and thus degradation of pipes (Hassinen et al., 2004; Andersson and Ifwarson, 2001). Based on the results of numerous model studies of accelerated PE aging under various conditions, including in the presence of chlorine in water, Whelton and Dietrich (2009) identified three main stages in the process. First, there is an attack of dissolved chlorine on the surface of the material resulting in an increase in surface oxidation and the presence of hydroxyl, vinyl and chlorinated products. The next stage involves the breaking of bonds between chains and the chains themselves, and further the formation of microcracks, which over time combine and spread into the pipe wall, causing it to rupture in extreme cases. Researchers have also observed visible striations and cracks as deep as 200 μm on the surface of materials treated with chlorine and chlorine dioxide (Devilleirs et al., 2018; Whelton and Dietrich, 2009; Kowalska et al., 2016). Consequently, PE aging processes can lead to the formation of solid fragments, classified, due to their very small size, as nanoplastics, less frequently as microplastics, as confirmed by the presented imaging results of the sections working in the actual distribution network.

Analysis of the ATR-FTIR spectra (Fig. 3) and determination of the carbonyl index (CI) provided more information on the aging process of the analyzed pipes. The results showed that during the operation of polyethylene water pipes, the degradation processes taking place extend into the deeper layers of the material. In addition to bands in the region of 2950–2850 cm⁻¹; 1460–1470 cm⁻¹ and 720–730 cm⁻¹, which are characteristic of PE and originate from stretching and bending vibrations of the -CH₂ groups building the polymer chain (Almond et al., 2020; Kerr et al., 2013; Abaroa et al., 2022; K  ppler et al., 2018), the spectra recorded the formation of new bands in the ranges of 3360–3370 cm⁻¹, 1700–1520 cm⁻¹ and 1180–900 cm⁻¹. Their intensity increased with the time of pipe operation and was particularly evident for pipes of small diameters. The presence of bands in the 3360–3370 cm⁻¹ range, attributed to the presence of the -OH group, is most likely

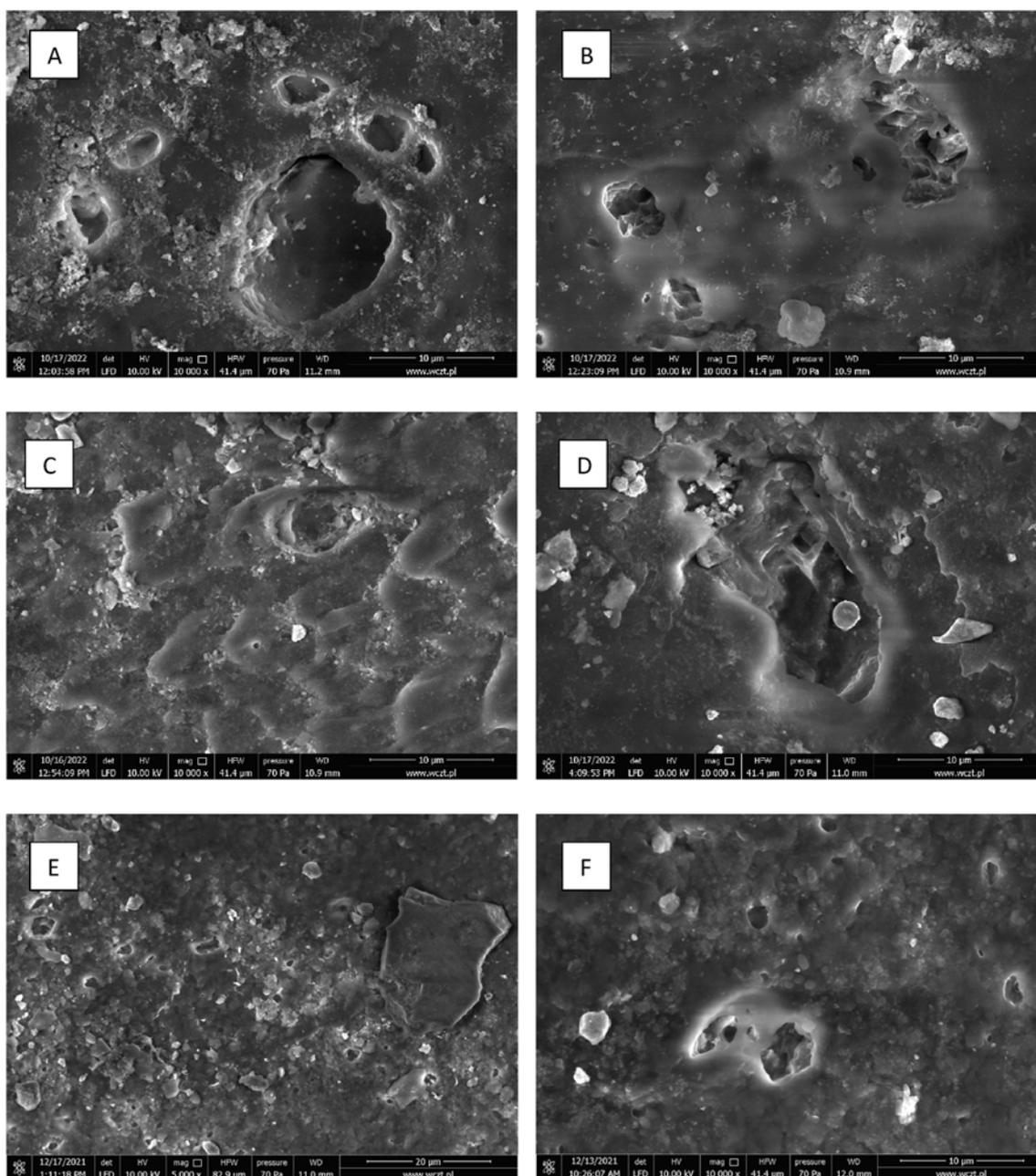


Fig. 8. Changes in the appearance of the inner wall surface of PVC pipes during operation; A) 16 years, B) 26 years, C) 28 years, D) 29 years, E) 36 years and F) 45 years of operation.

related to an increase in PE roughness, leaching of additives from deeper layers resulting in the formation of voids between polymer chains (formation of a sponge-like structure that facilitates the migration/diffusion of water and other contaminants, both from the transferred water and from the external environment (Whelton et al., 2011; Zhang et al., 2018; Song et al., 2021; Davis et al., 2007; Tang et al., 2013; Rysel et al., 2015)) and/or oxidative degradation of both the antioxidant coatings and the plastic itself. The co-occurrence of these, increasing over time, damages result in an increase in: 1) wettability, 2) water retention capacity of the polymer structure, and 3) hydrophilicity of the PE surface, associated with the formation of hydrophilic breakdown products of the protective coatings and the polymer itself (containing -COOH and -OH groups) in reaction with disinfectants present in distributed water (Fernández et al., 2021). On the other hand, the bands appearing in the range of $1700\text{--}1520\text{ cm}^{-1}$, with clearly marked maxima at 1650 cm^{-1} and 1550 cm^{-1} , are related to the appearance of an

increasing number of -C=O, -COOH and -CH=CH₂- groups, associated with oxidation and cleavage of the polymer chain occurring in the material, which results, among others, in the formation of biodegradable ketones and carboxylic acids (Fernández et al., 2021). The presence of a broad band with a maximum at $1010\text{--}1030\text{ cm}^{-1}$ attributed to the presence of an ether group (Kerr et al., 2013; Fernández et al., 2021) is also a direct indicator confirming the degradation of PE under the influence of factors causing oxidation and cleavage of bonds building the polymer backbone (such as residual disinfectant). This band is particularly well visible for pipes with small diameters and long service life (Fig. 3). Changes in intensity and width in the area of the band's characteristic of PE were also noted. In the case of pipes with a lower degree of degradation (shorter service life and/or larger diameters), their intensity increased, which is associated with an increase in disruptions of the crystallinity in polymer structure and thus an increase in amorphousness leading to increase in water binding capacity (Kerr et al.,

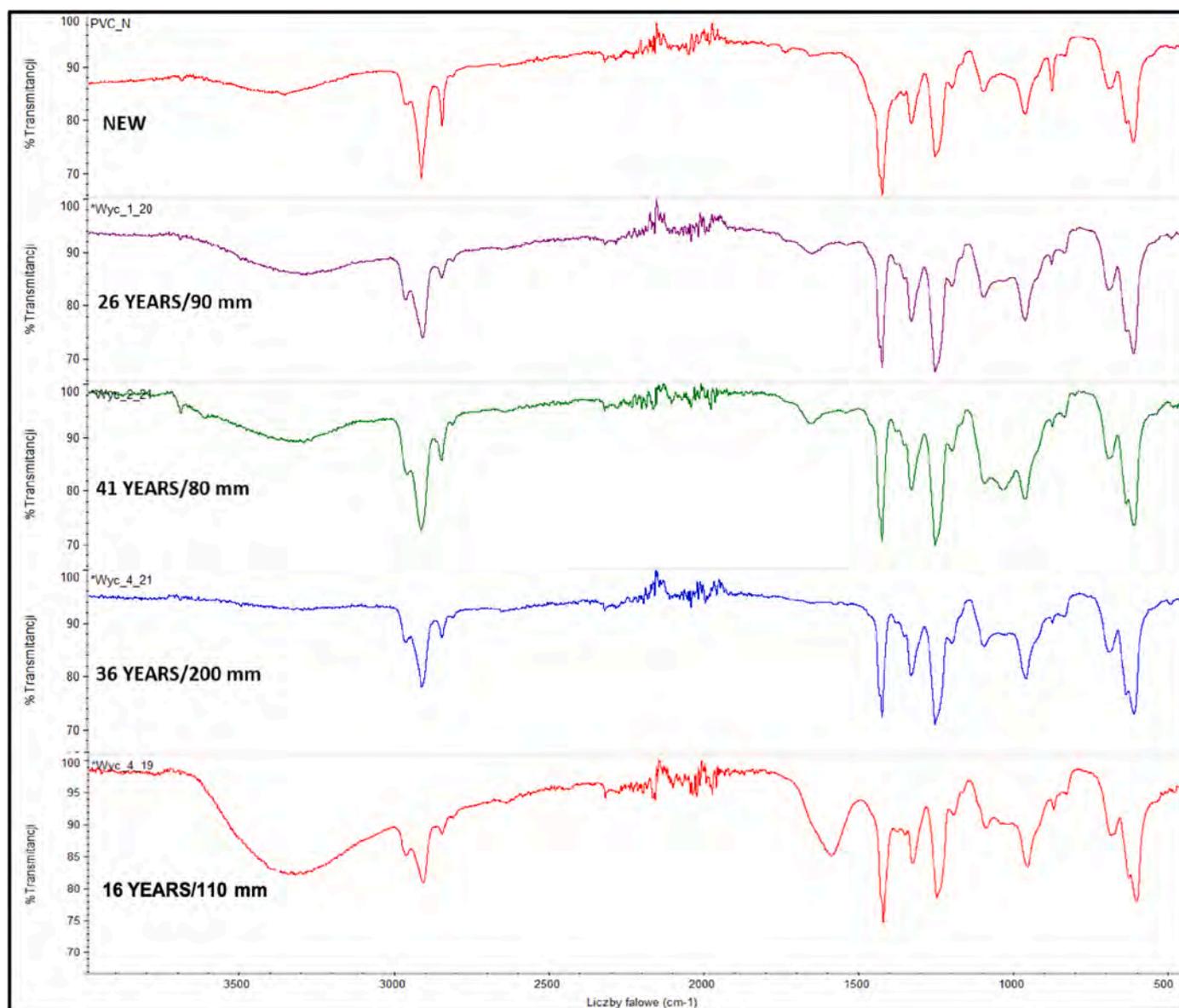


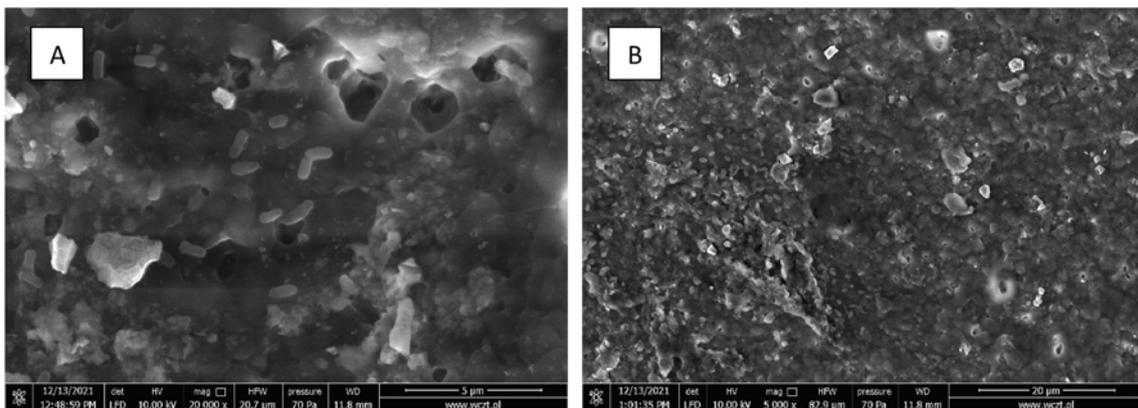
Fig. 9. ATR-FTIR spectra recorded for PVC pipes.

2013; K appler et al., 2018). This effect was less pronounced for the more heavily degraded materials, in which the formation of PE oxidation products manifested by the appearance of the previously described new bands occurred. The carbonyl index determined from the ATR-FTIR spectra confirmed the increasing degradation changes in the PE structure over time. The CI value increased with pipe age up to a value of 1, 426 and was higher for small diameters than for pipes with large cross-sections in the longest service life (Fig. 4A).

As a result of progressive degradation, soluble breakdown products of coatings and the plastic itself can also be released into the water, which in turn can provide a food source for microorganisms that populate distribution networks and/or are present in the transported water (Sim oes et al., 2006). A slippery, gelatinous deposit was present on the tested materials, indicating the presence of microorganisms. The thickness of the biological sediment layer correlated with the diameter of the pipes tested and was greater the smaller the pipe diameter. This is related to the flow velocity and water pressure, which are significantly higher in distribution pipes with large diameters, as well as to the presence of disinfectant. Pipes with a diameter of <120 mm are mainly used at the so-called ends of the network, where the daily water

distribution fluctuates periodically and there are periods of stagnation and/or very low flow, often accompanied by the disappearance of residual disinfectant. Studies by Flemming et al. (2002) and Gulati and Gosh (Gulati and Ghosh, 2017) have shown that about 95% of bacteria in drinking water distribution systems are attached to the inner surfaces of pipe walls, while only 5% are in the aqueous phase and are detected during analysis of water samples (Flemming et al., 2002; Gulati and Ghosh, 2017). Research also confirms that one of the key factors influencing the intensity of biofilm development in water distribution networks is the pipe material. The analysis of actual sections made it possible to observe not only the changes occurring on the inner walls of water pipes as a result of the operation, but also the increasing amount of biofilm developing inside the cavities created (Figs. 5 and 6). Metabolic processes carried out by microorganisms in the developed biofilms aggravate the material degradation process. Some of the microorganisms colonizing porous PE surfaces are so strongly associated with it that even after mineralization processes, bacterial cells are clearly visible on the surface of the plastic (Fig. 6).

Cleaned fragments



Uncleaned fragments

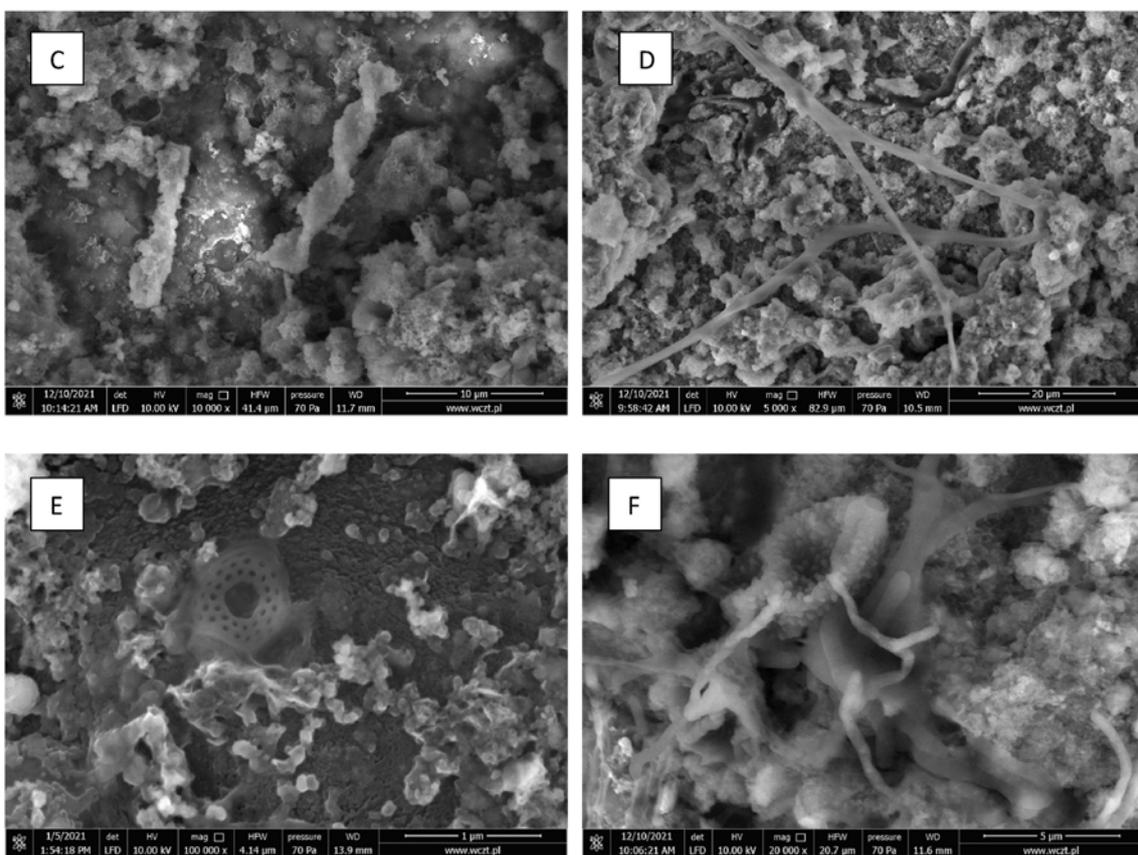


Fig. 10. Mineral and microbial deposits visible on the surface of PVC pipes; A) residual bacterial cells, B) mineral deposits, C) iron oxidizing bacteria (Garrison et al., 2019), D) and F) fungi (Alfonso et al., 2021) and E) representative of diatoms Thalassiosirophyceidae (Kociolek et al., 2015).

3.1.2. PVC pipes

Polyvinyl chloride is the material that has been used for drinking water transmission for the longest time. PVC is characterized by its high flexibility, abrasion resistance, chemical resistance, and low specific weight compared to cast iron pipes. Thanks to these properties, PVC is considered one of the more resistant materials to both chemical degradation and abrasion, which explains its wide use in water supply and wastewater networks. For the present study, 11 pipe sections made of PVC were obtained, and their service life ranged from 16 to 45 years. The materials taken were characterized by varying, but higher than PE, coverage of mineral-iron deposits carried by water from areas of the

network made of metal alloys, and the presence of a layer of slippery, gelatinous sediment, indicating the presence of microorganisms. In the case of PVC pipes, rusty mineral deposits covered basically the entire surface of the examined pipes regardless of diameter, while the thickness of biological deposits increased as the diameter of the pipe decreased.

The scanning microscopic images of the PVC surface recorded during the analyses indicate the presence of numerous depressions (pits/holes) and fine, torn plastic particles on the inner surface of the water pipe walls. As in the case of PE, the particles present on the PVC materials are characterized by a variety of shapes - from irregular plates with sharp edges to threads/fibers, but their number is higher than in the case of the

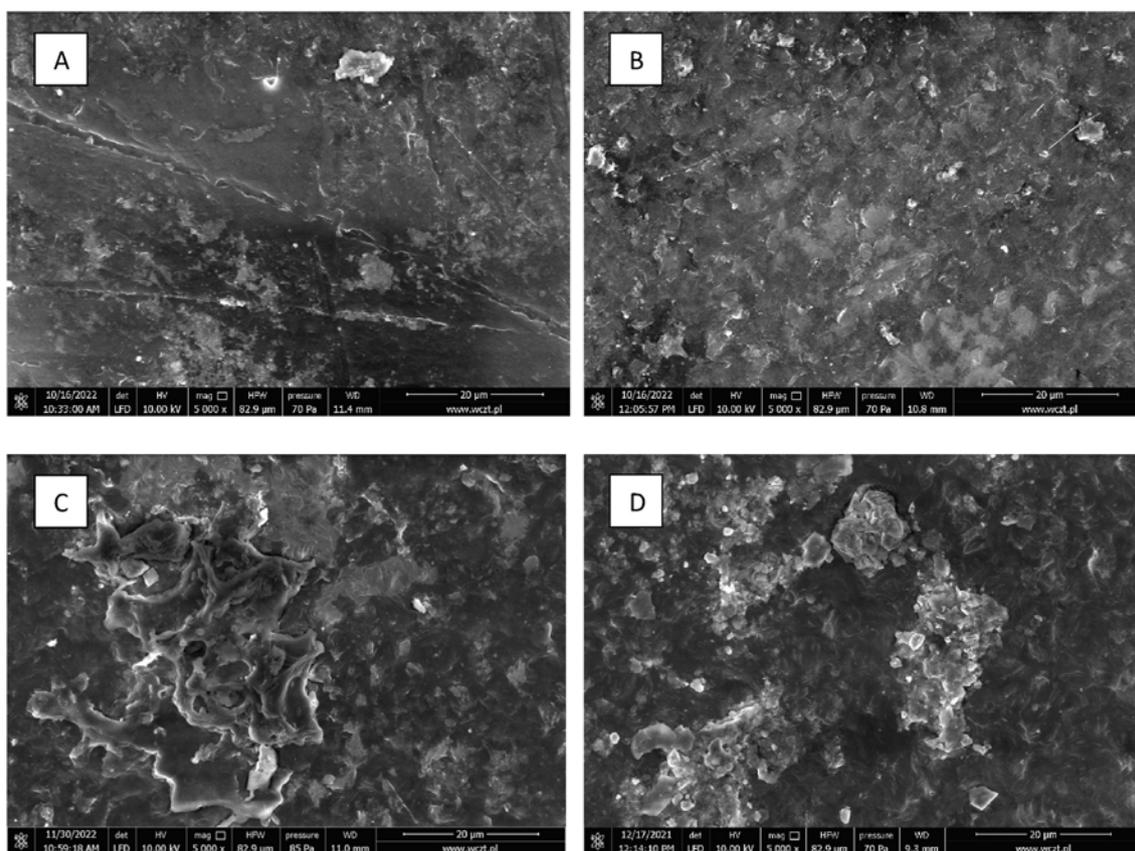


Fig. 11. Changes in the appearance of the inner surface of PE pipes depending on their diameter: A) ϕ 90 mm (13 years), B) ϕ 125 mm (15 years), C) ϕ 180 mm (13 years) and D) ϕ 225 mm (14 years).

PE samples examined (Fig. 7). This may be related to the longer service life of the plastic than PE, and thus the deeper degradation of the plastic. A significant difference from PE is the presence of numerous deep pits on the PVC surface. The characteristic pits were present on all analyzed sections regardless of their age and diameter. The number, distribution and shapes of the pits, as well as the accompanying numerous microfragments of eroded polymer, are very well illustrated by photos showing the surface of the PVC pipe (Fig. 8). The negative effect of water on the performance of this material has already been reported in the literature, indicating the formation of characteristic pits that promote sediment retention (Cerrato et al., 2006) and colonization by microorganisms. The results obtained in the presented series of studies confirmed the characteristic degradation path of PVC with the formation of depressions, inside which microorganisms can grow to form dense biofilms over time. In addition, analysis of microscopic images recorded for PVC sections after different periods of operation showed the extend damage to the plastic. Basically, the entire surface of long-used PVC is covered with pitting, which significantly increases the porosity of the polymer and the retention of deposits, clearly visible already during the macroscopic evaluation of the tested materials. Wetting angle measurements for these materials showed much more intense degradation of the PVC surface compared to PE. With operation, the CA of PVC decreased to values as high as $49,7^\circ$, with no clear correlation with the age of the pipe. A much clearer correlation was recorded between the wetting angle and the diameter of the tested pipes. In this case, pipes with larger diameters had a higher porosity, while those with $\phi < 90$ mm had a similar CA value ($80,1\text{--}87,7^\circ$) regardless of the length of service. This may be related to the quality of the plastic itself and/or to its lower resistance to the high pressures and higher flow rates prevailing in higher diameter pipes.

Analysis of the ATR-FTIR spectra of PVC pipe sections allowed the

identification of changes in the chemical structure of the material occurring during aging. All analyzed fragments had bands characteristic for PVC in the areas $2800\text{--}3000\text{ cm}^{-1}$ and $1200\text{--}1430\text{ cm}^{-1}$ originating from stretching and bending vibrations of -CH and =CH_2 groups, as well as bands 690 cm^{-1} and 610 cm^{-1} , which confirm the presence of -C-Cl group in the structure and serve to identify the polymer type (Kerr et al., 2013; K  ppler et al., 2018). Changes in the intensity of characteristic PVC bands and the appearance of new signals were observed, confirming the gradual degradation of the tested materials. In small-diameter pipes, the intensity of the bands at $2800\text{--}3000\text{ cm}^{-1}$ and $1200\text{--}1430\text{ cm}^{-1}$ increased with service time, while in larger-diameter pipes this effect was barely visible (Fig. 9). An increase in the intensity of the bands associated with -C-Cl stretching vibrations ($700\text{--}550\text{ cm}^{-1}$) was noted in all the materials analyzed, which may indicate degradation of the polymer structure. The spectra recorded for small-diameter tubes also showed the appearance of new signals in the range $1700\text{--}1500\text{ cm}^{-1}$ with maxima at 1650 and 1530 cm^{-1} , also visible on PE spectra, attributed to polymer chain scission and the formation of oxidative degradation products containing -C=O , -COOH and -CH=CH_2 groups (Fern  ndez et al., 2021). Additional confirmation of the degradation of PVC chains and the formation of its oxidation products are bands appearing in the $1100\text{--}980\text{ cm}^{-1}$ region originating from groups containing oxygen in their structure (Solovyeva et al., 2021). Their intensity increased with time of operation and was particularly pronounced for pipes with small cross-sections. Similarly to PE, bands originating from water ($3360\text{--}3370\text{ cm}^{-1}$) also appear on the spectra recorded for PVC (Solovyeva et al., 2021). Confirmation of the degradation of the PVC polymer backbone by oxidation, particularly intense in small diameter pipes, was provided by changes in the contact angle values. For PVC pipes, slightly higher CI values were observed for small diameters than for large diameters, amounting to $0,202\text{--}0,514$ and $0,11\text{--}0,207$,

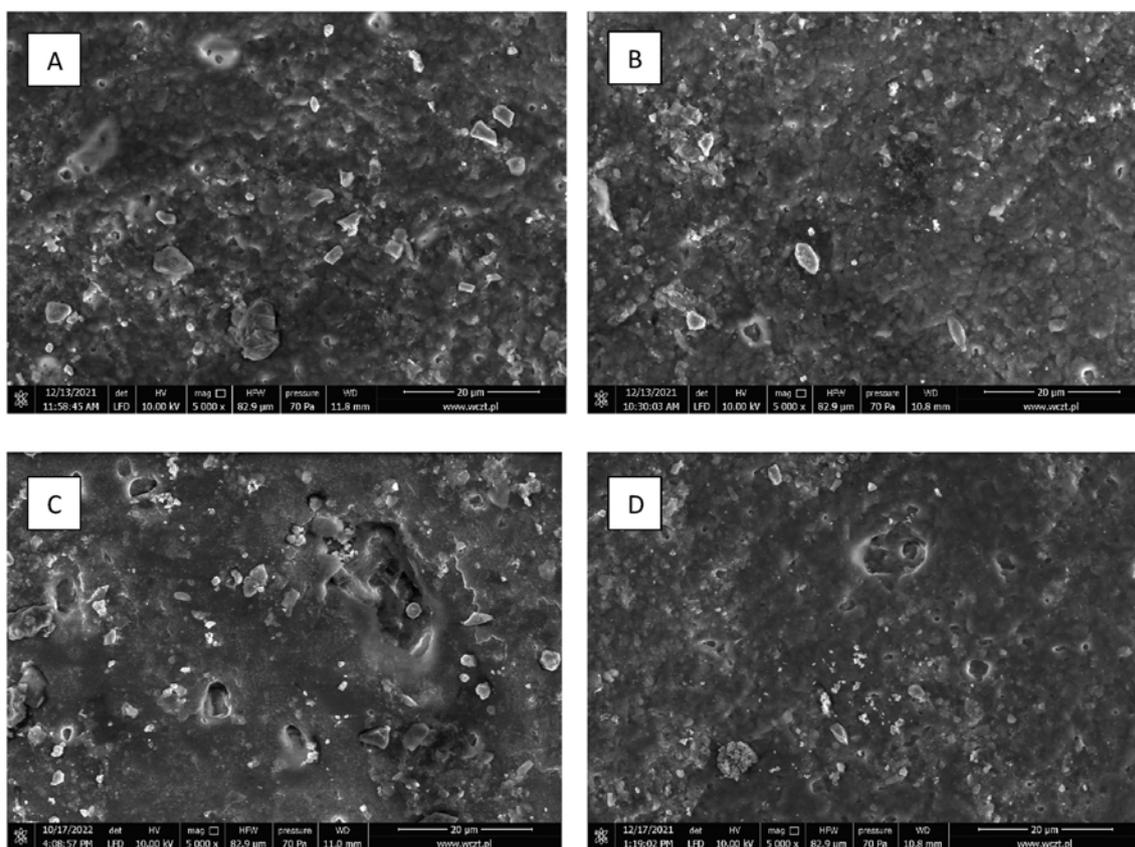


Fig. 12. Changes in the appearance of the inner surface of PVC pipes depending on their diameter: A) ϕ 80 mm (41 years), B) ϕ 100 mm (45 years), C) ϕ 160 mm (29 years) and D) ϕ 200 mm (36 years).

respectively (Fig. 4B). For large diameters, the CI values were very similar. An exception among the materials tested was a 110 mm diameter pipe that had been in service for 16 years and had the highest CI (2, 483), indicating a very high degree of degradation of this material. It is likely that the pipe was made of low-quality PVC. For pipes operating for about 30 years, similar CI values were obtained (in the range of 0,075–0, 202), while for the longest-operating pipes these indices increased, taking values of 0,371 after 41 years of operation and 0,514 for a 45-year-old pipe.

The youngest of the analyzed cuttings worked in the distribution network for only 16 years, which indicates that degradation processes occur on the surface of PVC from the first years of their operation (Fig. 9). In addition, during the formation of cavities, most likely as a result of peeling and detachment of the polymeric material, numerous fragments of varying morphology are formed, classified mainly as nanoplastics, which can be released into the distributed water. Thus, the aging process of PVC under operating conditions can be a significant source of microplastics in tap water.

Visible defects on the surface of PVC, can provide an excellent place for microorganisms to grow and thus biodegrade the polymer. They are also an extensive reservoir of mineral deposits, which over time can also provide a substantial breeding ground for bacteria. A study by Biedroń et al. (2017) found that the biofilm formed on PVC is thicker and has a more complex structure with multiple channels, compared to the biofilm observed on the surface of pipes made of polyethylene. This may be a consequence precisely of the presence of numerous irregularities on the PVC surface, which favor the undisturbed proliferation of microorganisms in isolated areas with impeded access to flowing water containing active disinfectant. Images showing mineral deposits, individual microorganisms and biofilm fragments on PVC are shown in Fig. 10.

3.2. Influence of service time, diameter and hydraulic conditions on the degradation process of PE and PVC pipes

A consequence of the progressive operational aging of water pipes are changes in such parameters as the roughness and internal diameter of the pipe. However, the narrowing of diameters occurs mainly in pipes made of metal alloys, which have corroded under operating conditions and their inner surface has become covered with tubercles (Nawrocki and Świetlik, 2011; Świetlik et al., 2012). Age-related changes in the roughness of metal pipes can have a major impact on the capacity and pressure drop of the distribution system, and thus adversely affect water flow (Christensen, 2009). In the case of pipes made of plastic, surface phenomena occur on a much smaller scale, which does not restrict the flow of transported water. However, in the case of plastics, their service life can be significantly affected by the ambient temperature causing a reduction in elasticity that can contribute to pipe rupture. The risk of failure increases with age and the degree of degradation of the plastic. In the case of low negative pressure in pipes, contaminated water, sewage and other impurities present on the outside of aging pipes can also be drawn into the interior through cracks and/or their degenerated "spongy" structure. It is extremely important that there is a laminar flow of water at a constant velocity in the pipeline, so that the pressure and quality of the water being transferred can be kept constant. Such conditions are easier to maintain in pipelines with smaller diameters, while pipelines with larger cross-sections are most often characterized by turbulent flow. In practice, it is very difficult to determine the hydraulic conditions prevailing in extensive distribution networks, as they depend on the diameter of the pipe, its location (center, dead end) and water demand. In the systems studied, the water pressure in the transmission pipes can vary from 2 to 4 Ba, depending on the pipe profile. However, water flow rates can vary over wide ranges from 0 to 18 m³/h for pipes with averages <100 mm to as much as 280 m³/h for pipes with

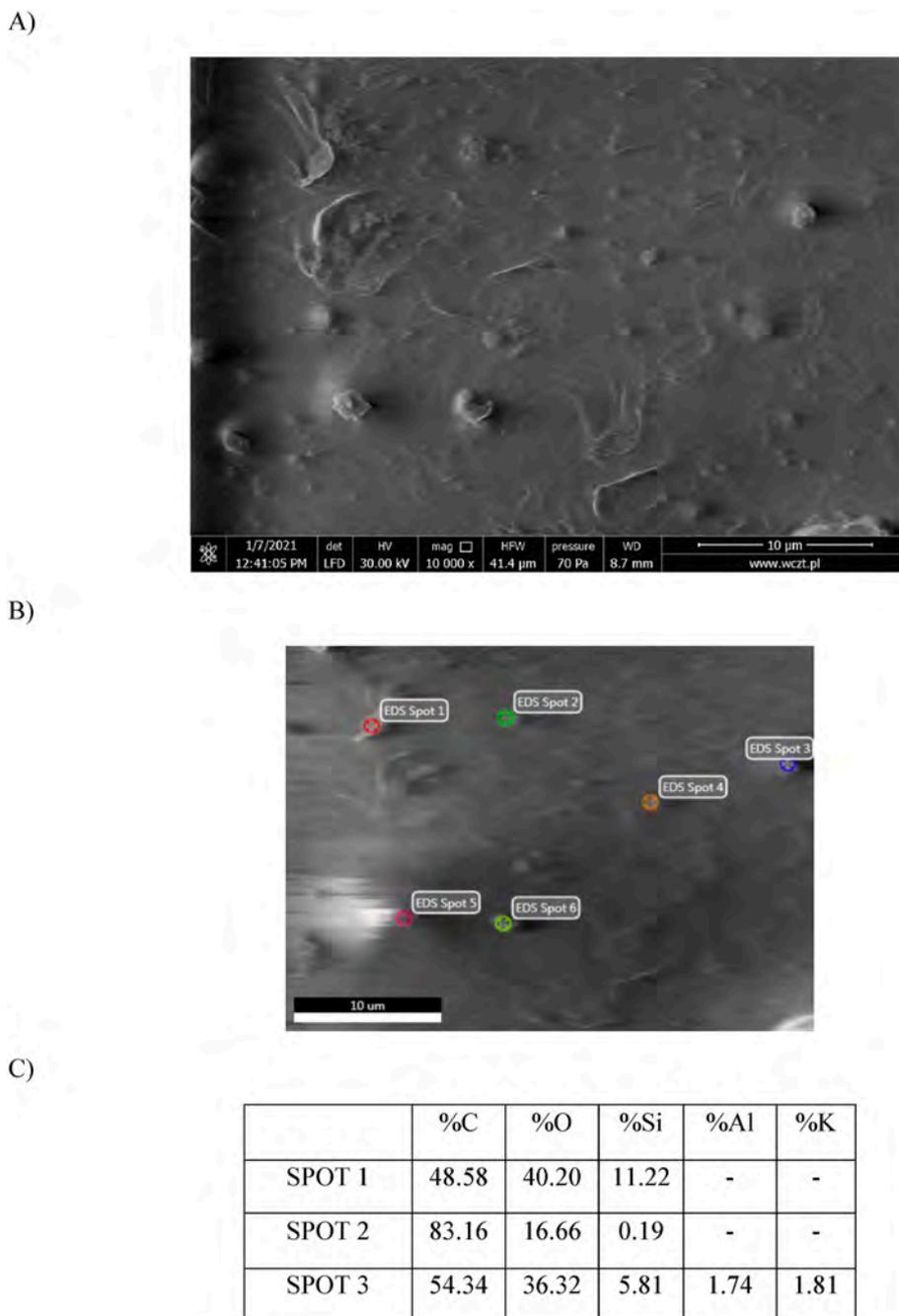


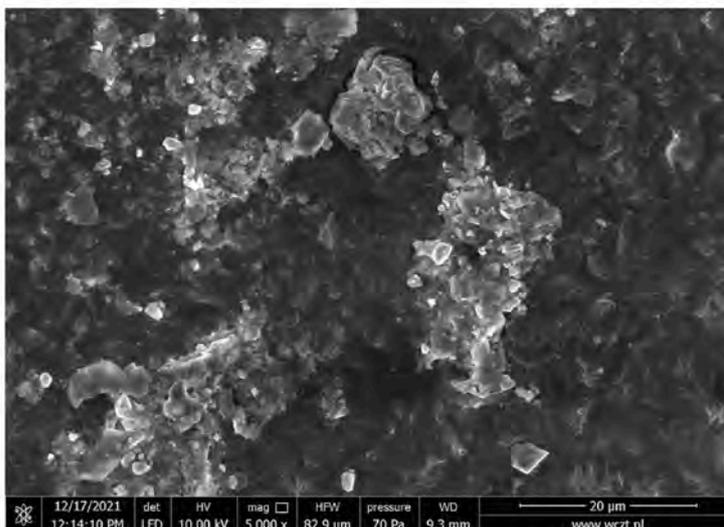
Fig. 13. A) SEM image of the surface of PE after 4 years of exploitation (180 mm), B) and C) EDS analysis of selected particles.

diameters >300 mm. In practice, flow rates are lower and the prevailing pressure in the pipes fluctuates over diurnal cycles. The lowest water flow rates, in the range of 0–1 m³/h, occur in the smallest diameter pipelines supplying the ends of distribution networks with variable water consumption. In these lines, the pressure is maintained at 2–2,5 Ba. Regardless of the cause, disturbed water flow can not only promote the migration of contaminants from the environment into the pipe, but also cause the detachment of sediments, biofilms and fragments of the polymers themselves located on the inner walls of the pipes and damage to the system (Liu et al., 2017). In order to determine the progression and intensity of degradation processes occurring in in-service water pipes made of PE and PVC, the service life and diameter of the pipes were also analyzed in detail. It was assumed that the diameter of the pipes studied is directly related to the hydraulic conditions prevailing in the distribution network.

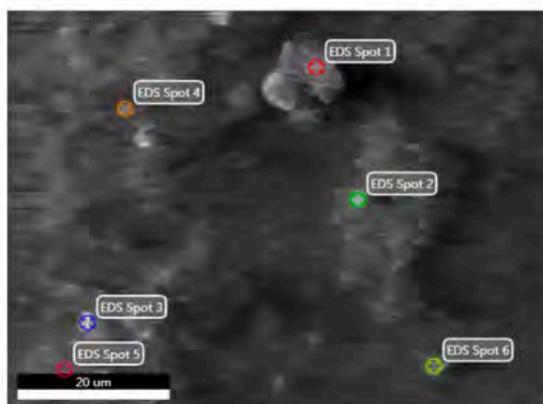
3.2.1. PE pipes

The microscopic images presented earlier (Figs. 1 and 2) show that the degree of degradation of the PE surface clearly increases with elongation of service life. The course of the polyethylene aging process is also significantly affected by the diameter of the pipes in service and the associated flow rate and water pressure [Świetlik and Magnucka, 2024 and references in it]. Fig. 11 clearly shows that in pipes of higher diameters the aging and surface degradation process is more intense than in those of small diameters. Despite the longer service life, the damage visible on the surfaces of small-diameter pipes is shallower and characterized by the coexistence of a smaller number of torn fragments, which is most likely related to the much slower, more laminar flow and lower water pressure acting on the walls. In pipes with large cross sections, the visible damage is deeper, covers larger areas of the surface and has a greater potential to release exfoliated plastic micro-fragments into

A)



B)



C)

	%C	%O	%Al	%Si	%Mg	%Na	%K	%Fe
SPOT 1	31.92	47.32	4.31	11.71	-	4.08	0.32	0.35
SPOT 2	50.18	45.33	1.06	2.38	0.20	-	0.20	0.65
SPOT 4	81.21	17.49	0.29	0.81	-	-	0.21	-
SPOT 5	52.58	44.67	1.00	1.76	-	-	-	-
SPOT 6	89.40	10.38	-	0.21	-	-	-	-

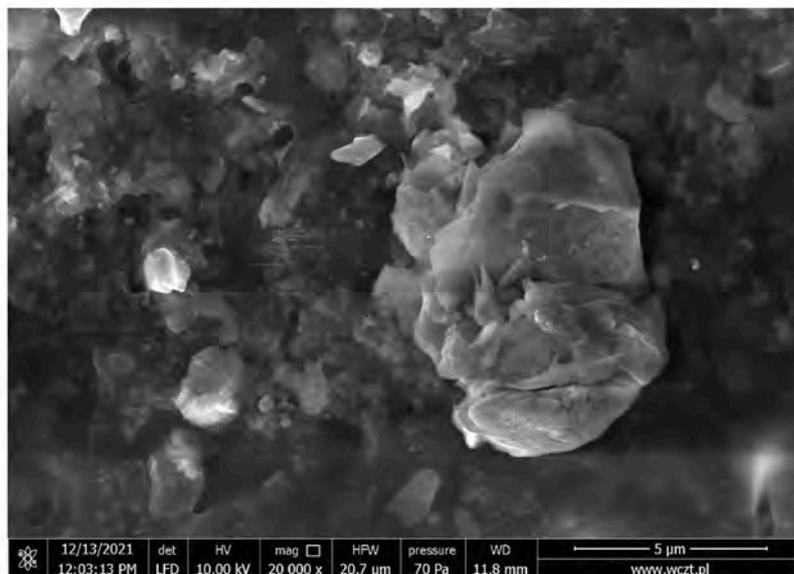
Fig. 14. A) SEM image of the surface of PE after 14 years of exploitation (225 mm), B) and C) EDS analysis of selected particles.

the water. However, the high flow rate and high water pressure limit the proliferation of microorganisms on the PE surface, indicating that despite the numerous defects, this type of pipe will be less susceptible to microbial degradation than small-diameter pipes. Therefore, it can be concluded that in the case of polyethylene, the degradation processes that deepen over time affect all water pipes in operation, while in small-diameter pipes the oxidative aging of the polymer can be intensified by bacteria in biofilms, and in large-diameter pipes mainly by the rate and type of flow and water pressure.

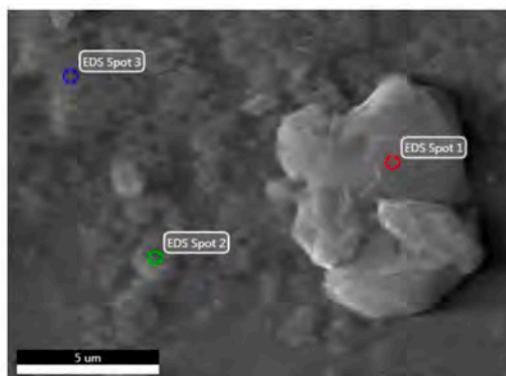
3.2.2. PVC pipes

For the analysis of PVC cuttings, similar relationships were observed for PE. Due to the age of the acquired sections (16–45 years), the rate of degradation of this polymer could not be accurately estimated. The microscopic images in Figs. 6 and 7 clearly show the advanced degradation of all the fragments, making the intensification of the process as a function of time difficult to grasp. Analysis of images recorded for larger areas of the surface (not shown) confirmed the progressive nature of the processes in question. On the PVC surface, however, clear changes in the appearance and depth of damage were observed depending on the diameter of the pipe (Fig. 12).

A)



B)



C)

	%C	%O	%Cl	%Al	%Si	%Ca	%Pb
SPOT 1	10.99	48.30	22.87	0.74	16.65	0.45	-
SPOT 2	14.74	36.39	45.40	0.62	1.40	0.67	0.78
SPOT 3	21.85	30.35	45.53	0.32	0.74	0.63	0.58

Fig. 15. A) SEM images of the surface of PVC after 41 years of exploitation (80 mm), B) and C) EDS analysis of selected particles.

On the surface of small-diameter pipes, numerous particles and fragments of torn material were visible, as well as numerous small pits. Wider pipes, on the other hand, were characterized primarily by the presence of numerous deep pits around which peeling areas and plastic particles were visible. This confirms the degrading effect of flow rate and water pressure on PVC, as was the case with PE. Of concern, however, is the large, increasing number of loose particles present on the PVC surface with small diameters. This is because it indicates that as PVC pipes age, they have an increasing potential to release nano- and microplastic particles into the water. In the case of small diameters, such pipelines are mainly found at the ends of the network and deliver drinking water directly to consumers' homes, significantly exposing them to the introduction of unwanted microplastics into their bodies via tap water as well. A detailed analysis of the observed microplastics will be presented

in the next chapter.

3.3. Chemical characterization and elemental analysis of microfragments present on the inner walls of PE and PVC pipes

Sections of water pipes made of polyethylene and polyvinyl chloride, after cleaning and etching (ensuring complete purification of the walls from mineral and organic deposits), were subjected to full characterization using the EDS technique. The elemental composition of the microfragments present on the tested materials was analyzed to determine and/or confirm their origin.

3.3.1. PE pipes

Elemental analysis of PE cuttings showed that most of the particles

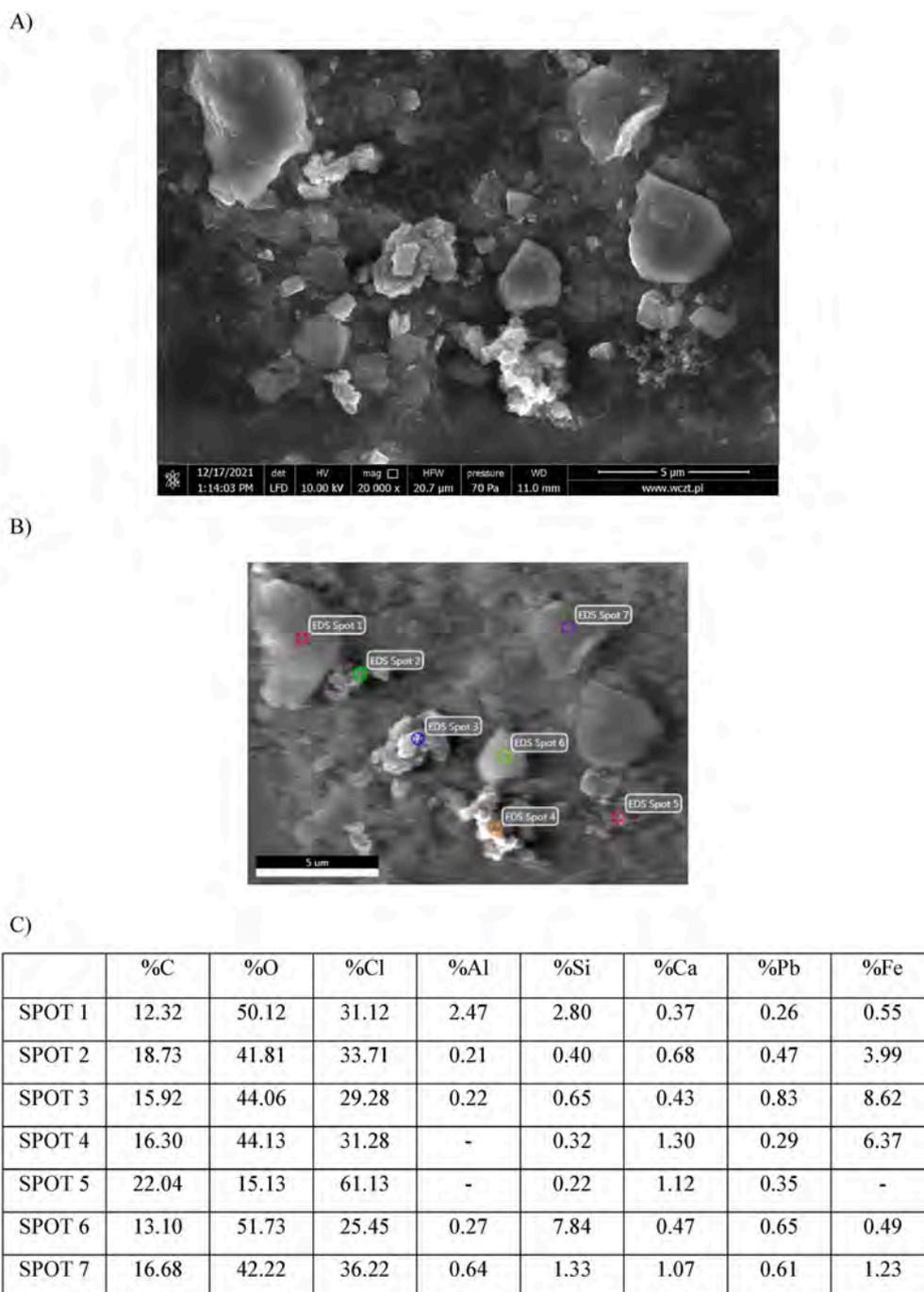


Fig. 16. A) SEM image of the surface of PVC after 36 years of exploitation (200 mm), B) and C) EDS analysis of selected particles.

found on the inner walls of the pipes are fragments of peeling antioxidant coatings and polyethylene itself. The elemental composition of the analyzed particles varied due to different degrees of oxidation, a consequence of the degree of peeling and residence time in the network. Some of the fragments were characterized by a low percentage of oxygen (a few - several percent), indicating that the particles were detached from the polymer relatively recently. On the other hand, in the case of particles that were in contact with flowing water for a longer period of time, the percentage oxygen content increased to 40% or even more. Figs. 13 and 14 show examples of EDS analyses obtained for PE after 4 and 14 years in service, respectively. On the surface of the shorter-exposed materials, surface wrinkles were clearly visible, resulting from the degradation of the antioxidant coatings (Fig. 13). The elemental composition of these "wrinkles" confirmed that they are most

likely torn/separated antioxidant coatings, characterized by high carbon content, reaching up to 80% C in a few cases (Fig. 13), while the content of this element in the unoxidized polymer is about up to 86%. This confirms the earlier hypothesis, according to which the antioxidant coating is first broken/damaged, followed by peeling and exposure of the polymer. As a consequence, its gradual degradation occurs, resulting in the phenomenon of the release of microfragments of plastic into the water. The torn and partially detached PE particles undergo further oxidative processes, as indicated by the increasing percentage of oxygen in the particles most loosely bound to the polymer surface (Figs. 13 and 14).

3.3.2. PVC pipes

The images of the analyzed PVC particles shown in Figs. 15 and 16

present a variety of their shapes. Fragments with sharp irregular edges predominate, but the presence of fragments with fibrous shapes was also noted (not shown). The elemental composition of the scales and particles, present on the walls of PVC pipes, subjected to elemental analysis varied with respect to oxygen content, indicating the degree of oxidation of the polymer, as well as with respect to chlorine content. Fragments detached from PVC relatively recently were characterized by a chlorine content of >45% (Fig. 15), while for particles that had been in contact with flowing water for a longer period of time, the % chlorine content decreased depending on the degree of degradation and averaged from 42 to about 25% (Fig. 16). As the chlorine content decreased, the oxygen content of the particles increased, which directly indicates that the fragments detached from the polymer matrix are successively oxidized over time by oxidants present in the flowing water (dissolved oxygen, disinfectants). In addition, almost all analyzed PVC microfragments showed the presence of admixtures of lead, used in the manufacture of PVC pipes to increase the resistance of the plastic to temperature changes. The percentage content of Pb most often ranged from 0 to <1%, but in a few cases it was higher, reaching from 1,2 to 6,6%. Also of great interest were the particles represented by the "Spot 5" visible in Fig. 14, for which a relatively high carbon content and a very high percentage of chlorine, exceeding 60%, were determined, along with a low percentage of oxygen. Given the claimed long life of the pipes (in this case 36 years, Fig. 16), these particles were most likely trapped under a layer of mineral and biological deposits that had accumulated over the years, resulting in limited oxygen/disinfectant access. This proves that in the case of PVC pipes with a very long service life, in addition to particles released systematically into the transmitted water, there is also an accumulation of microplastic fragments permanently bound to the deposits present on the internal surfaces of the pipes. The presence of such microplastic particles trapped in sediments deposited on the inner walls of distribution pipes was confirmed by Chu et al. (2022).

4. Conclusions

Investigations of PE and PVC distribution pipe sections, operated for a long time in working DWSN, confirmed that both plastics are gradually degrading, causing increasing damage to their inner walls. Progressive degradation of antioxidant coatings and the plastic itself was observed, leading to an increase in porosity, cracking and flaking of the PE surface, causing the release of small fragments of plastic classified as fine microplastics and nanoplastics into the water. Elemental analysis of the scales and micro-fragments confirmed that these are PE particles with varying degrees of oxidation. Spectroscopic studies of long-life pipes showed that PE aging results in the cleavage of polymer chains, an increase in the material's amorphousness and the formation of oxidized degradation byproducts. In the case of PVC, the presence of numerous pits/gaps/holes was observed, distributed fairly evenly across the entire inner surface of the walls of the analyzed water pipes. Pitting was present on all sections regardless of their age and diameter, but their number and depth increased with increasing pipe service life. On the surface of PVC there were more numerous exfoliations and loosely bound fragments of different morphologies than in the case of PE. The elemental composition of the particles confirmed that they were PVC fragments with varying degrees of oxidation, which could be classified as nano- and microplastics. Spectroscopic analysis showed that the aging of PVC is mainly through oxidation, which results in the cleavage of polymer chains, the exposure of deeper polymer layers and the formation of degradation products containing oxygen-rich groups. The degree of surface degradation of PE and PVC increased with increasing service life, with the course of the process and the type of damage caused depending on the diameter of the pipes in service and the operating conditions, such as pressure, flow rate and periods of stagnant water. Long-term operation of PE and PVC pipes and the associated progressive degradation of their surfaces leads to the formation of an increasing number of irregularities conducive to colonization by microorganisms,

which, combined with a decrease in the speed of water flow and the disappearance of residual disinfectants, can lead to the development of biological coatings (biofilms) on the walls of pipes. The aging process of pipes made of thermoplastics occurs relatively quickly. Significant surface damage occurs already in the first decade of operation, which significantly shortens the life of pipes, and increases the risk of secondary contamination of drinking water with nano- and microplastics. In the case of both plastics, the number of nano- and microplastic particles with high release potential into the water increased as the diameter of the pipes decreased, indicating that customers at the ends of the network to which drinking water is supplied by small-diameter pipes are most vulnerable. Microplastics and nanoplastics released into drinking water from PE and PVC pipes may pose a potential health risk to consumers due to their ability to bind and carry toxic contaminants on their surface and their small size indicating that they can penetrate human and animal tissues and bloodstream, as described in the literature. Therefore, aging PE and PVC pipes should be considered a real and very important source of micro- and nanoplastics in tap water, and drinking water producers should include monitoring of plastic content in regular quality control of the distributed product.

Data availability

The datasets generated during and/or analyzed during the current study are available from the corresponding author on reasonable request.

CRediT authorship contribution statement

Joanna Świetlik: Writing – review & editing, Writing – original draft, Supervision, Project administration, Methodology, Funding acquisition, Data curation, Conceptualization. **Marta Magnucka:** Writing – original draft, Visualization, Methodology, Investigation.

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