*IOSR Journal of Applied Chemistry (IOSR-JAC)*
*e-ISSN: 2278-5736.Volume 15, Issue 4 Ser. I (April 2022), PP 20-29*
*www.iosrjournals.org*

# Comparative study of the evolution of copper, iron, lead and zinc levels in tap water after stagnation of the same duration in different pipes as a function of pH

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***Abstract***
*Tap water may have a quality far removed from that from the distribution service due to contamination by metallic trace elements. This is why this work focused on the study of the effect of pH on the evolution of the concentration of copper, iron, lead and zinc in tap water. Thus, three blocks of four types of pipe made of cast iron, galvanized steel, polyvinyl chloride (PVC) and Polypropylene (PPR) pipes and accessories are supplied with water of a composition similar to that of the public water supply of Niamey (Niger). The analyzes of the water samples were carried out by flame atomic absorption spectrometry. The study generally showed that the concentration of these metals is higher at the start of stagnation at a slightly acidic pH (5.11). While over a long period of water stagnation, the concentration of these metals is higher at slightly alkaline pH (9.11). At neutral pH (7.04) the concentration of these metals oscillates between extreme values (found at pH 5.11 and pH 9.11) throughout the duration of the experiment.*

***Keywords****: Tap water, metallic trace elements, stagnation, Niamey*

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Date of Submission: 06-04-2022 Date of Acceptance: 21-04-2022

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## I. Introduction

The water treated on leaving the factory and served by the municipal network only contains copper, iron, lead and zinc in traces. Their concentrations increase at the outlet of the tap when water stagnates in the service entrance and in the internal piping of buildings [1-3]. Tap water can then contain a much higher content because of the use of Cu, Fe, Pb and zinc materials in plumbing [4]. It thus constitutes a persistent source of exposure to heavy metals [5]. These concentrations are explained by high stagnation times (weekends, school holidays) conducive to galvanic corrosion of welds and brass containing these metals in the internal piping of buildings [6-9]. However, consumer exposure to these metallic trace elements through tap water remains poorly characterized due to the lack of concentration data reflecting exposure through tap water. Indeed, the concentrations of these metals in tap water vary according to the location of their source in relation to the tap, the stagnation time, the water consumption preceding the stagnation, the water temperature, the diameter and the pipe length. This work studies the evolution of the contents of copper, iron, lead and zinc in tap water after stagnation of the same duration in polyvinyl chloride (PVC), galvanized steel, polypropylene (PPR) and cast iron pipes depending on the pH.

## II. Materials And Methods

### 2.1. Reagents and solutions

The reagents used are of analytical quality. The solutions are prepared in distilled water. The solutions supplying the pipes are prepared from the following salts: $CaCO_3$, $KNO_3$, NaCl and $MgSO_4.7H_2O$. Sodium Chloride (NaCl) is from Merck/Indonesia. Nitric acid $HNO_3$ 65% from VWR/France is used to acidify standard solutions and samples. The calcium carbonate ($CaCO_3$, 98%) comes from Micheltronche. Potassium nitrate ($KNO_3$ 99.5-100%) comes from Prolabo/France. Magnesium sulfate heptahydrate ($MgSO_4.7H2O \geq 99.5\%$) is from VWR Chemicals/France. The hydrochloric acid solution (prepared from 35% HCl, VWR/France) 0.1 M was used to bring the pH back to 7.04 and to 5.11 which was initially at 9.11. The 1000 $mg.L^{-1}$ standard solutions for copper, iron, zinc and lead AAS come from VWR/France.

The pH 9.11 solution is prepared from the above salts. Then, the solutions of pH 7.04 and that of pH 5.11 are obtained from the solution of pH 9.11 by lowering its pH using a dilute solution of hydrochloric acid. The concentrations of the chemical parameters of the pH 7.11 solution represent on average the concentrations of these same parameters in the public water supply of Niamey at the time of work.
Following the analysis tests carried out by atomic absorption, no trace of Cu, Pb, Zn and Fe was detected in these solutions.

**2.2. Experimental procedure**

The assembly consists of three systems, consisting of four pipes each. The four pipes are made of polyvinyl chloride (PVC), galvanized steel, Polypropylene (PPR) and cast iron. The PVC pipe has a PVC elbow and tap. The PPR pipe has an elbow and a PPR valve. For the galvanized steel pipe and the cast iron pipe, each is equipped with a galvanized steel elbow and a brass tap.
The four pipes of system 1, 2 and 3 are supplied with the solution of pH 5.11; 7.04 and 9.11 respectively.

**2.3. Sampling**

To monitor the evolution of the concentration of Cu, Fe, Pb and Zn in the 4 types of pipes in the laboratory as a function of pH, new and sterile 60 mL polyethylene bottles were used to take the samples. Thus, two hundred and four (204) water samples were taken and analyzed by atomic absorption spectrometry as described by [10].

The determination of Cu and Fe levels in the laboratory is carried out in the water contained in each pipe after the following stagnation times: 4 hours, 8 hours, 12 hours, 16 hours, 20 hours and 24 hours (1 day), 2 days, 3 days, 4 days, 5 days, 6 days, 7 days (1 week), 2 weeks, 3 weeks, 4 weeks (28 days), 60 days and 90 days. Seventeen (17) samples were taken from each pipe.

## III. Results and discussion

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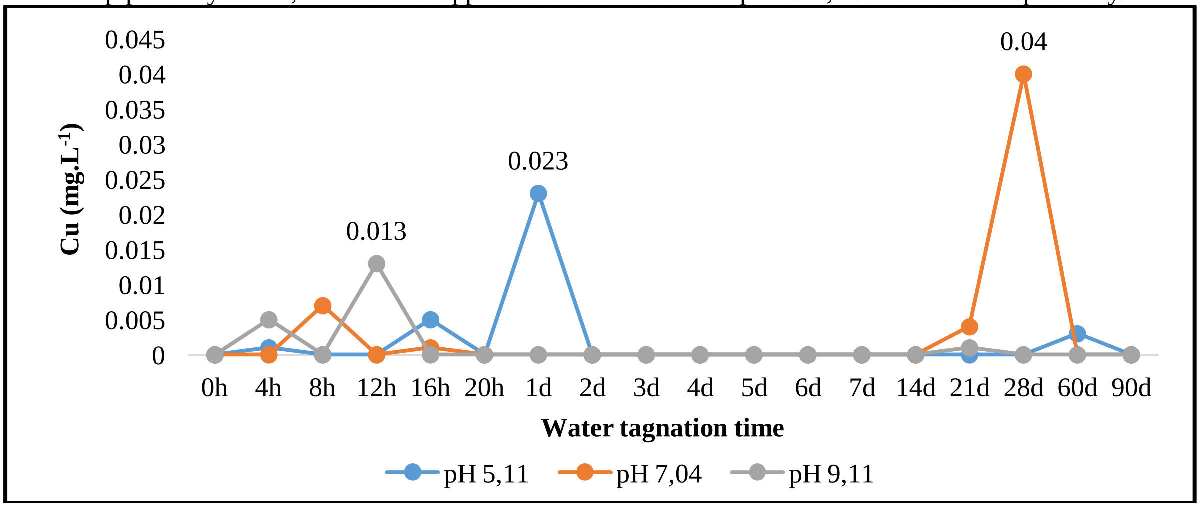
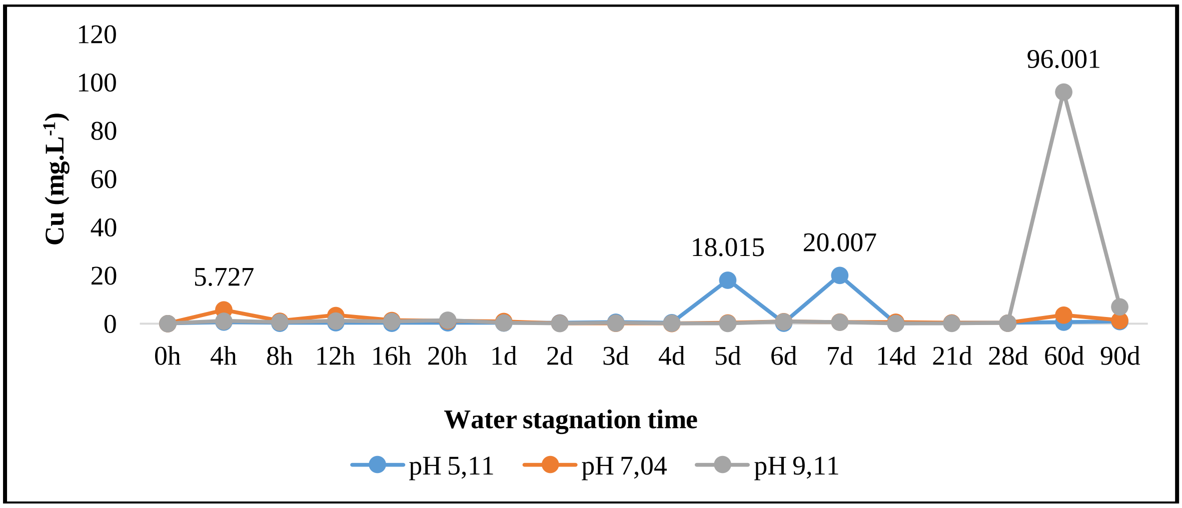



**Figure 1**: Effect of pH on the evolution of the copper concentration in the water remaining in the PVC pipe.

**3.1.2. galvanized steel pipe**

Figure 2 shows that the galvanized steel pipe releases more copper at pH 9.11 on the 60th day of stagnation. This concentration may be due to a detachment of the encrustations formed within the pipe. But at a more acidic pH, the concentration of copper is higher at the start of stagnation. At pH 7.04 the copper concentrations are the lowest over the entire stagnation period.

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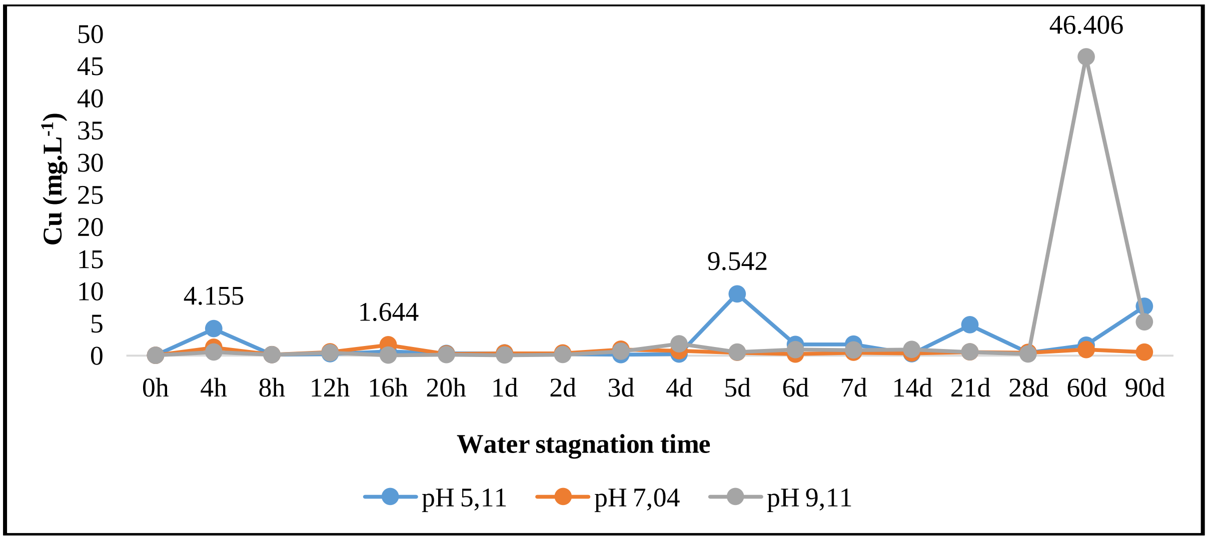


**Figure 3**: Effect of pH on the evolution of the copper concentration in the water remaining in the PPR pipe.

**3.1.4. cast iron pipe**

Figure 4 shows that the cast iron pipe releases more copper at the start of stagnation (from 4 to 20 h) whatever the pH (for the three pH values studied). Thus, the concentration of copper at pH 5.11 (5.517 $mg.L^{-1}$) is double that found at pH 7.04 (2.518 $mg.L^{-1}$) and four times that found at pH 9.11 (1.082 $mg\ .L^{-1}$) after 4 h of water stagnation.

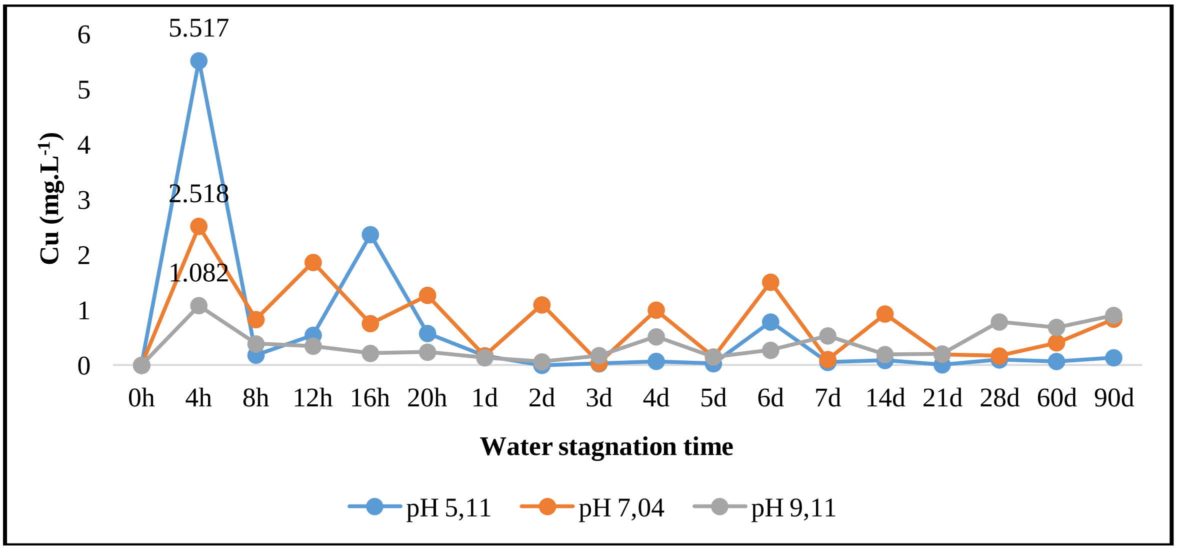


**Figure 4**: Effect of pH on the evolution of the copper concentration in the water remaining in the cast iron pipe.

### 3.2. Iron content

#### 3.2.1. Polyvinyl chloride pipe

Iron content is low in PVC pipe. 0.419 $mg.L^{-1}$ (at pH 9.11) is the highest concentration obtained. Figure 5 shows that iron is mainly released in solution during the first two days of water stagnation regardless of the pH (for the three pH values studied). For a prolonged period (> 2 days), the concentration of iron is practically zero apart from a rise in concentration of 0.241 $mg.L^{-1}$ observed on the 28th day of water stagnation. These results would mean that the iron leached at the start of stagnation is subsequently transformed into an insoluble product deposited on the wall of the pipe. The appearance of an iron concentration on the 28th day shows that the deposited iron can however come off at any time during the stagnation.

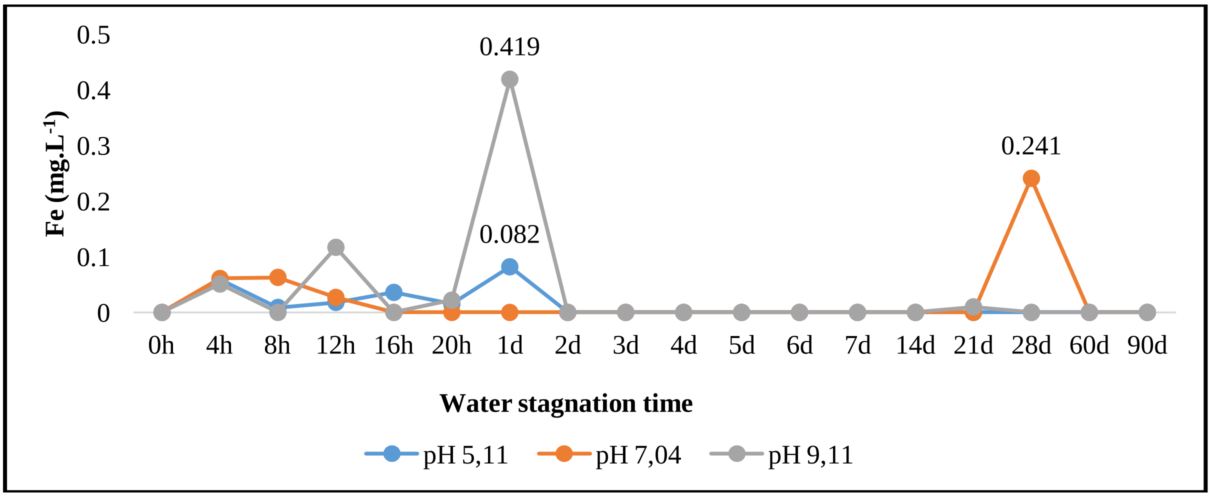


**Figure 5**: Effect of pH on the evolution of the iron concentration in the water remaining in the PVC pipe.

#### 3.2.2. galvanized steel pipe

Figure 6 shows the effect of pH on the variation of iron in standing water in the galvanized steel pipe. The results obtained show that whatever the pH (for the three pH values studied), the iron concentration is very low at the start of water stagnation (≤ 4 days). But, at the end of the 5th day, 6th day and 60th day of stagnation, the concentration of iron increases sharply respectively in a slightly acidic medium (pH 5.11), in a neutral medium (pH 7.04) and in a slightly alkaline medium (pH 9 ,11). The concentration of iron is higher in a slightly acidic medium (60.951 $mg.L^{-1}$ on the 6th day) and alkaline (55.639 $mg.L^{-1}$ on the 60th day) than in a neutral medium (47.625 $mg.L^{-1}$ on the 6th day ). Indeed, the galvanized steel pipe as its name suggests is a steel pipe covered with a layer of zinc. Thus, iron is not in direct contact with water in this condition, which is why its release into water requires prolonged contact (here 4 days at least).

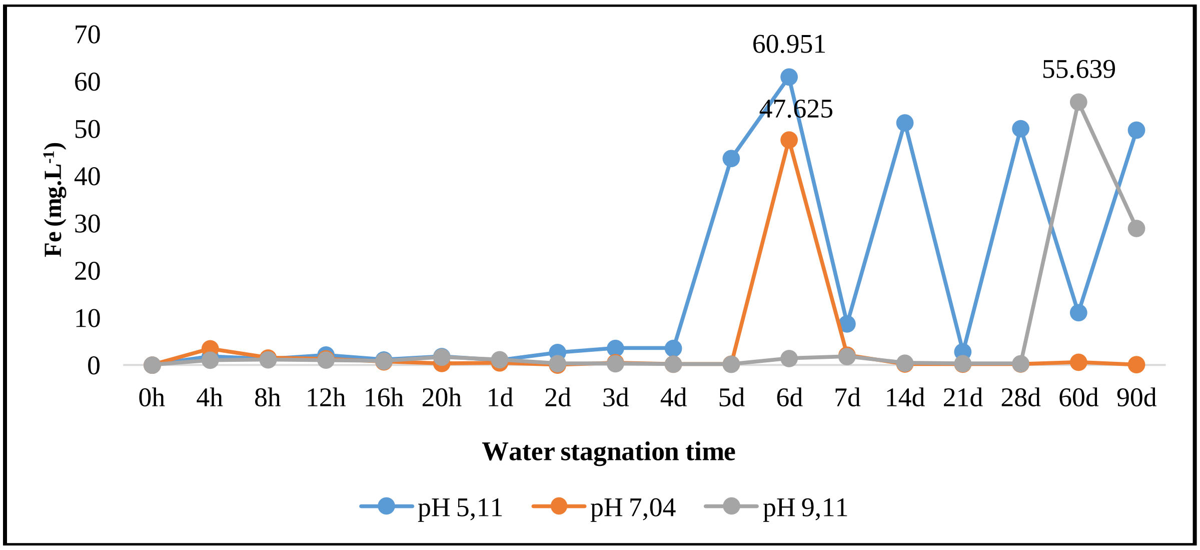


**Figure 6**: Effect of pH on the evolution of the iron concentration in the water remaining in the galvanized steel pipe.

### 3.2.3. Polypropylene pipe

Iron concentration is very low in stagnant water in PPR pipe. Figure 7 illustrates the effect of water pH on the change in iron concentration during water stagnation in the PPR pipe. Throughout the duration of the experiment, the concentration of iron is low regardless of the pH of the water (for the three pH values studied). The alkaline medium (2.242 mg.L$^{-1}$) has, after prolonged contact with the pipe, the highest concentration of iron than the acid medium (0.181 mg.L$^{-1}$) and neutral (0.175 mg.L$^{-1}$) . In addition, the concentration of iron is higher after 4 days of water stagnation. Thus, the presence of iron in the water in contact with the PPR water pipeline would be mainly due to the PPR gate valve which includes a metal part. This part would be made of an iron alloy covered by a layer of zinc like the galvanized steel pipe, because the stagnation of water on this metal part presents the same behavior (change in Fe concentration) as that observed for the TG.

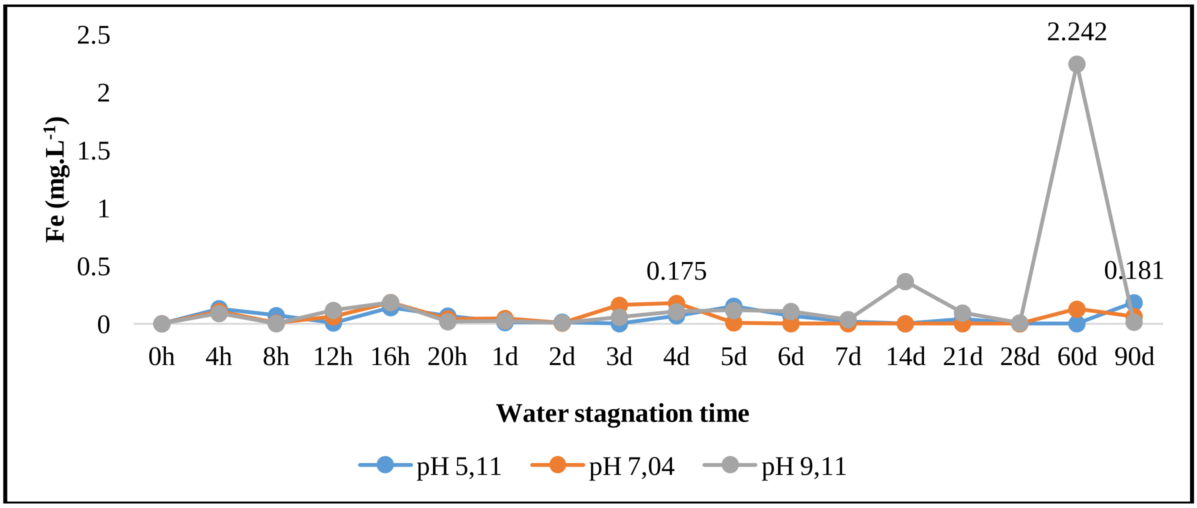


**Figure 7**: Effect of pH on the evolution of the iron concentration in the water remaining in the PPR pipe.

### 3.2.4. cast iron pipe

Cast iron pipe is the material that releases more iron into the water (Figure 8). The iron concentration in standing water in this pipe is low for 4 to 12 h for all three pH values. However, after 16 hours of stagnation, the iron concentration increases considerably at pH 5.11 (43.037 mg.L$^{-1}$) and at pH 9.11 (63.681 mg.L$^{-1}$). On the other hand, at neutral pH, the concentration of iron only reaches the same order of magnitude (43.409 mg.L$^{-1}$) after 6 days of water stagnation. For a prolonged period of stagnation (> 28 days), the concentration of iron increases in a neutral medium and at a slightly alkaline pH to reach very high values of 189.878 mg.L$^{-1}$ and 554.011 mg.L$^{-1}$ after 90 days of stagnation respectively while it decreases in acid medium.

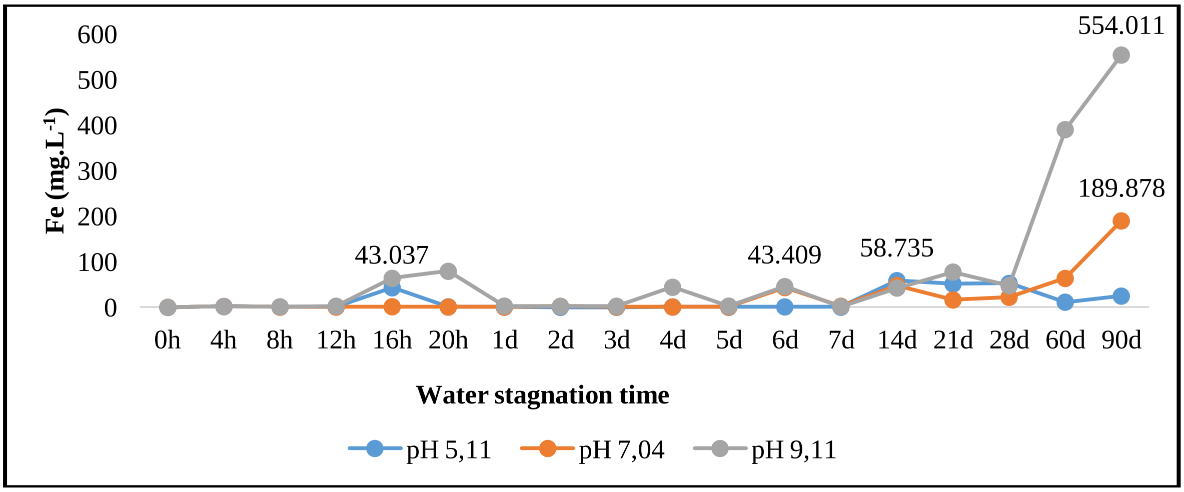


**Figure 8**: Effect of pH on the evolution of the iron concentration in the water remaining in the cast iron pipe.

**3.3. lead content**

**3.3.1. Polyvinyl chloride pipe**

Figure 9 shows the impact of pH on the evolution of the concentration of lead in stagnant water in the PVC pipe. The analysis of this figure shows that the concentration of lead at pH 5.11 evolves with higher values than those obtained at pH 7.04 and at pH 9.11 over practically the entire duration of the experiment. The concentration of Pb increases during stagnation at each pH considered and does not cancel out because lead would be used as an additive for the manufacture of PVC pipe. In contact with water, lead goes into solution and deposits (corrosion products) are very rare to form a barrier that can limit the contact of water with the wall of the pipe.

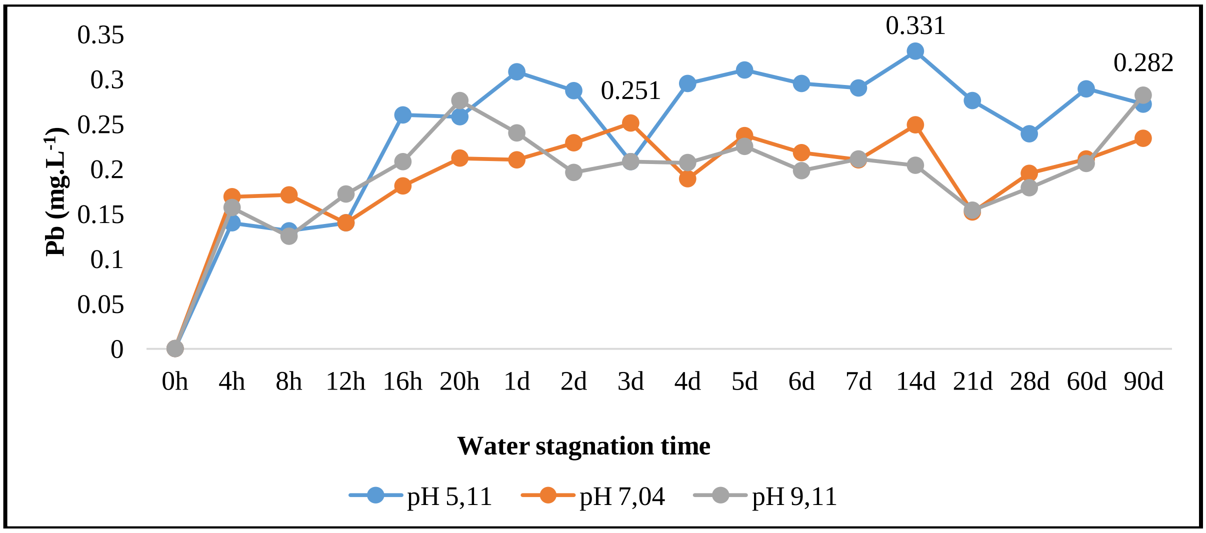


**Figure 9**: Effect of pH on the evolution of the lead concentration in the water remaining in the PVC pipe.

**3.3.2. galvanized steel pipe**

The lead concentration is very high at the start of stagnation (6.168 $mg.L^{-1}$ after 4 h) of water with a pH of 7.04 in the TG but gradually decreases as a function of time (Figure 10). In a slightly acid medium (pH 5.11), the Pb concentration fluctuates around lower values (1.176 - 2.935 $mg.L^{-1}$) but over a long period (7 days of stagnation) before falling to lower values. In a slightly alkaline environment (pH 9.11), the concentration of lead is lower (0.034 - 1.495 $mg.L^{-1}$) during the first 28 days of water stagnation compared to its concentration in environments with a pH of 5.11 and 7.04. The concentration peak (6,730 $mg.L^{-1}$) observed after 60 days of water stagnation would be due to detachment of corrosion product.

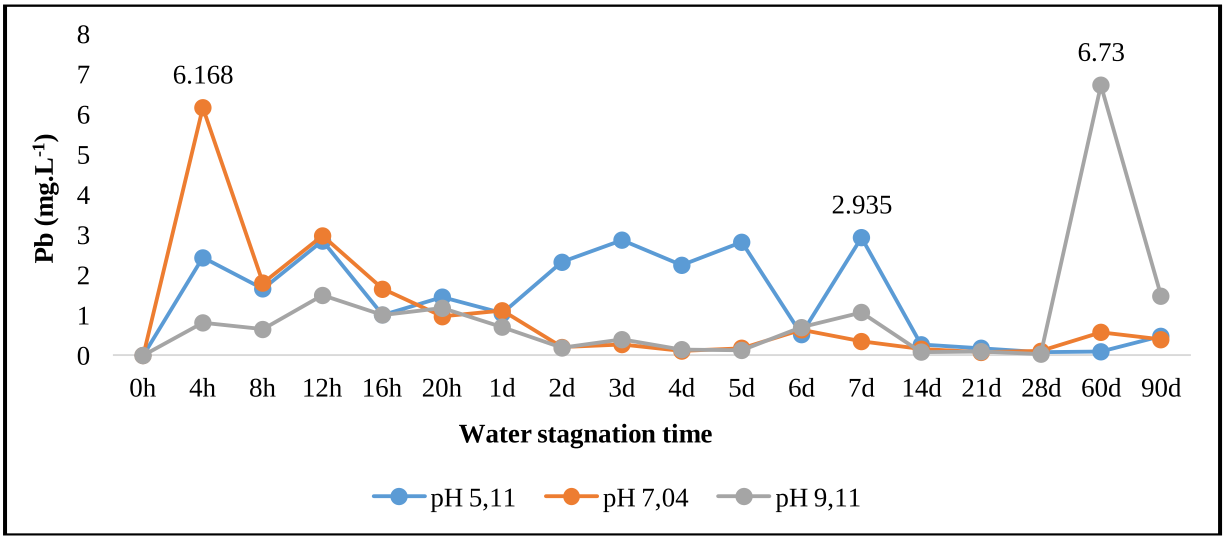


**Figure 10**: Effect of pH on the evolution of the lead concentration in the water remaining in the galvanized steel pipe.

### 3.3.3. Polypropylene pipe

The evolution of the lead concentration in the water remaining in the PPR pipe shows that the water with pH 9.11 has the highest lead concentration than in the case of pH 5.11 (0.203 - 1.941 mg.L$^{-1}$) and pH 7.04 (0.271 - 4.351 mg.L$^{-1}$) over the entire duration of the experiment (Figure 11). This concentration varies from 5.043 to 1.006 mg.L$^{-1}$ (from 4 h to the 2nd day) and from 9.256 to 2.502 mg.L$^{-1}$ (from the 3rd to the 28th day) of stagnation before evolving to a peak of 94.449 mg.L$^{-1}$ after 60 days.

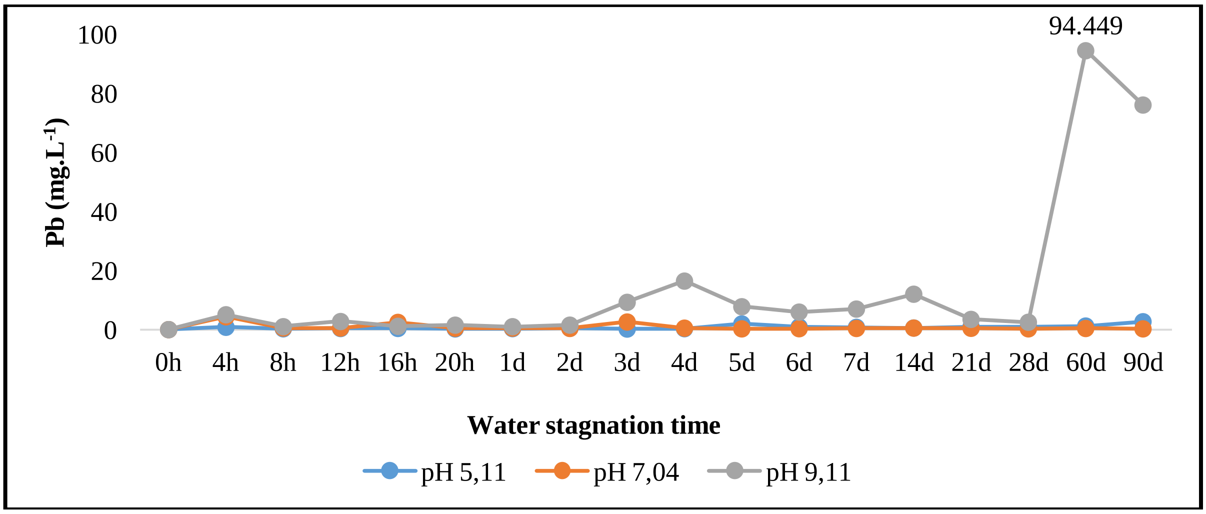


**Figure 11**: Effect of pH on the evolution of the lead concentration in the water remaining in the PPR pipe.

### 3.3.4. cast iron pipe

The lead concentration is very high (6.522 mg.L$^{-1}$; 5.881 mg.L$^{-1}$ and 4.022 mg.L$^{-1}$ respectively at pH 5.11; 7.04 and 9.11 after 4 hours of contact with water) in the cast iron pipe at the start of stagnation (Figure 12). It decreases during stagnation and reaches values around 0.300 mg.L$^{-1}$ after 3 days. This concentration remains low for the rest of the water stay but increases slightly to pH 7.04 and 9.11 from the 28th day. In general, the concentration of lead evolves with higher values when the pH decreases at the beginning of water stagnation in the cast iron pipe. But for a long period of water stagnation the concentration of lead gradually decreases during water stagnation. This decrease would be due to the formation of sparingly soluble lead hydroxycarbonate [11].

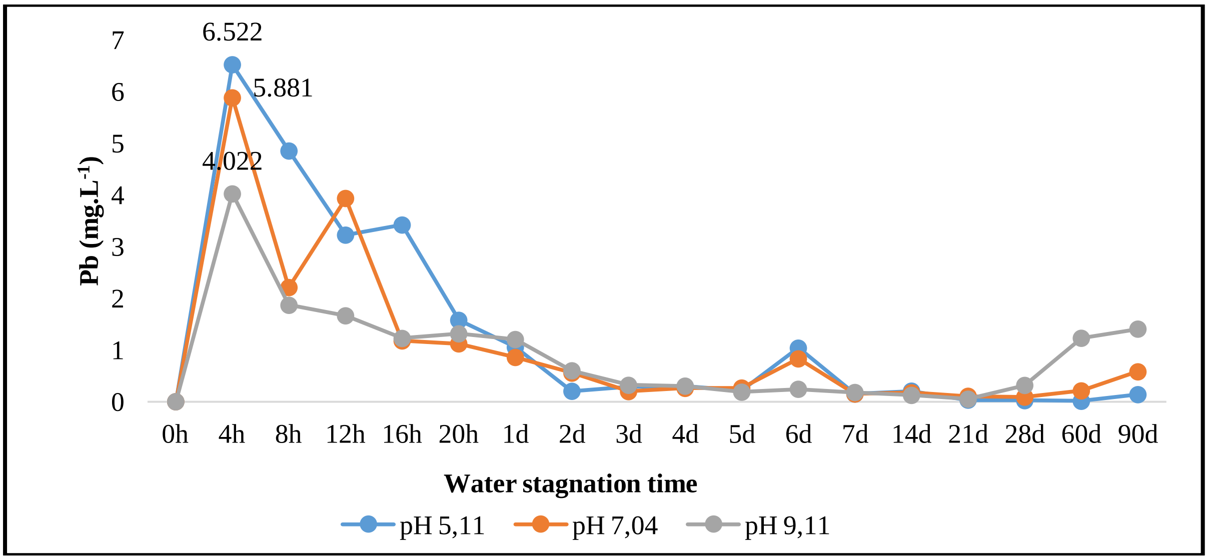


**Figure 12**: Effect of pH on the evolution of the lead concentration in the water remaining in the cast iron pipe.

### 3.4. Zinc content

#### 3.4.1. Polyvinyl chloride pipe

Figure 13 shows that zinc is in low concentration in PVC pipe. For the three pH values considered, the zinc concentration is higher at the beginning (4 h - 2 d) and for a prolonged period (21 - 60 days) of water stagnation. Zinc concentration evolves similarly at pH 5.11 and 7.04 during water stagnation with slightly elevated values at pH 7.04. In a slightly alkaline environment, the concentration of zinc evolves during stagnation with lower values compared to environments where the pH values are lower. However, this evolution is characterized by fluctuations, at the start of the water stay, between the lowest and highest concentration values. These fluctuations would be due to the displacement of the carbonates formed within the pipe at pH 9.11. Because carbonates can limit the contact of water with the walls of the pipe.

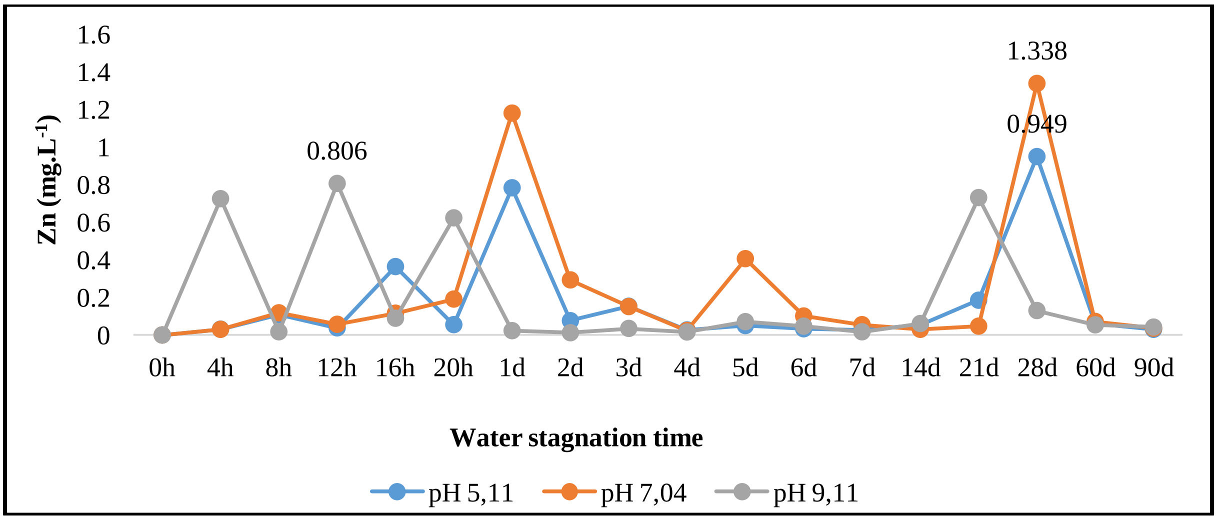


**Figure 13**: Effect of pH on the evolution of the zinc concentration in the water remaining in the PVC pipe.

#### 3.4.2. galvanized steel pipe

Galvanized steel is one of the materials that release more zinc into the water. This occurs most intensely at pH 7.04 and a prolonged period of stagnation (Figure 14). For pH 5.11 and 9.11 the zinc concentrations obtained are similar during the 90 days of stagnation.

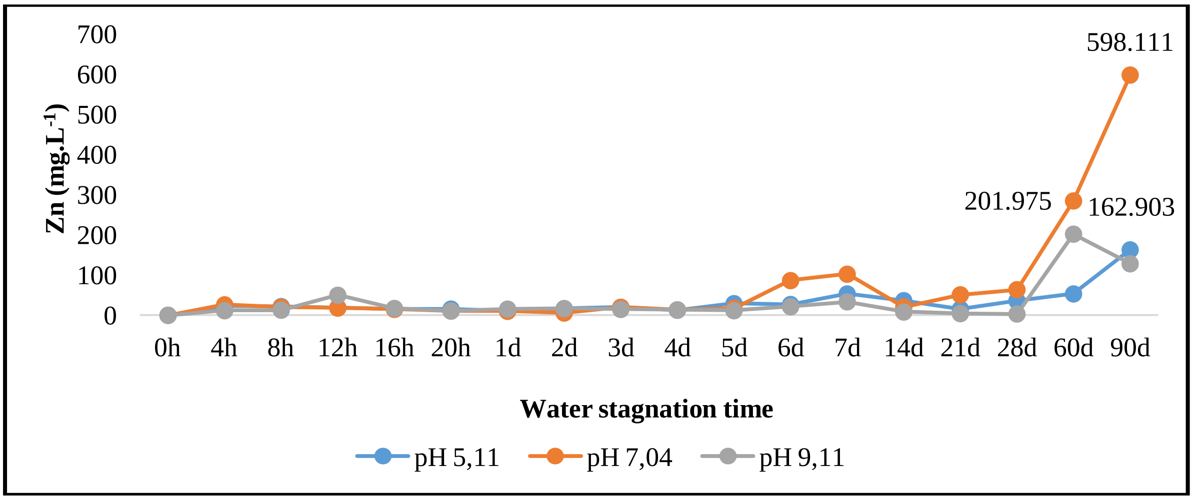


**Figure 14**: Effect of pH on the evolution of the zinc concentration in the water remaining in the galvanized steel pipe.

### 3.4.3. Polypropylene pipe

Figure 15 shows the effect of pH on the evolution of Zn concentration during water stagnation in the PPR pipe. These results show that the acid medium has Zn concentrations slightly above those found in neutral medium and in slightly alkaline medium up to 28 days of water stagnation. Then, water with a pH of 9.11 has higher Zn concentrations beyond 28 days.

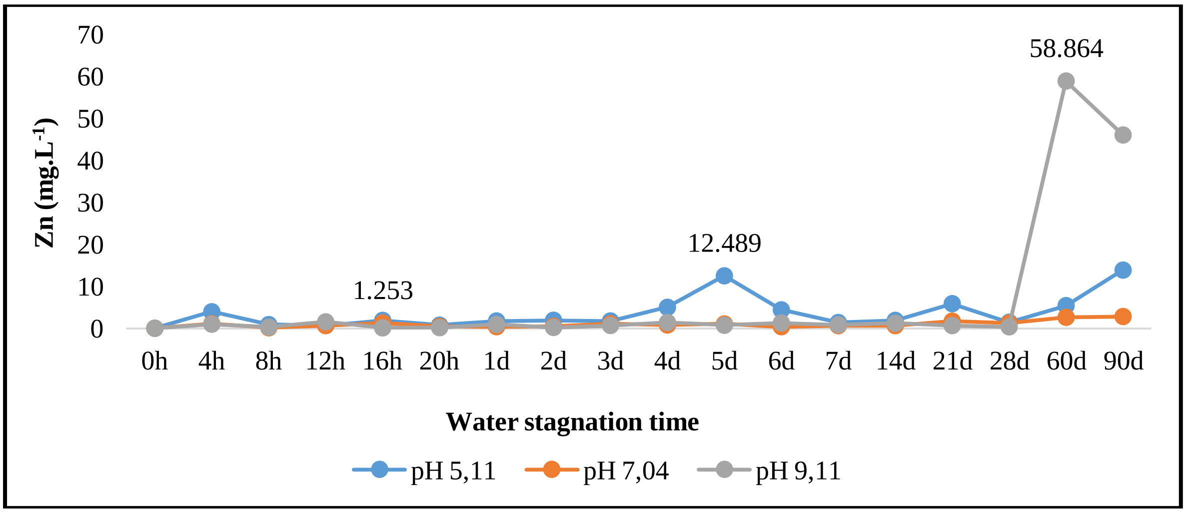


**Figure 15**: Effect of pH on the evolution of the zinc concentration in the water remaining in the PPR pipe.

### 3.4.4. cast iron pipe

The impact of pH on the evolution of the zinc concentration in the water remaining in the cast iron pipe is presented in Figure 16. It appears that the zinc concentration evolves towards higher values with stagnation time. pH 9.11 is the pH at which the concentration of Zn is higher than that found at pH 7.04, itself higher than that found at pH 5.11 over almost the entire duration of the experiment.

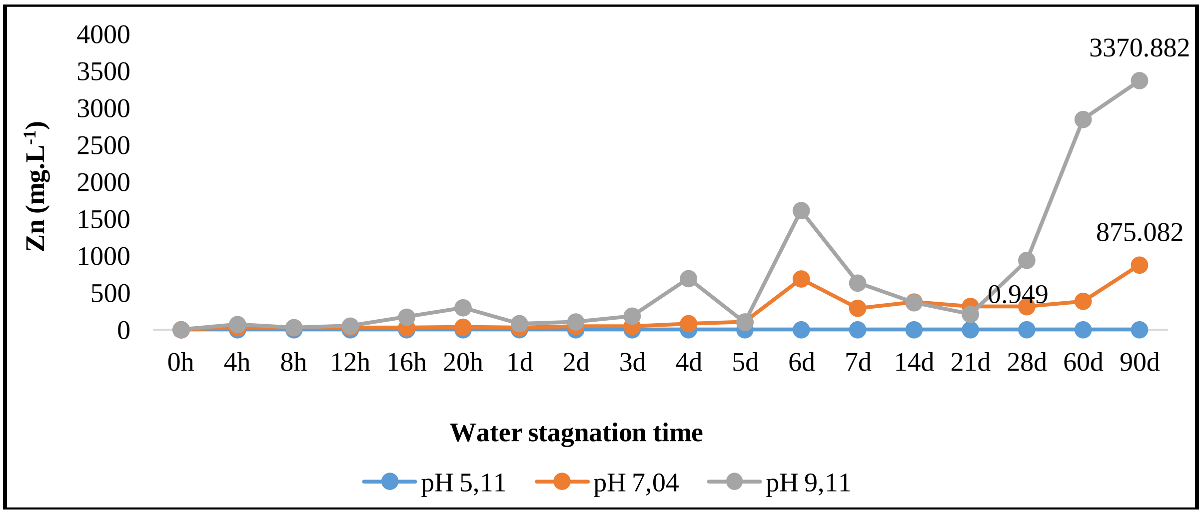


**Figure 16**: Effect of pH on the evolution of the zinc concentration in the water remaining in the cast iron pipe.

## IV. Conclusion

The evolution of the Cu, Fe, Pb and Zn contents as a function of the stagnation time of the water in the polyvinyl chloride, galvanized steel, polypropylene and cast iron pipe in the laboratory was studied at pH 5.11, 7.04 and 9.11. It appears from this study that all the new pipes made up of these pipes and accessories release Cu, Fe, Pb and Zn into the water under the conditions of the experiment. Metal pipes release much more of these four metals (except Pb) in front of plastic pipes. The comparative study of the effect of pH on the evolution of the concentrations of Cu, Fe, Pb and Zn generally showed that these concentrations are higher at the start of stagnation at slightly acidic pH (5.11). While over a long period of water stagnation, the concentration of these metals is higher at slightly alkaline pH (9.11). At neutral pH (7.04) the concentration of these metals fluctuates between extreme values throughout the duration of the experiment.

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MIJITABA SAHIROU Bachir, et. al. “ Comparative study of the evolution of copper, iron, lead and zinc levels in tap water after stagnation of the same duration in different pipes as a function of pH.” *IOSR Journal of Applied Chemistry (IOSR-JAC)*, 15(04), (2022): pp 20-29.