Gravimetric, mechanical and chemical characterization of different materials used in sewers systems: Polyvinyl chloride (PVC), polypropylene (PP) and high density polyethylene (HDPE), aged in sulfuric acid at 60°C

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ABSTRACT - *The effect of aging on mechanical and structural behaviors of PVC, PP, and HDPE was investigated by acid solution at 60°C. The curves of gravimetric measurements for PVC and HDPE during testing time reveal that the water absorption follows the Fickian process, while the curve of water absorption for PP follows a non-Fickian behavior. While the degradation progresses, an increase in tensile strength, a great drop in elongation at break and a clear brittle failure mode for PVC, PP and HDPE specimens was observed. The increase in tensile properties is due to the increase in crystallinity and cross-linking density. The decrease of tensile properties is due to the scission of molecular chain. The changes in functional groups were monitored by using the Fourier transform infrared spectroscopy (FTIR). These results show that the principal functions of polymers are the aliphatic hydrocarbons. After aging, the change and appearance of new peaks signify an oxidation of hydrocarbon groups (CC, CH) due to the acid attack, effect of temperature and aging time. These results show that there is a good relationship between the diffusion of solvent as well as the increase of time exposure and temperature on the mechanical properties and function of polymers.*
Keywords – FTIR, mechanical properties, polymer materials, sulfuric acid, water absorption.

1. INTRODUCTION

The environment in some sewer structures can become very acidic due to the formation of sulfuric acid converted from the hydrogen sulfide which is generated by bacterial activities [1-5]. Corrosion deterioration has long-term effects on the environment, economy, and society [5] and may result in serious problems such as the loss of ability to transport sewerage, contamination of ground and groundwater and collapses [4]. Significant deterioration of concrete in such harsh environments has been reported worldwide [4]. Consequently, it has been reported that some new materials can be more resistant to acid such as polymer materials. They are recently more and more frequently used in sewer systems and became an indispensable element of the human life [6,7]. Polyvinyl chloride (PVC), polypropylene (PP) and high density polyethylene (HDPE) are materials for polymer pipes which are used for sewer networks. Their massive use is a matter of great environmental and economic concern [7], due to their low production costs, light weight and efficient performances [8].
For example, a PP water pipe has a variety of advantages, such as the high performance-to-cost, the light weight, the thermal stability and chemical resistance [10]. Furthermore, according to Maiara et al. [9], high density polyethylene (HDPE) has balanced mechanical properties, chemical resistance, impermeability to water and low costs. Moreover, according to TIKKA et al. [11] polyvinyl chloride (PVC) is one of the most widely used thermoplastics in the world and its production costs are low.

On the other hand, as reported by A. Boubakri et al. [6], the durability of polymer materials became an object of interest when they were used in aggressive environments such as temperature, aggressive chemicals and mechanical stresses. Depending on the degree of exposure conditions this may lead to a decreasing of the polymer lifetime. As reported by A. Boubakri et al. [6], the mechanical and chemical effects are the main reasons for the degradation of the polymer.

Therefore, there is a considerable interest to know in which way the acid degradation has an effect on the mechanical and structural behaviors of polymeric materials.According to J. Weon [10], the long-term exposure for a PE pipe to an elevated temperature condition causes adverse effects on the mechanical and thermal properties. When the aging time increases, an increases in crystallinity, molecular weight, chain scission, hydroxyl, in carbonyl, and/or vinyl group are found; and a reduction of the antioxidant and the amorphous region are observed. According to J. Andrew Whelton et al. [12], the long-term exposure for a PE pipe to water containing free available chlorine causes adverse effects on the mechanical and surface characteristics. They have found a reduction of the antioxidant levels, an increased crystalline content and a reduction the in amorphous region, a chain scission and an increased hydroxyl, carbonyl, and/or vinyl group content.

According to R.K. Rowe et al. [13], the tensile properties of HDPE GM in a laboratory immersion test is useful to understand how it will react to physical stress and to evaluate oxidative stability due to aging. As a consequence of the oxidation that occurs during degradation, the tensile properties begin to change. According to Zaki et al. [14], the diffusion in the polymeric materials is of fundamental importance in many applications. The diffusion of chemicals into a polymer component may affect the mechanical performances and degrade the material. As reported by A. Boubakri et al. [6], the degradation process of the polymer material leads to several physico–chemical and mechanical modifications and is influenced by several factors such as chemical properties, aging temperature, aging time and immersion solution. The present paper attempts to study how the aging in the acid solution at 60°C has an effect on the mechanical and structural behaviors of PVC, PP and HDPE in order to acquire a fundamental understanding of the degradation mechanism of these materials.

II. MATERIALS AND METHOD

2.1 Materials

The materials tested in this study as cited in Lasfar et al. [1] are: PVC, PP and HDPE; and they are used in sewage systems. The density of the HDPE was 0.96 (NM ISO 1183-1) and the melt index (190°C, 5Kg) was 2.38 g/10 min (ISO 1133). The density of the PP was 0.91 and the melt index (190°C, 5Kg) was 0.71g/10 min. The density of the PVC was 1.44 and the VICAT softening temperature (VICAT) was 85°C (NM EN 727 2013). The specimens were obtained according to NM ISO 6259-2 and were cut from the longitudinal direction for the PVC sewer pipe (Ø 400 × 300 × 10 mm³) and from the transversal direction of the inner wall for the PP and HDPE annulated sewer pipes (Ø 1000 mm).

2.2 Exposure

The polymer samples were degraded by a continuous immersion in 4 liters of sulfuric acid (H_2SO_4) solution 5000ppm of SO_4^{2-} with a pH=1 at 60±1°C. The maximum immersion time considered in this test is equal to 103 days.

2.2.1 Gravimetric measurements

The gravimetric method is one of the most used methods to measure the absorption of a solution by materials [14].

The weight of each specimen was measured and recorded every week, after drying the samples with a cloth, by using a technical balance with an accuracy of ±0.01 mg. The weight gain m(t) was calculated from the specimens initial weight M(0) and the specimens weight before exposure to the sulfuric acid M(t) as follows (1) [1-3,13-15] :

$$m(t) = \frac{M(t) - M(0)}{M(0)} \times 100 \qquad (1)$$

To ensure the reliability of the measurement, five specimens of each material were weighed.

2.2.2 Tensile test

The tensile properties were evaluated following the guidelines of the EN ISO 6259-1 standard. The tensile test was carried out using a Zwick Roell Z010 testing machine at room temperature with a strain rate of 5 mm/min till rupture for the PVC specimens and with a strain rate of 100 mm/min till rupture for the PP and HDPE

specimens. An extensometer was employed to measure the displacement in the gauge length region. The average value was recorded after the testing of three specimens of each material.

2.2.3 Analysis by Infrared Spectroscopy (FTIR)

The infrared (IR) spectra were measured by attenuated total reflection (ATR) on the surface of the specimens. The FTIR instrument used for polymer materials was a model NICOLET IS10.

III. RESULTS AND DISCUSSIONS

3.1 Gravimetric measurements

Water diffusion characteristics in polymers are usually characterized by weight gain measurements [16]. Fig.1 shows the evolution of the mass of PVC, PP and HDPE pipe specimens immersed in sulfuric acid (H_2SO_4) depending on the square root of time at 60°C.
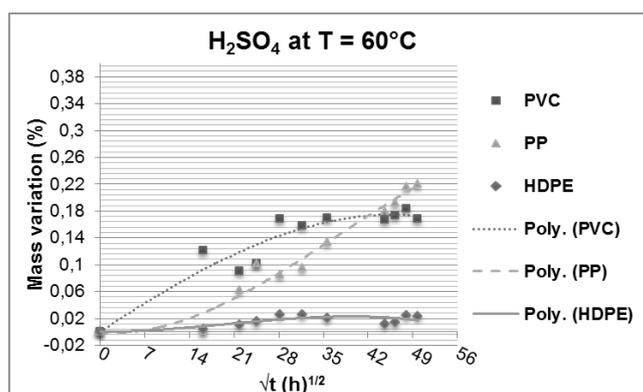

Fig. 1 Weight change as a function of time ($\sqrt{t}$ ($h^{1/2}$)) for PVC, PP and HDPE exposed to H_2SO_4 at 60°C

As plotted in fig.1, the curves for PVC and HDPE during the testing time reveal that the water absorption follows the Fickian process. The curves show first a linear relation between the absorption rate and the square root of the duration of immersion, characterized firstly by a phase of water absorption and weight gain, indicating the penetration of water into the amorphous parts of the material till saturation [1-3,17], and secondly by a relatively stable phase characterizing a "pseudo plate" of the diffusion. Furthermore, as plotted in fig.1, the curve for PP reveals that the water absorption follows a non-Fickian behavior, characterized by a continuous absorption over time, which is usually accompanied by significant deformations and damages within the material [1.18].

On the other hand, for the absorption of HDPE, a Fickian behavior during the testing time, while this material is generally known by a non-Fickian behavior, characterized by a weight loss after a certain period of aging. Several authors have found the same behavior [1,2,19]. Such behavior is explained by an internal reorganization of the polymer chains, by a leaching of some components of low molecular weight of the material and is also explained by the migration of crosslinking agents to the surface during aging [1,2,19].

3.2 Mechanical characterization

3.2.1 Tensile curves

Tensile curves of HDPE, PP and PVC pipe specimens in the initial state and in degraded state (t> 12 days) were selected. Figs.2, 3 and 4 show the curves of stress versus elongation for HDPE, PP and PVC specimens respectively, before and after aging in sulfuric acid at 60°C.
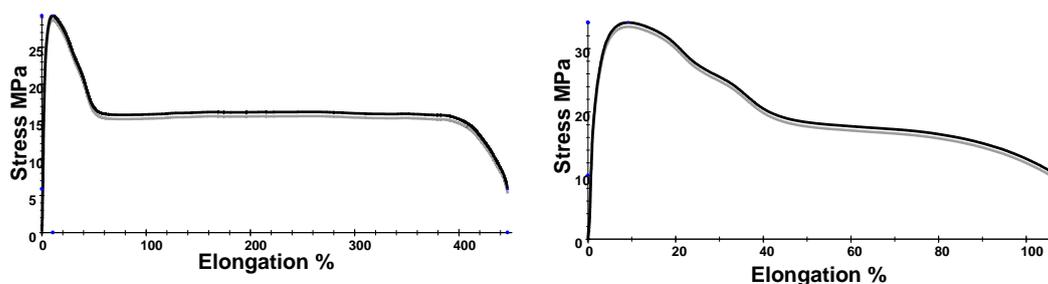
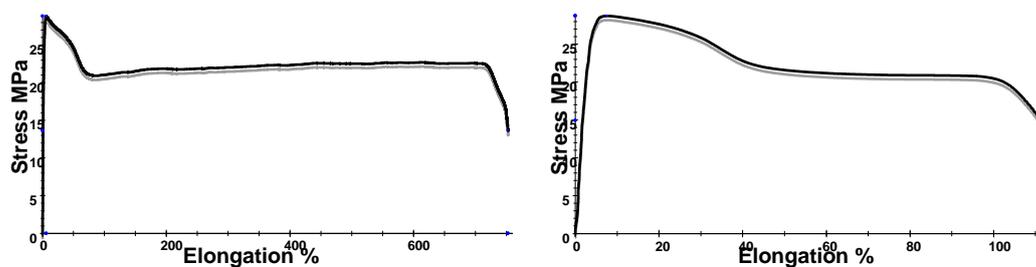Fig. 2 Curves stress/elongation for HDPE before and after aging in H_2SO_4 at 60°C

Fig. 3 Curves stress/elongation for PP before and after aging in H_2SO_4 at 60°C
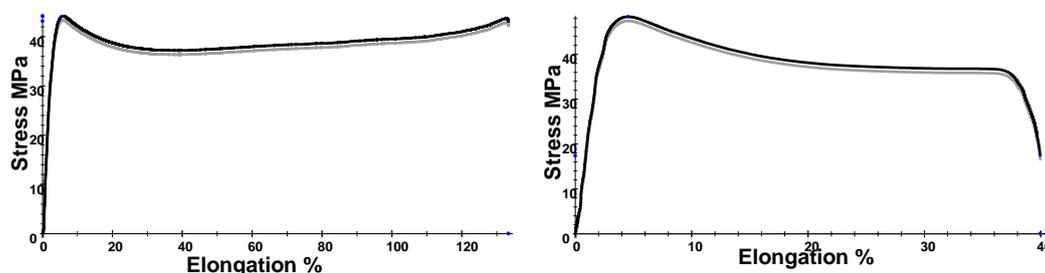

Fig. 4 Curves stress/elongation for PVC before and after aging in H_2SO_4 at 60°C

The stress-strain curves of plastic materials (figs.2, 3 and 4) can be divided into four parts:

A first part of the curve, ascending and linear up to about 42.58 MPa for PVC, to 29.9 MPa for the PP and up to 28.38 MPa for HDPE, allows to determine the modulus of apparent elasticity of the material. In this part the deformation remains reversible. After this point, a first transformation of the specimen due to a phenomenon of necking was observed. The module "falls" quickly to achieve a plate of deformation with an almost constant stress. The necking spreads across the entire sample causing a chain orientation before an increase of stress followed by a total rupture [17,18]. The four steps are the same for all three materials. The behavior of the PVC, PP and HDPE at the initial state is a ductile behavior with an elongation at break equal to 126.2% for PVC, 706.53% for PP, 386.43% for HDPE. After aging in H_2SO_4 at 60°C, brittle behavior was found (immediate break after aging). Andrew Whelton et al. [12] have found a great variability of the results of the elongation at break and a clear brittle failure mode for HDPE pipe samples aged in 250 ppm of Cl_2 at 37°C in comparison to the ductile fracture for new specimens. This ductile failure can be attributed to the chain fracture as well as the chain disentanglement, which were caused by the oxidation of the induced chlorinated water.

As reported by R. K. Rowe et al. [13], as degradation progresses the HDPE GM becomes increasingly fragile. The decrease in tensile properties at break signifies the transition from a ductile to a brittle material as a result of degradation.

Generally, temperature accelerates the diffusion rate of the aging process [6].

3.2.2 Results of mechanical characterization

The mechanical properties of PVC, PP and HDEP samples aged in sulfuric acid at 60°C were investigated. The key mechanical property (i.e., tensile strength and elongation at break) depending on time is shown in figs.5 and 6 respectively.
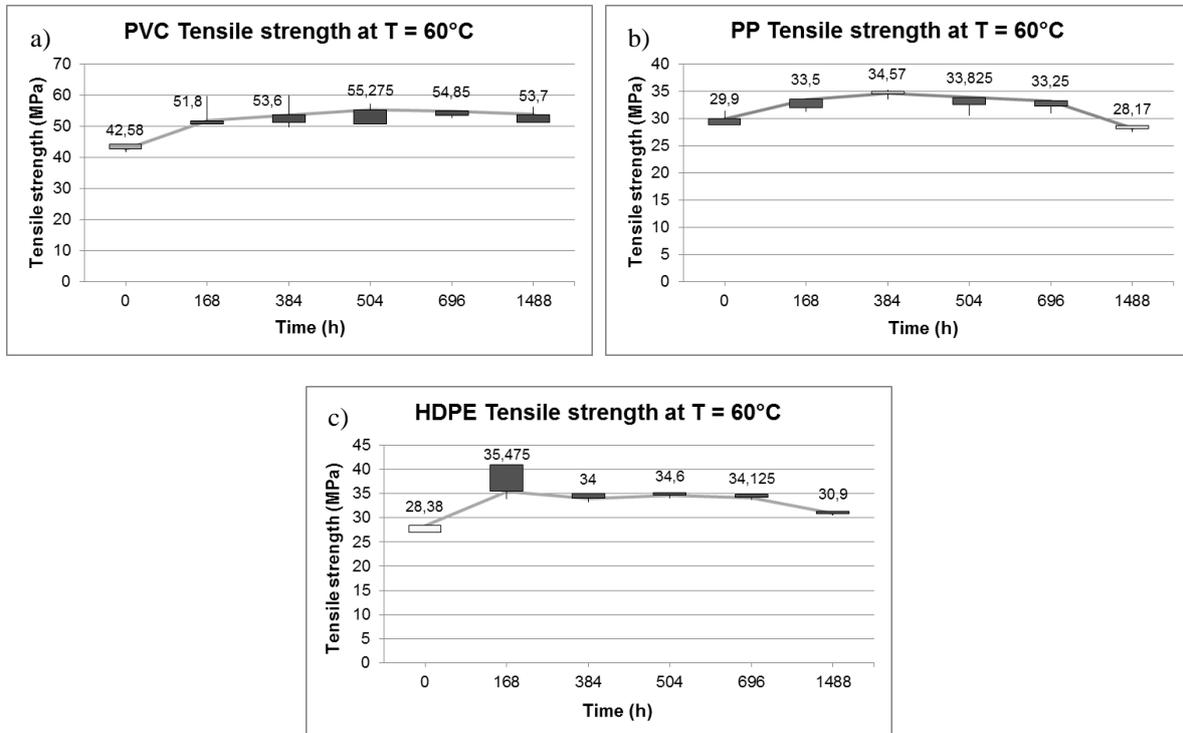

Fig. 5 Tensile strength as function of time for: a) PVC, b) PP and c) HDPE, pipe samples aging in sulfuric acid solution at 60°C
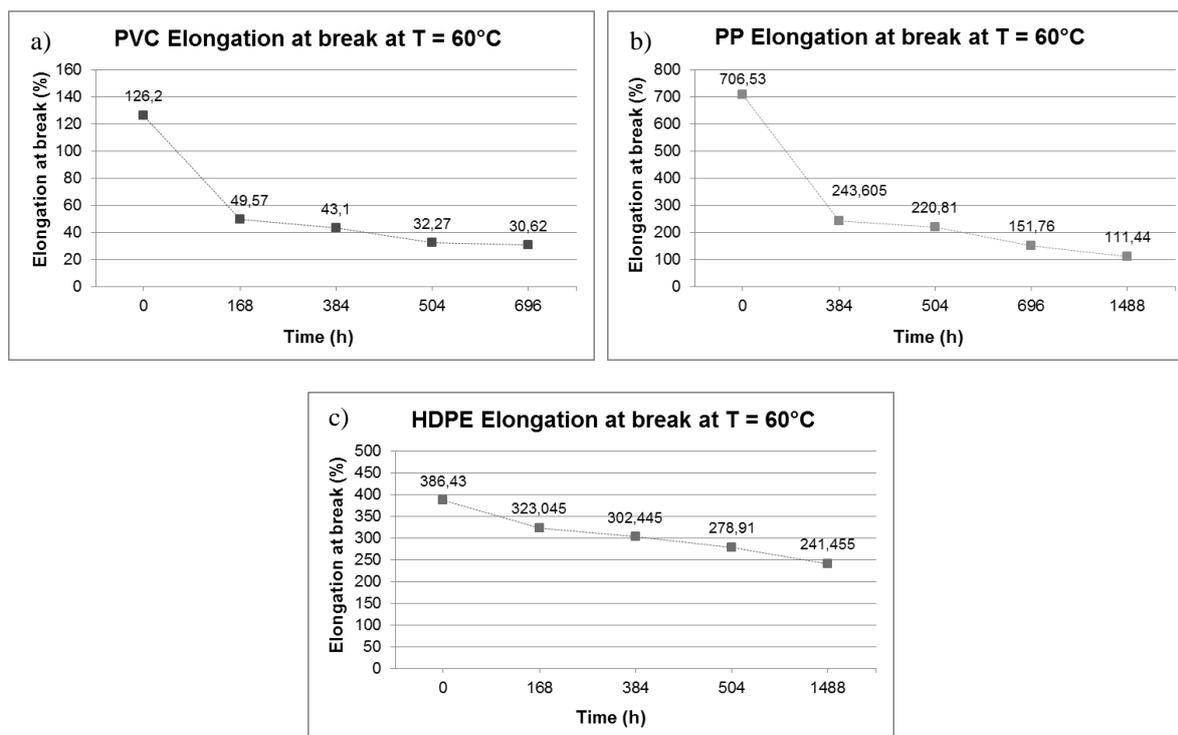
Fig. 6 Elongation at break as function of time for: a) PVC, b) PP and c) HDPE, pipe samples aging in sulfuric acid solution at 60°C

According to figs.5 and 6 for all three materials an increase followed by a decrease in tensile strength at the end of the test was observed. A decrease of the elongation at break was also found: after 600h, a drop of 126.2% to 30.62% for PVC, a drop of 706.53% to 151.76% for the PP and a drop of 386.43% to 241.455% for HDPE. A strong decrease in the elongation at break as a function of aging time and temperature for the materials aged in sulfuric acid was concluded.

These results are explained by the increase in crystallinity, followed by an increase in crosslinking density, due to the diffusion of the solvent into the material, the effect of the temperature and an increasing aging time [1]. Several authors have confirmed these results. To better explain these phenomena, J. Weon et al. [10] gave clear interpretations: They found that the prolongation of the thermal exposure time for the linear low density polyethylene (LLDPE) sample causes a progressive increase in tensile strength and a decrease in elongation at break. These results can be explained by the increase of crystallinity, followed by the increase of crosslinking density due to thermal oxidation. The elevated temperature leads to the increases in chain alignment and lamellar thickness of polymers by the increase of the chain mobility. This leads to the increase of tensile strength by the increase of the secondary intermolecular bonds. Then, the oxidation reactions develop the crosslinking process which provokes an amelioration of resistance, accompanied by deleterious effects on the mechanical properties due to chain scission. Finally, as reported by J. Weon et al. [10], the phenomena which are responsible for the decrease of elongation at break during thermal aging are a reduction in chain segment by molecular chain scission, in density of chain entanglement, in chain mobility and elasticity.

Following these interpretations, Mendes et al. [20] show that the increase in tensile strength and the decrease in elongation at break of polyethylene confirm the presence of two competing degradation mechanisms: the scission and the crosslinking. Furthermore, according to R. K. Rowe et al. [13], they show that the increase in the mechanical properties (yield strength) can be attributed to the physical aging indicated by the increase in crystallinity.

B. Abbès et al. [21] show that the diffusion of solvent and the increase of temperature affect the mechanical behavior of the PP polymer, by increasing the movement of the amorphous part, which leads to diminished mechanical properties.

According to D. Bouguedad [22], the increase in mechanical properties (tensile strength) is due to the increase in the crosslinking density while the decrease in mechanical properties (tensile strength and elongation at break) is due to the molecular chains scission. This last phenomenon causes a decrease in the average molecular weight and in the degree of crosslinking as well as a loss of plasticizers by the augmentation of the chain mobility. Again according to D. Bouguedad [22], the degradation rate increases with increased temperature. According to P.Y. Le Gac et al. [16], for polydicyclopentadiene (pDCPD) aging in seawater at temperatures below Tg, the only mechanism involved in the degradation is oxidation. This oxidation leads to an increase of the network density in the material, which results in a modification of the tensile behavior and a decrease of the elongation at break.

3.3 Spectroscopy FTIR
FTIR analyses were conducted to evaluate the degree of degradation for HDPE, PP and PVC pipe specimens removed after aging in H_2SO_4 at 60°C.

3.3.1 Spectra of HDPE, PP and PVC in the initial state
Figs.7.a, 7.b and 7.c show the absorbance spectra of the materials HDPE, PP and PVC at the initial state respectively as function of wavenumbers. These spectra are characteristic of the material and are used as reference spectra for the study after aging.
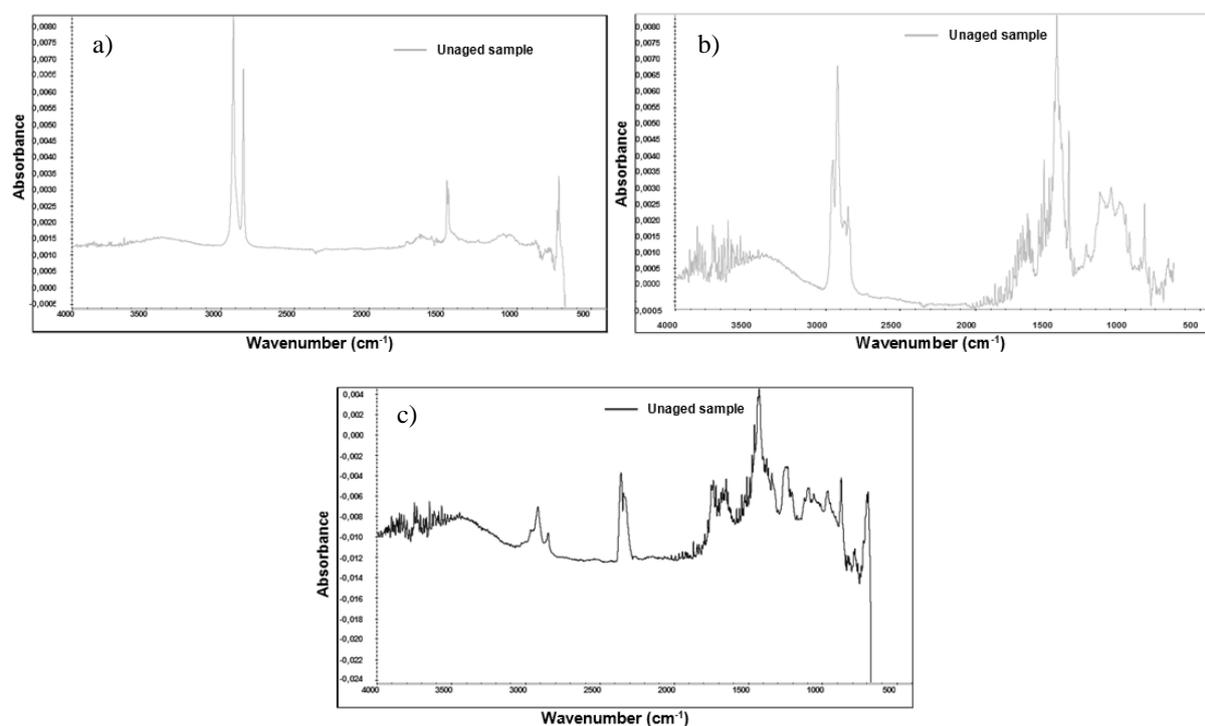

Fig. 7 FT-IR spectra at initial state of: a) HDPE, b) PP, and c) PVC

According to figs.7.a, 7.b and 7.c, the three materials have characteristic peaks of the aliphatic hydrocarbon function. This is the main function of these materials in the initial state. The most important vibration modes are the CH stretching around 3000 cm^{-1} and CH deformation modes around 1460 cm^{-1} and 1380 cm^{-1} [23,24].

According to C-J Chen et al. [25], FTIR spectroscopy has been proved to be a useful technique for the characterization of intermolecular interactions (mainly hydrogen bonding and dipole–dipole) between groups in the self-polymer or different polymer molecules in blend systems, leading to changes in absorbance of specific functional groups. According to A. Kositchaiyong et al. [26], characteristic peaks of PVC are the peak intensities at wave numbers of 2960-2850 cm^{-1}, 1470-1350 cm^{-1} and 800-600 cm^{-1} (C-H stretching, C-H vibrations, and C-Cl stretching). Furthermore, according to I.S. Elashmawi et al. [27], the absorption bands for pure PVC are observed at 3440 (OH stretching), 2913 (C-H stretching), 1631 (C=O), 1251 (C-H deformation), 964 and 611cm^{-1}. More representative bands of PVC are the bands at 1096 (C-C streching) and at 966 (CH$_2$ rocking). According to H. Rajandas et al. [28], HDPE is made of the elements carbon (C) and hydrogen (H), which form chains of repeating CH$_2$ units. Bonds which are naturally present in the polymer are CH$_2$ asymmetrical stretching, CH$_2$ symmetrical stretching, bending deformation, CH$_3$ symmetric deformation and rocking deformation. Again, according to V. Parthasarathi et al. [29], important peaks of a new HDPE are: peaks between 2600 and 3000cm^{-1} (C-H stretching vibrations), vibration of peaks at 1490cm^{-1} (C-H bending vibration), and a sharp peak at 730cm^{-1} (C-H out of plane bending vibration).

Furthermore, according to E. Parparita et al. [30], the FTIR spectrum of PP displays the bands assigned to different stretching vibrations of the methyl and methylene groups. For example, the four prominent bands at 2959, 2920, 2873 and 2839 cm^{-1} attributed to asymmetric and symmetric stretching vibrations of the methyl and methylene groups. Bands at 1458 and 1377 cm^{-1} (CH$_3$ asymmetric and symmetric bending), bands at 1361 cm^{-1} (CH bending and CH$_2$ wagging vibrations) and other important bands are also described by [30].

3.3.2 Spectra of HDPE, PP and PVC after aging in sulfuric acid at 60 °C

Figs.8.a, 8.b and 8.c show the transmittance spectra of the materials HDPE, PP and PVC respectively as functions of wavenumbers after 83 days of aging in H$_2$SO$_4$ at 60°C. Transmittance results were given, because usually the spectral interpretations are based on the transmittance as a function of wavenumbers, even for the absorbance spectra in the initial state.
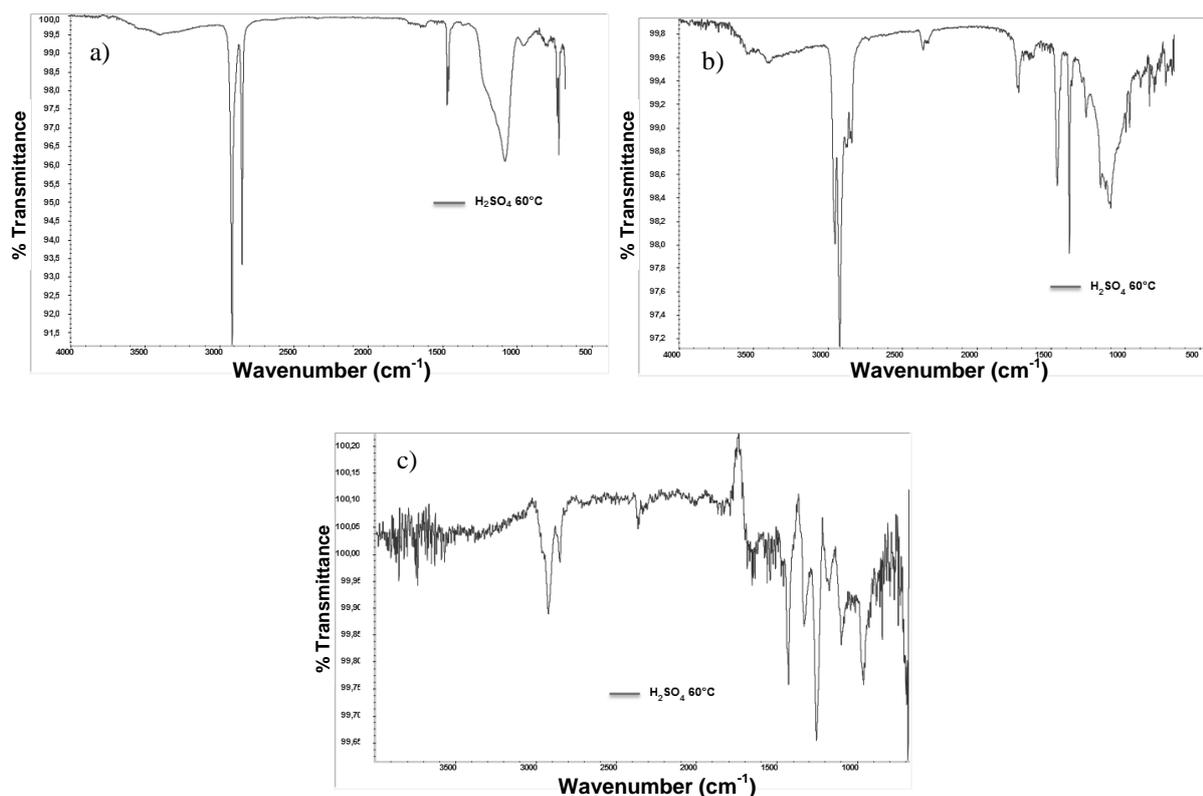

Fig. 8 FT-IR spectra at degraded state of: a) HDPE, b) PP, and c) PVC

For the HDPE aged for 83 days in H_2SO_4 at 60°C (fig.8.a), the appearance of a new spectrum at about 1100 cm^{-1} and the existence of the characteristic peaks of the aliphatic hydrocarbons function were observed. For the PP aged for 83 days in H_2SO_4 at 60°C (figure 8.b), the appearance of new peaks, which are characteristic of the carboxylic acid function, and the existence of characteristic peaks of the aliphatic hydrocarbons function were discovered. The characteristic peaks of the carboxylic function are: 1725-1700 cm^{-1} (C=O stretching), 1440-1395 cm^{-1} and 1320-1210 cm^{-1} (C-O stretching), and 950-900 cm^{-1} (OH deformation). For the PVC aged for 83 days in H_2SO_4 at 60°C (figure 8.c), the disappearance of the characteristic peaks of the hydrocarbons function was observed and new peaks characterizing new functions were not found.

These results show that the diffusion of solvent and the effect of time and temperature have an influence on the mechanical behavior and lead to chemical transformations. The modification and the appearance of new peaks, signify the oxidation of the hydrocarbon groups (CC, CH) [16,10], due to the attack of the acid and the effect of temperature and aging time.

According to J. Weon et al. [10], the main peaks of polyethylene are the C-H stretching (2900 cm^{-1}), the CH_2 deformation bending (1460 cm^{-1}) and the CH rocking bending (717 cm^{-1}). After thermal aging, a band around 1680-1780 cm^{-1} (C=O) originated from carboxyl functional groups appears. They also observed a peak at 3200-3400 cm^{-1} (O-H) derived from the hydroxyl functional group. This result show that the thermal aging for the LLDPE sample leads to local oxidation for the hydrocarbon groups (C-C, C-H). Once more according to P.Y. Le Gac et al. [16], in order to understand the chemical mechanisms involved in the polydicyclopentadiene (pDCPD) after immersion in hot seawater, FTIR measurements were performed and thereby they proved the appearance of two new characteristic bands situated at 1100 (C-O) and 1750 cm^{-1} (C=O). This is explained by the oxidation of the polymer. According to N. Nandakumar et al. [31], the exposition of Ethylene–propylene–diene terpolymers samples to aggressive 60% sulfuric acid, leads to important changes in the region of hydroxylated and carbonylated products and deformation vibrations. As a result of chemical degradation by acid, a new band appears in the spectra at 3200–3600 cm^{-1} due to the hydroxylated products and other changes in the chemical structure. These results show that the primary process involved in acid degradation is decrosslinking via hydrolysis.

According to Montes Castillo et al. [32], changes in the chemical structure of the polymer are observed by infrared spectroscopy. The apparition of hydroxyl (~3400 cm^{-1}) and carbonyl (~1700cm^{-1}) peaks signify a polymer oxidation. They also show that the crystalline component of polymer is assumed to be preserved during degradation; only the amorphous component is degraded, usually leading to an augmentation of the polymers crystallinity.

IV. CONCLUSION

The effects of acid solution aging at 60°C on the mechanical and structural behaviors of PVC, PP, and HDPE pipe specimens have been examined and lead to the following conclusions:

- The gravimetric measurements for PVC and HDPE during the testing time reveal that the water absorption follows the Fickian process. This is characterized firstly, by a phase of water absorption and weight gain, indicating the penetration of water into the amorphous parts of the material till saturation, and secondly by a relatively constant phase characterizing a "pseudo plate" of diffusion.

- The gravimetric measurements for PP reveal that the water absorption follows a non-Fickian behavior, characterized by a continuous absorption over time, which is usually accompanied by significant deformations and damage within the material.

- After aging in H_2SO_4 at 60°C, brittle behaviors for HDPE, PP and PVC pipe specimens were found (immediate break after aging) as a result of degradation.

- After aging in H_2SO_4 at 60°C, an increase in tensile strength and a decrease in elongation at break were found. The increase in mechanical properties is due to the increase in cristallinity and crosslinking density while the decrease in mechanical properties is due to the molecular chains scission.

- According to IFTR analyzes, the modification and appearance of new peaks signify the oxidation of the hydrocarbon groups (CC, CH), due to the attack by the acid and the effect of temperature and aging time.

These results show that the long term exposure to an aggressive solution and high temperature has a deleterious effect on the mechanical properties and leads to chemical transformations.

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