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Migration of Minerals and Organic Compounds between Bottled Water and its Plastic Packaging

Wafa Cheddadi[1]*, Yamina Berredjeme[1], Achraf Hamrouni[2], Zhour Hattab[3], Abdelhak Gheid[1], Raouia Hamrouni[4]

[1] Water and environment science and technology laboratory, Faculty of Science and Technology, Mohammed CherifMessadia University, Souk Ahras 41000, Algeria.
[2]Mines Metallurgy Materials Laboratory L3M National Hight school of Mines and Metallurgy, Amar Laskri ,B.P.233,W129,Sidi Amar, R.P. Annaba 23000 Algeria
[3]Laboratory of Water Treatment and Valorization of the Industrial Waste, Faculty of Sciences, Department of Chemistry, Badji-Mokhtar University, B.P.12, Annaba 23000, Algeria.
[4]LEREC, Department of physics, Badji-Mokhtar University, B.P.12, Annaba 23000, Algeria.

ABSTRACT

Several studies have shown the bottle's ingredients have been transported in water, this study aims to investigate experimentally the migration of several minerals from water to PET bottles. A series of water bottles were examined after exposure to extreme conditions of temperature(30, 60 and 90 days)(25, 35 and 45°C).The (AAS) to determine the concentration of Ca, Mg, Na, k, Pb after 90 days showed a significant decrease and increased in the amount of minerals water .the (TOC) concentration increases in mineral water when the temperature increases ,this is confirmed by the FTIR analysis, proving the existence of minerals compounds from the water to PET,and proven by analysis with (XRD).the study proves that the minerals have been successfully implanted into the PET surface, as the (WLI), and hence this result suggests the improvement of the hydrophilic property of the modified PET.

Keywords: Polymer, PET, Mineral Water, Pollution, Packaging.

Introduction

Packaging materials in contact with food play an unavoidable role in the conservation and protection of foodstuffs. Also, they appeared a constant innovation like the development of active and intelligent materials. The world consumption of bottled water is in constant evolution due to the increased need for consumers to have access to the safe drinking water of an efficient chemical and microbiological quality.In recent years, the packaged water sector in Algeria has experienced exceptional development thanks to investment encouragement. This development results in the establishment of dozens of exploitation and production units of packaged waters throughout the national territory. Also, it was accompanied by an exceptional increase in the consumption, whose part per capita has remarkably evolved in twenty years (4 L/capita/year in 1989 to 22 L/capita/year in 2007). In fact, thermoplastic packaging has grown considerably in popularity since the beginning of the 1990s, and has become part of everyday life. Interestingly, polyethylene terephthalate (PET) is a family of polyesters, a semi-crystalline plastic polymer **(**C. Bach et al.,2012), and has gradually replaced other materials, such as polyvinyl chloride and glass in the packaging of mineral water and beverages (Layal al Rayes et al.,2012). Owed to its chemical inertness and physical properties, including transparency, strength, malleability,

impermeability to gases, liquids and microorganisms, recyclability, lightness, and often lower cost, it seems to be basically suitable for food packaging applications, especially in beverages and drinking water. Further, PET is industrially synthesized by either two ways: the transesterification of dimethyl terephthalate with ethylene glycol followed by polycondensation(Konkol.2004) or the esterification of terephthalic acid and ethylene glycol, and noteworthy, the polycondensation step is common to both synthesis pathways. In addition, PET can be catalyzed by metal salts widely-used in the packaging in the food industry, and specifically for water packaging, including natural mineral waters and spring waters whose exploitation and protection are well-described in the National Executive DecreeN° 04-196, 2004. Hence, natural mineral waters are defined as microbiologically healthy waters and obviously distinguished from other waters intended for human consumption by their natural properties, including purity, and typical content of mineral salts, and trace elements or other constituents. These properties are assessed on geological, hydrogeological, physical, chemical, physicochemical, microbiological, and pharmacological levels, making these natural mineral waters therapeutically effective to human health. The qualification of these two types of waters established in terms of this decree is alike to that provided by the guideline text related to the natural mineral waters and spring waters. Whilst, several studies have shown that food contains substances whose origin is controversial, and this refers to a probable hypothesis, indicating that their presence is due to the migration of PET constituents such as monomers, additives and unintentionally added substances (UAS) to bottled water (Fabrizio et al.,2018).Further, the deterioration of the packaging and/or its contents during aging leads to a quality loss, product returns, and consequently to a decrease in the company's brand image. Several types of interactions due to physicochemical mechanisms occurred between a package (container) and the packaged product (contents) are shown in fig.1 (Isabelle. S et al.,2011). The inertia of packaging is rarely total since this polymer is not inert and may exchange and migrate between the plastic and the food matrix, i.e. transfer to the food of constituents of the packaging material (additives, monomers, neoformed compounds ...) (Westerhoff et al.,2008).Moreover, the interaction between the container and the contents can lead to material transfers, causing, for example, an alteration of the organoleptic properties of the food, and as a result, a toxicological problem may arise (Djelloul et al.,1994). This contact container/content can also affect the packaging mechanical properties (Konkol.2004).

• **Sorption phenomena** for which constituents of the food can be absorbed by the packaging, which on the one hand harms the quality of the food and, on the other hand, can lead to packaging deterioration.

• **Permeation processes** of which the sorption is overall, the first step characterizing the transfer of volatile substances through the material; there may have either loss of aroma from the food or food contamination by substances originating from the environment (gases, odours) or from the external packaging wall (such as inks located on the surface of the packaging) which can have negative effects both toxicologically and organoleptically(Isabelle.S et al., 2011).

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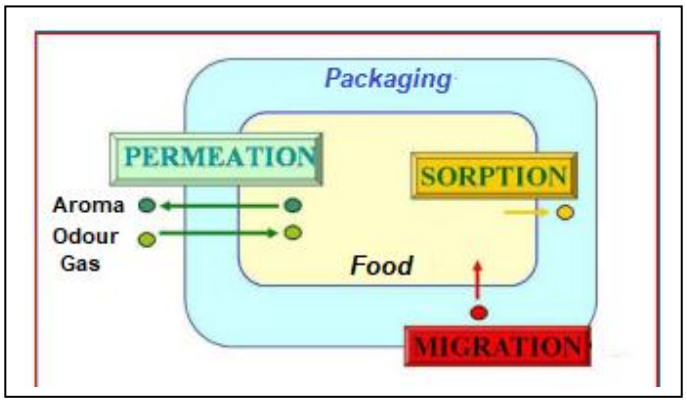

Fig1. Packaging/food material transfers.

These exchanges are affected by several factors, including the storage time and temperature, the concentration of the migrant in the polymer, the type and nature of the migrant and the drink, and the solubility of the migrant in the liquid (Rungchang et al., 2013), and whose activities can toxicologically and organoleptically affect the food health and quality, and the packaging properties. Nevertheless, the manufacturing process or unsafe storage conditions results in the appearance of some contaminants in bottled water. In Algeria, the consumption of mineral water and natural springs in PET bottles Algeria has increased considerably in recent years, and hence the no less than 40 brands on the market results in incentives and support measures for investors in this field. According to an investigation conducted by the Association of Algerian Beverage Producers (AABP), the water consumption has increased from 4 L/capita in1990 to 23.4 L/capita in2011,and although of this development, the lack of legislation on the use of plastic packaging for food contact has led to excessive use of plastic packaging during production and storage, and consequently threaten the consumer health. Therefore, the current legislation must guarantee the protection of public health and, ensure the consumer interests with regard to the marketing launch of materials and articles intended to be in contact with food. As previously reported (Yingying et al., 2014,StefanieMaaß et al.,2017),PET is not inert and shows amass transfer with water or soil. Additives containing impurities, which may infiltrate the packaging of chemical mixtures with oestrogenic activity have been found in PET bottled mineral water (Yang et al.,2011). Furthermore, the investigation of the migration of plastic packaging to foodstuffs is one of the most important research interests in the field of food safety. Recently, the specific migration of antimony in PET bottled water for all the studied bottles was reported to do not exceed the value required by European and world legislation for different investigation temperatures: 6°C, 25 °C and 40 ° C and for a duration ranging from 6 hours to 365 days of use. The antimony content is higher in smaller bottles because it depends on the contact surface related to the water volume, and of note, the diffusion coefficients were determined for temperatures between 5 and 40°C (B..Zmit et al .,2019).Some inorganic species may be present as residues of catalysts or additives used to produce PET. Antimony is considered the most important inorganic compound that can migrate from the PET bottle to beverages. Regulation N° [10/2011] sets the specific migration limit (SML) for the antimony of 0.04mg.kg^{-1}, and the Environmental Protection Agency(EPA) sets an AML in drinking water of 6 µg.L-1 .Further, the migration of the antimony (Sb) from PET into water increases rapidly during the first storage

period and then slows down to a limit concentration (Keresztes et al., 2009). The addition of plasticizers (phthalates and adipates) to plastic packaging is widely used to improve the flexibility, workability and durability of polymers, particularly in polyvinyl chloride (PVC)(Djelloul et al.,1994,Fierens et al., 2012).In addition, phthalates in food contact materials are subjected to strict regulations and do not seem to be used in the manufacture of PET bottles. As reported (C. Bach et al., 2012), the migration potential of stabilizers is not remarkable due to their insolubility in water. and therefore, some research work focuses on the study of the existence of more volatile, saturated and unsaturated carbonyl compounds responsible for the bad taste of water bottles and humans death(LoPachin et al., 2014).These studies focused on the release of initial PET reagents (monomers and catalysts), reaction by-products, and plastic additives into bottled water. The properties of chemical additives present in plastics, including migration, release, fate and environmental impact during use, disposal and recycling have been well-documented (JN Hahladakis et al.,2018). Also, other study has reported the presence of benzene, toluene, ethylbenzene, styrene (BTES), formaldehyde, acetaldehyde and benzaldehyde in Lebanese polyethylene terephthalate (PET) bottled water (Layal al Rayes et al.,2012).Moreover, Algeria is a country whose climate differs between the north and the south (Sahara), but it faces incredible environmental contamination by various hazardous contaminants threatening human health. Thus, we assume the necessity to acquire experimental data and to share our results on this totally unknown problem in Algeria with the various actors (authorities, consumers) and the scientific community. Here, we would focus on the study of the possible contamination caused by plastic packaging used to package bottled water marketed in Algeria. Several studies have shown that some of the bottle's ingredients have been transported in water, but the discussion of whether this water could lose some of its basic mineral components or not remains unanswered. Thus, the objective of this research work is to assess the material transfer phenomena (migration/absorption) of the constituents (monomers, catalysts, impurities, additives, and neo-forms) from the packaging to the water, with respect to the following quantitative, qualitative and sanitary point of view:

- The characteristics of bottled water,
- The physicochemical properties and structure of the polymer, making up the bottles.
- The storage conditions of the bottles (time, temperature, and sun exposure),
- The correlation between the physical properties of water packaging and the presence of certain compounds in bottled water.

The present study was aimed to experimentally investigate the transfer of several minerals to (PET) bottles. Herein, the purchased samples of two types of water (mineral water and spring water) were stored in two different types of bottles (polyethylene terephthalate (PET) and glass), and exposed to full sun for 30, 60 and 90 days, and T°C 25, 35 and 45 ° C. A Shimadzu (Germany) atomic absorption spectrophotometer (AA 6300G) was used to determine mineral concentrations in the mineral water, and a diffractometer (XRD) was used to detect the presence of minerals in the bottles when exposed to water. In the second part of the study, a concentration of organic matter was found in the mineral water using (TOC-LCSH / TOC-LCSN (hereafter referred to as TOC-L) total organic carbon (TOC), white light interferometry (WLI) and Fourier transform infrared analysis (FTIR) showed a change in the composition of the topography of a bottle surface.

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3. Results and Discussion

3.1. White Light Microscope Interferometer FRT (WLI)

Fig.2 illustrates the surface topology variation of a PET bottle filled with water during 90. Here, various bottle parts (neck, body and feet of the Type A bottle) were subjected to White Light Microscopic Interferometer (WLI) examination. A difference in the shape of the surfaces can be seen when changing the sampling location and, also the presence of new deposit components on the surface, leading to its thickening. This is likely due to the transfer of minerals from the water to the PET bottle. Further, a topography of peak-coherence interferometric surface was designed and implemented for surface profiling. In this study, we noticed slight discrepancies in the results of the prototype interferometer as evidenced by the validation process, while the overall surface mapping is satisfactory and can be improved by the suggested items from the observations. Once the initial configuration is completed, the measurement of a surface and its processing becomes simple, even if the processing is not optimized for time. A non-homogeneous physical property, the dispersion of water in the different PET and PET/water–polymeric matrix has been observed by WLI. The observations carried out on the different nanocomposites(Abbas Ghanbari.2012) revealed a set of isolated folds distributed homogeneously in the nanocomposites and with, however, the existence of small stacks for the other part of the polymer. The folds seem to be aligned in a privileged direction. Another consequence of the penetration of solvent into the polymer is the swelling, damaging the internal structure of the polymer, which implies that the sorption process of other food constituents is facilitated (Konkol. 2004).

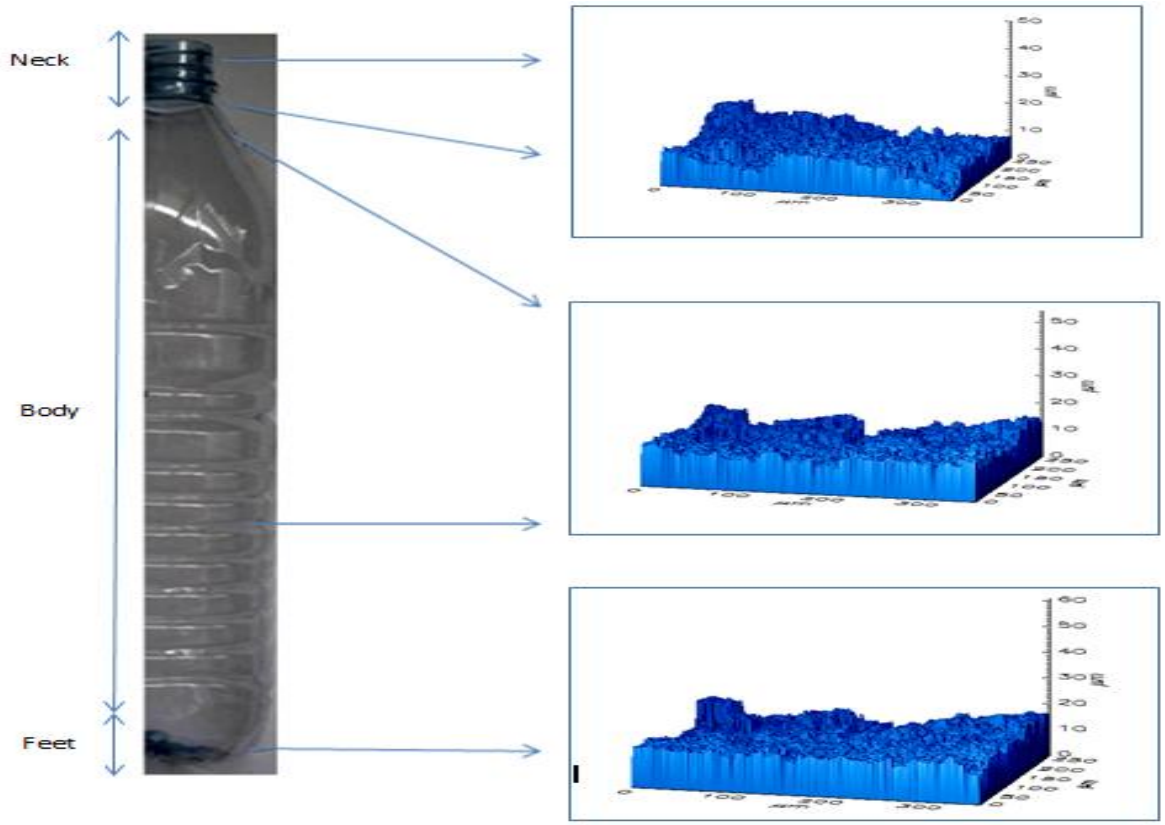

Fig.2. White light interferometer (WLI) of the surface PET bottle filled with water for 90 Days at 25°C.

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3.2. Atomic Absorption Spectrophotometer (AAS)

3.2.1 Effects of storage time and temperature on the transfer characteristics between the polyethylene terephthalate (PET) bottle and water.

Fig.3 illustrates the evolution of the transient concentration at various temperatures. We note that as time increases, the value of the mineral concentration decreases. The phenomenon is based on the equilibrium process between the mass transfer of minerals from the water sample to the adsorbent (PET bottles). A contact time between the adsorbent and the water reveals the following element concentrations obtained at 25°C : [Ca] 67.12 mg/L, 16.19 mg/L [Mg], 17.6mg/L [Na] and 0.08 mg/L [K] for 60 days, and 0.17 mg/L of [Pb](Krishan et al. ,2017, Shweta et al. ,2019). It can be seen that as the temperature value highly increases, the mineral concentration highly decreases, and hence, after 90 days the [Ca] value of 67.01 mg/L at 25°C is gradually reduced to 50.20 mg/L at 35°C and 25.06 mg/L at 45°C (Natarianto et al., 2019). This result can be explained by the transfer of minerals to the PET bottle, while the water showed the presence of lead (Pb) whose concentration is calculated to prove the source of migration from the packaged polymer. Hence, the presence of Pb can be explained by reverse migration (PET to water), where Pb is one of the additives used in the manufacture of PET (C. Bach et al., 2012).
Fig.3 depicts the effect of time and temperature on the concentration of water-dissolved minerals for PET bottles of type (A). The following results can be highlighted:

- The effect of the time parameter on the transport of the mineral elements (Ca, Mg, Na and K) is greater at a temperature between 45 and 35 °C than that at 25 °C (Fig.3 A).
- The water concentration of each element (Ca, Mg Na and K) decreases during the same time period with increasing temperature.
- After 90 days, the Ca value is reduced by 67.1, 50.20 and 25.06 mg/L at 25, 35 and 45 °C respectively.
- The concentration of Pb released from the PET bottle increases with increasing time period and temperature.
- Lead (Pb) release increased from 0.1 mg/L at 25 °C to 0.17 mg/L at 35 °C, and 0.29 mg/L at 45 °C after 4 weeks of storage, from 0.16 mg/L at 25°C to 0.18 mg/L at 35°C and 0.31 mg/L at 45°C after 8 weeks of storage, and from 0.17, 0.21 and 0.35 mg/L at 25, 35, and 45 °C respectively, after 12 weeks of storage.

In this work, the absence of bacteria was found in the tested waters, and as a result, the possibility of biological decomposition was ruled out due to the fact that sun exposure destroys the microorganisms present in the water. The SODIS method (Solar Water Desinfection) is a process based on the disinfection of water by solar irradiation to obtain drinking water free of infectious germs (C. Bach. et al., 2012).Furthermore, the element found in water bottled in PET and glass has been identified as exogenous, and this proves that lead (Pb) is a metal absent in glass-bottled water. However, PET- bottled water (brand A and C) is remarkable because it contains Pb at levels ranging from 0.13 to 0.29 mg/L. This metal is therefore derived from PET packaging, but does not increase after 90 days at different exposure temperatures (25, 35 and 45°C).As expected, Pb is characteristic of PET- bottled water, and whose content evolves with different exposure temperatures, i.e. its presence is not due to the geology of the soil, but it is effective and well made from the manufacture of PET (J.N. Hahladakis et al., 2018).In addition, all Pb levels detected in this study do not exceed the regulatory threshold of 40 mg/kg for the regulation 10/2011.

3.2.2 Effect of bottle type on mineral degradation at 45°C.

As shown in Fig.3(A6), a big difference between the two types of the manufactured bottles (PET, glass). The obtained results were compared at a temperature of 45°C.

- At the time of contact between adsorbent and water, the decrease is 6.08 mg/L Ca in the PET bottle and 0.99 mg/L Ca in the glass bottle for 60 days.
- The decrease in Ca concentration is greater as the contact time increases for the PET bottle, since the Ca concentration becomes constant in the glass bottle.
- Conclusively, the glass has better characteristics for water conservation (reducing the effect of material transfer). This result concord with the published research literature (Layegh et al., 2019).

3.2.3. Effect of water type on mineral degradation at 35°C

The comparison of the evolution of mineral and Pb concentration for two types of water (mineral water and spring water) as a function of time is shown in Fig.3(A5). Several physicochemical parameters play a key role during the process of mineral transport in water.

- Pb release increases from 0.22 mg/L to 0.27 mg/L and 0.29 mg/L for 4 weeks, 8 weeks and 12 weeks respectively, in mineral water at 35°C.
- Pb release increases from 0.17 to 0.18 and 0.21 mg/L for 4, 8 and 12 weeks respectively, in spring water at 35°C.

As can be deduced from fig.3, the rate of degradation increases when the concentration of minerals increases, and this is evidenced by the Ca values. This result can be explained by the presence of diffusion phenomena (Pauline et al.,2018). Monomers, known as stabilizers and technological adjuvants are usually lipophilic molecules, and even though their possible diffusion in aqueous foods, they migrate more in fatty foods. In terms of interaction, there is a possibility of penetration of the contact medium into the polymer packaging depending on their affinity, for example fat in polyolefins or water in PET.

3.3. Total Organic Carbon (TOC)

The total organic carbon (TOC) content is a key parameter in controlling the stability of bottled water. Fig.3 (B1-B2) illustrating the impact of sunlight and temperature on PET- and glass-bottled water shows that the TOC concentration gradually increases as the temperature rises. Also, TOC is found in type A and C bottled water in considerable quantities, but a small amount and just traces are detected in the type B bottle. This result is likely explained by the migration of TOC from PET bottles to mineral water, as confirmed by infrared absorption analysis. However, the presence of the same compound in the glass-bottled water suggests that the origin of this substance may also be related to the (polyethylene) stopper of the bottle (Romão et al., 2009).Importantly, an equilibrium of chemical potential of PET packaging and water as far as organic matter is concerned. The impact of sunlight and temperature on PET-and glass-bottled water was assessed using the Total Organic Carbon (TOC) analysis method. In a first approach, only bottled water that has undergone the maximum conditions of sun exposure (90 days of contact with a temperature variation of 25 to 45°C) was analysed. This approach is based on the hypothesis that under these conditions, the migration of the elements would be more important due to a temperature close to the glass transition temperature (Tg) of PET. This introduction of molecules into all the macromolecular chains leads to a decrease in the Tg value of the polymer

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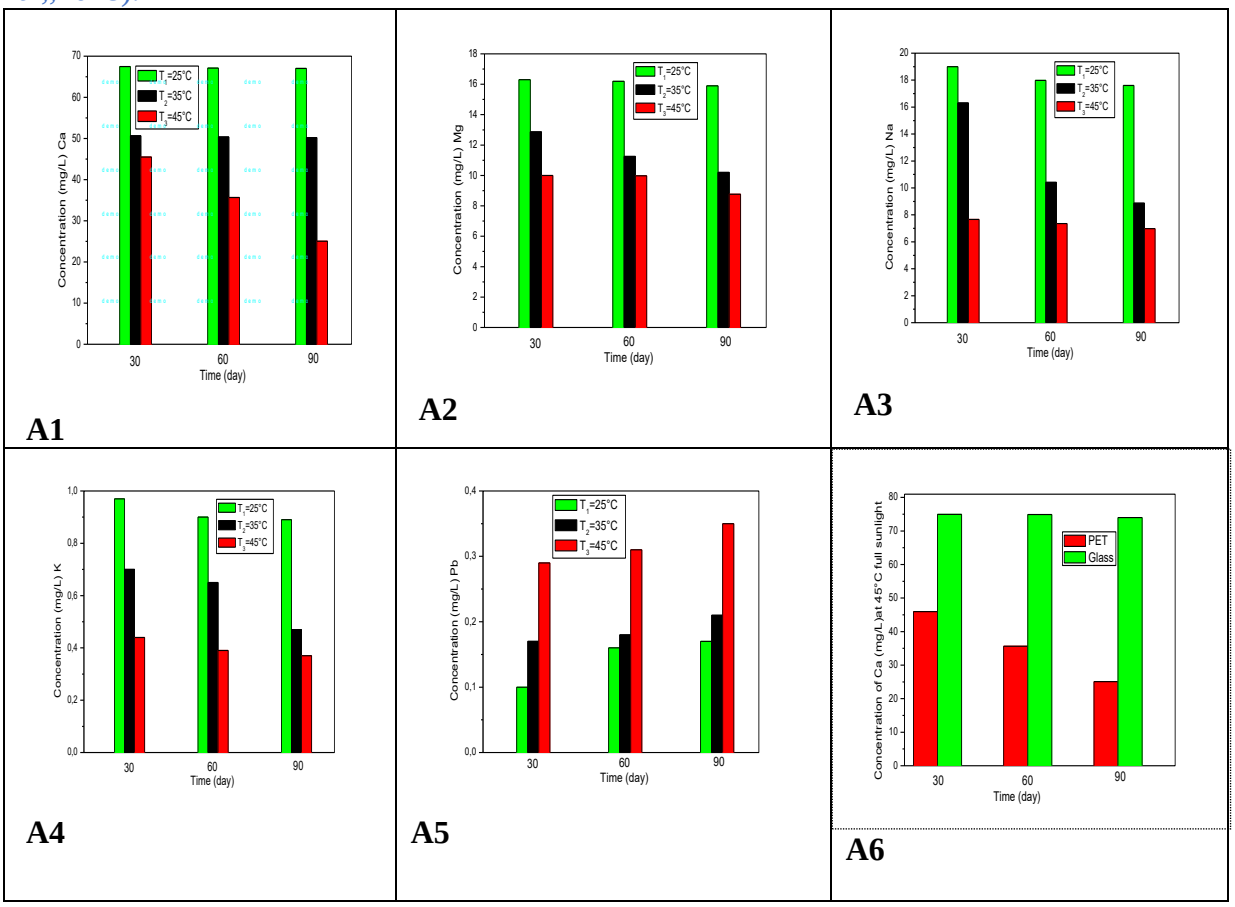
and accelerates the diffusion of organic macromolecules from PET to water (Zaki.2008).Moreover, during storage and transport of the bottles under these extreme exposure conditions, TOC was found in the bottled water of group A and C with considerable quantity, but its small amounts and traces were observed in the bottle of group B. Whilst, the concentrations in the water of the other molecules previously detected during the qualitative analysis were proven according to the impact of temperature, and that the origin of this compound is related to the packaging material. Nevertheless, the presence of this same compound in glass-bottled water suggests that the origin of this substance may also be related to the material of the bottle's corks. Of note, group A and B bottles are sealed with polyethylene (PET) caps, and the glass bottle caps consist of an aluminum (Al) cap with a plastic seal. Only carbonyl compounds were detected in the PET bottled water of both brands (A and C), although the impact of sun exposure. The absence of these compounds in the glass-bottled water during both types of exposure confirms that the occurrence of carbonyl compounds is related to the PET material. In this regard, several authors have reported that both substances are generated by thermal and oxidative degradation of the PET material (Romão et al., 2009).In comparison to the literature, the results of this study regarding the migration of acetaldehyde in mineral water after 10 days at 40°C are in good agreement with those previously reported (Ceretti et al., 2010). Additionally, the effect of water typology on the migration of these two molecules by the action of sunlight may explain the absence of carbonyl compounds. On the one hand, reported that some mineral waters contain heterotrophic bacteria using formaldehyde and acetaldehyde as a carbon source (Khaldi et al,,2018).



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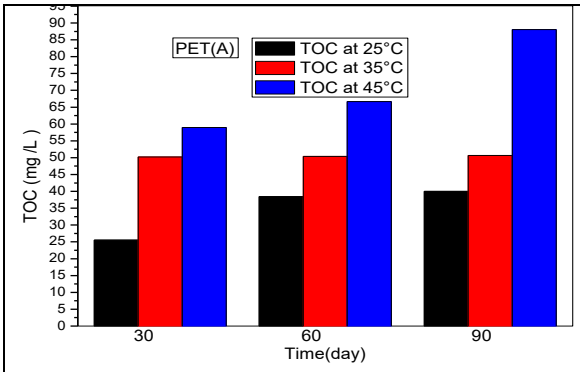
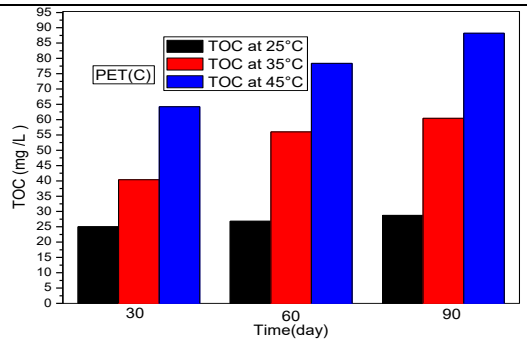


B1



B2

Fig.3 Effect of the temperature and time on the transient concentration for PET bottle type (A): **A-** of the minerals (Ca, Mg, Na ,K and Pb) in mineral water. **A6**-Transient concentration of the calcium of mineral water in PET bottle and glass bottle at 45 ° C. **B**-Transient concentration of total organic carbon in PET- and glass-bottled mineral water at a different time.

3.4 Fourier Transform Infrared Spectrum (FTIR) analysis

Fourier Transform Infrared Spectroscopy (FTIR) is a widely used analysis method to study the nature of chemical bonds in polymers and their structural state by characterizing vibrational modes. We can easily monitor the chemical and morphological transformations of polymers under different stresses (M. Djebara et al.,2012). The infrared spectrum is formed as a result of the absorption of electromagnetic radiation at frequencies corresponding to the vibration of specific sets of chemical bonds within a molecule .As depicted in Figure 8 showing the FTIR spectra of plastic PET bottles, the characteristic bands of the FTIR spectrum show that plastics are made from PET. Further, the absorption bands Fig.4 at 3100-2800 cm-1 have been assigned to an aromatic and aliphatic -C - H stretch bond, 1720 cm^{-1} to the carbonyl ester stretch bond, 1300 cm^{-1} to the ester stretch group and 1100 cm^{-1} to the methylene group. Most bands differ depending on that the PET sample was extruded and quenched, heat-treated and then drawn, i.e. whether the sample is amorphous and oriented, or crystalline and drawn. The differences are due to the configuration of the ethylene glycol group and also phenylene carbonyl bonds (cis / trans conformers) (Ziyu et al.,2012).Importantly, chemical additives present in trace amounts and related to the manufacture of the polymer or vial are not observed, but these traces and degradation products could migrate from the bottle to the water. Thus, analytical methods enabling the detection of these substances in water with low detection limits have been developed. To achieve this goal, a screening analytical method has been designed, as well as the separation and identification of these compounds are in progress (JN Hahladakis et al.,2018).The FTIR spectrum analysis method for the bottle without water shows a wide band in the region of 1000 to 1500 cm^{-1}. This may be due to the additives present in the PET bottle Fig.4.Whilst, other absorption bands were detected at 3440 cm^{-1}(O-H, stretch), 2516, 2348 and 1796 cm^{-1}(C = O; calcite, $CaCO_3$), 1640 cm^{-1}(H-O-H, bond), 1423 cm^{-1}(C = O e OH, stretch), 1000 (Si-O.); silicate, C-S-H), 874 cm^{-1}(C = O, drawing, calcite, $CaCO_3$) ,776 cm^{-1}(Si-O-Si, symmetrical drawing), 713 cm^{-1}(CO_3, drawing), 678-648 cm^{-1}(SiO_4, drawing) and 583-414 cm^{-1}(Fe-O-Fe, drawing) (Ana Paula et al., 2017).

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Fig.4 shows the FTIR spectra of the A and C brand bottles respectively before and after natural exposure under controlled light conditions for 90 days. Before exposure to sunlight, the characteristic bands of PET at 1340 cm^{-1} and 1370 cm^{-1} are obviously observed from the experimental results. The presence of copolymers or additives related to the bottle manufacturing process was not detected. On the other hand, the decrease in the spectral bands' intensity between 2918 cm^{-1} and 2850 cm^{-1} related to the presence ofCH_2 groups in the polymer chain is observed by comparing the spectra before and after exposure of Brand A PET. A decrease in the intensity of peaks after natural irradiation would be due to a split in the polymer chain, causing a decrease in the concentration of these CH_2 groups (Naït.2008). In accelerated exposure, the spectra of PET from brand A and C bottles evolve and show the appearance of massive classical oxidation, more particularly heavy carbonyl products adsorbing at 1875 cm^{-1} and benzoic acids adsorbing around 1690 cm^{-1}. The increase in transmittance may be due to the leaching of compounds (organic carbon) from the plastic bottled water.

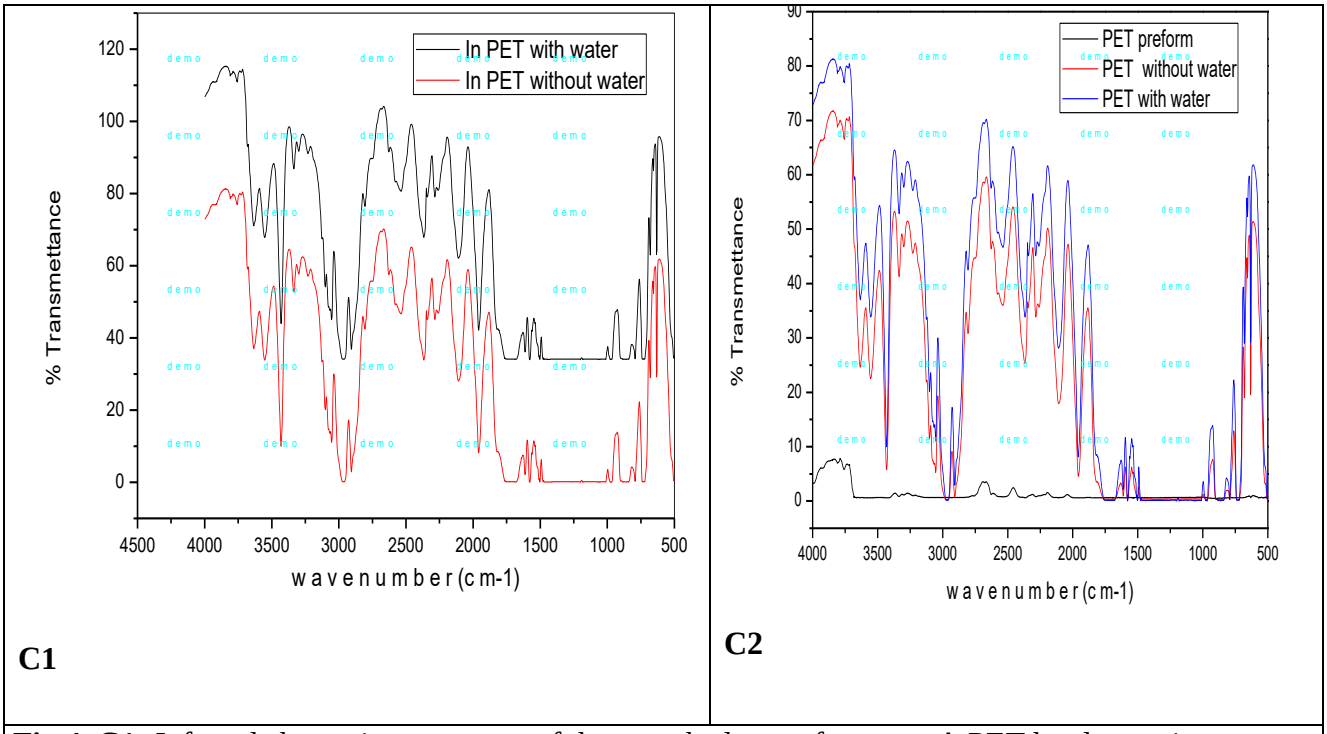

Fig.4 C1- Infrared absorption spectrum of the stretched part of a group A PET bottle part in sun exposure and part in contact with water.**C2-** Infrared absorption spectrum of the stretched part of a PET preform, a PET bottle without water, and a PET bottle of group A part in contact with water.

3.5. X-ray diffraction analysis

The polyethylene terephthalate (PET), a polymer used in beverage packaging for the bottles production by injection stretch blow molding is defined as polyester with an amorphous or semi-crystalline structure, based on the processing history information. Of note, the crystallinity of PET can be measured by various methods , including X-ray diffraction (common technique for crystallinity measurement), providing microstructural details on the tested sample, such as crystal size and orientation .In the present study, the X-ray diffraction patterns were collected

from the PET bottles of brand A and C under certain experimental conditions (contact time, ordinary temperature and selected temperature, exposure to sunlight).The different diffraction peaks can be observed in Fig.5showing a wide band and, also, good diffraction peaks at 2 θ = 11° and 12° as reported elsewhere(H. Liu et al.,2018), 2 θ = 8.9° and 19.7°, indicating the coexistence of amorphous and crystalline regions in the structure, while the broadest peaks of the order 2 θ = 12-34° are attributed to amorphous PET (G. Galo Silva et al.,2019,Z. Zhang et al .,2019,Y. Zhang et al.,2020).A previous study(AirongXuet al.2020) has suggested that the crystallinity of Methylcellulose(MC) and PET is reduced after hybridization of MC with PET. Furthermore, in relation to the pure MC and PET, the values 2 θ of MC and PET in MC / PET films remain invariable, suggesting that MC interacts little and/or weakly with PET, in addition that a PET spectrum represents anamorphous structure as reported by (V. Shabafrooz ,and al.2018) . (Johra FT and al .2014)who have found the graphene peak (002) was observed at 2θ= 26.54°, indicating the presence of platelets (Johra FT and al .2014).The intensity of the peak increased with the increase of the GrapheneNanoplatelets (GNP) Concentration, and here, the nanocomposite draw-barsexhibit a wide-hump around 2 θ = 19°. As indicated by the XRD analysis, the GNPs exhibit an orientation along the axial direction of the PET tensile stress. This orientation of the GNPs more effectively, reinforced the PET matrix by providing superior mechanical properties compared to a random orientation distribution(V. Shabafrooz ,and al.2018)of the typical XRD spectra of PET samples before and after Ag+ implantation. As well, the XRD model of the implanted silver ion PET shows a significant difference from the control PET. The diffraction peaks at 2θ=38.1°, 64.7° and 77.5° correspond to the crystallinity of Ag_2CO_3, Ag_2O. This XRD analysis confirms that silver has been implanted on the surface of the PET (J.X. Li, and al.2007).The XRD units in all composite samples showed marked peaks that the huntite-hydromagnesite particles were homogeneously dispersed and buried within the matrix polymer (E.Basturk, and al.2015).As expected, the presence of crystalline structures in the degraded polymer may cause the appearance of a white powder-like substance on the surface of the degraded transparency sheet. These values (2θ) were matched with the XRD of published phthalate-related compounds, since the XRD of the degraded polyethylene terephthalate revealed the presence of three types of phthalate derivatives(C. Sharon, and al. 2012).Furthermore, the crystallinity or more packing in the mainline of our commercial chitosan has been modified using a deacetylation process. In this case, the first reflection is associated with two different types of crystals. According to (Hwang KT, and al.2003).A number of crystalline peaks were observed for pure PET (without water), which can be attributed to the semi-crystalline structure of this polymer, meanwhile, only a small visible difference was observed between the model PET XRDs with the appearance of the following molecules: SiO_2quarte, Green cinnabar (Cr_2O_3), sucrose ($C_{12}H_{22}O_{11}$)Fig.5 D1, since the XRDs of the PET bottle of Group A shows the presence of Calcite ($CaCO_3$), Bornite (Cu_5FeS_4),Sodium Erbium $NaErF_4$ Fig.5 D2,and the XRD of the PET bottle of Group C reveals the presence of Halite NaCl ,Fluorite (CaF_2), Calcite ($CaCO_3$)shown in detail in Fig.5 D3. Additionally, the X-ray diffraction models shows that the spectra are quite similar and resemble to the XRD PET model, showing the difference in the additives and stabilizers that have entered in the PET manufacturing process according to very precise conditions. The amount of biopolymer reduces the intensity of diffraction peaks, characterizing the spectra as semi-crystalline phases.

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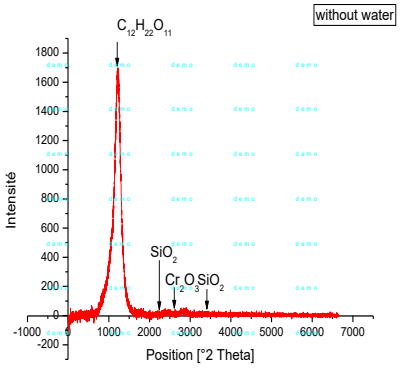
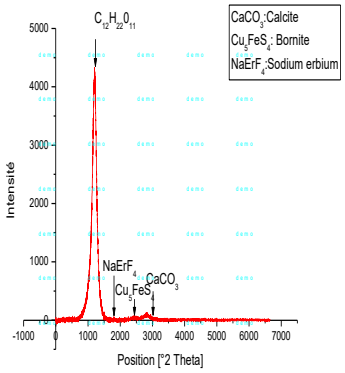
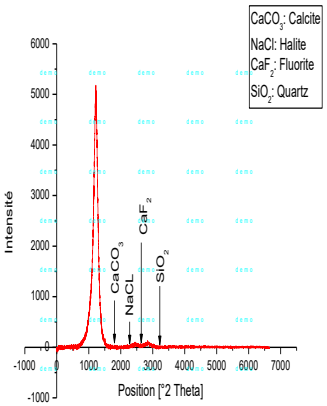


D1



D2



D3

Fig.5 D1- X-ray diffractogram of PET bottled without water,**D2-** X-ray diffractogram of PET bottled water from group A, **D 3-** Optical RX diffractogram for the PET bottle with group C

4. Conclusion

The present work has highlighted the migration of minerals in various bottled waters subjected to different temperatures and exposure durations, in addition to the organic compounds and minerals that were expected to be detected in PET bottled water. It was shown also that high temperature (45°C) and long storage time (90 days) promote the migration process. The results of the white light interferometric and FTIR analyses showed a change in the composition of the topography of the bottle surface, showing a difference in the surfaces shape in the case when the sampling locations are changed. Further, the atomic absorption spectrophotometer analysis has evidenced the effects of storage time and temperature on the water transfer characteristics in PET bottles, and on the degradation of minerals of PET-bottled water. It has been shown that the effect of time parameter on the mineral elements (Na, K, Ca and Mg) transport is greater for temperatures of 45 and 35 °C than that for 25 °C. At the same time, the concentration of each element (Na, K, Ca and Mg) in water decreases with increasing temperature. After 90 days, the Ca value decreases significantly by 67.1, 50.20 and 25.06 mg/L at 25, 35 and 45 °C respectively, while the concentration of lead (Pb) in water increases slightly with temperature and time, and so increases from 0.17, 0.21 and 0.35 mg/L at 25, 35 and 45 °C respectively. This result can be explained by the transfer of minerals to the PET-bottles (water- PET), and the reverse transfer of Pb to water (PET - Water).Regarding the effect of bottle type on mineral degradation at 45°C in full sunlight, the decrease in Ca concentration was found to be greater when the contact time increases in the PET bottle, and constant in the glass bottles, and consequently, the glass bottle has better characteristics for conservation in accordance with the literature. At 35° C, as the concentration of minerals increases, the rate of degradation intensifies due to the phenomenon of diffusion, in addition, that the TOC concentration increases progressively in mineral water as the temperature rises. Moreover, the FTIR analysis obviously indicates the increase in transmission from the bottle to the water due to the leaching of compounds (Organic Carbon) from the plastic bottle to the water. Interestingly, X-ray diffraction (XRD) was used to characterize the surface structure and composition. The results indicated that the minerals have been successfully

implanted into the PET surface, suggesting that the hydrophilic property of the modified PET is enhanced.

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