

INFLUENCE OF TEMPERATURE AND PH ON THE DECOMPOSITION KINETICS OF PERACETIC ACID IN AQUEOUS SOLUTIONS

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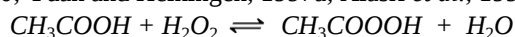
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Abstract— Peracetic acid (PAA) is a strong oxidant used by the food industry as a sanitizer, in medical area as a disinfectant and by the textiles and paper industries as a bleacher. Its decomposition rate is an important parameter in these applications. The main purpose of this paper is to study the decomposition kinetics of PAA between 25 and 45 °C in solutions with pH 3.11, 5.0 and 7.0. The decomposition of PAA is a first-order reaction for all solutions and temperatures studied. The rate constants were between $2.08 \cdot 10^{-3}$ and $9.44 \cdot 10^{-3} \text{ h}^{-1}$ (pH 3.11), between $2.61 \cdot 10^{-3}$ and $16.69 \cdot 10^{-3} \text{ h}^{-1}$ (pH 5.0) and between $7.50 \cdot 10^{-3}$ and $47.63 \cdot 10^{-3} \text{ h}^{-1}$ (pH 7.0). The activation energy of PAA decomposition in aqueous solutions are 58.36 and 72.89 kJ·mol⁻¹ when pH was 3.11 and 5.0, respectively.

Keywords— peracetic acid, decomposition kinetics, temperature, pH.

I. INTRODUCTION

PAA is a powerful sanitizer widely used by the food industry and is commercially available in the form of a quaternary equilibrium mixture containing acetic acid, hydrogen peroxide (HP), water and PAA as shown by the chemical equation (Zhao *et al.*, 2008; Falsanisi *et al.*, 2006; Kitis, 2004; Gehr *et al.*, 2003; Musante *et al.*, 2000; Yuan and Heiningen, 1997a; Alasri *et al.*, 1992):



Gehr *et al.* (2003) have reported that PAA is less stable than HP. A 40% PAA solution loses 1 to 2% of its active ingredients per month, while HP (30 to 90% solution) loses less than 1% per year. Block (1991) reported that dilute PAA solutions are even more unstable: a 1% solution loses half its strength through hydrolysis in 6 days. Gehr *et al.* (2003) recommends that PAA should be stored at cool temperatures in original containers in order to improve its stability. It was reported that PAA in solution may be consumed by hydrolysis, spontaneous decomposition or decomposition catalyzed by transition metal ions (Yuan and Heiningen, 1997b; Gehr *et al.*, 2003; Kitis, 2003; Veschetti *et al.*, 2003; and Zhao *et al.*, 2008).

Spontaneous decomposition of PAA occurred at a pH range of 5.5-10.2 (Gehr *et al.*, 2002 *apud* Kitis 2003; Koubek *et al.*, 1963 *apud* Zhao *et al.*, 2008). Zhao *et al.* (2008) reported that in an acid condition and at a temperature range of 55 to 95°C the spontaneous decomposition of PAA is a second-order reaction with respect to PAA concentration. However at a temperature below 55

°C the spontaneous decomposition of PAA is so insignificant that it could be negligible and hydrolysis became the predominant consumption factor for PAA. Zhao *et al.* (2007) reported that the hydrolysis of PAA in an acidic environment is first order with respect to PAA concentration, water and H⁺ concentration.

Rucker and Cates (1988) reported that the decomposition of peracetic acid is affected by the solution's pH and the observed decomposition rate of peracetic acid at 30 °C increases with pH ranging from $0.921 \cdot 10^{-3} \text{ h}^{-1}$ at 5.34 to $15.5 \cdot 10^{-3} \text{ h}^{-1}$ at 8.90.

Temperature also affects the decomposition of PAA. Greenspan and MacKellar (1955) observed that at room temperature, diluted solutions of PAA (10 g/L), lost half of their sanitation power within 6 days. Kunigk *et al.* (2001) have shown that increasing the temperature, a more rapid decomposition occurred. At 45 °C the concentration was halved in 72 hours, but at 25 °C the loss in 240 hours was only 33%. They concluded that temperature has an important role in the shelf life of PAA solutions.

The main purpose of this paper is to present results obtained in experiments carried out to study the influence of temperature and pH on the decomposition of PAA in aqueous solutions.

II. METHOD

A commercial formulation of PAA sanitizer containing around 200 g·L⁻¹ PAA, 300 g·L⁻¹ acetic acid, and 100 g·L⁻¹ HP was used in the experiments. The above product was diluted with distilled water in order to obtain concentrations of PAA between 240 and 900 mg·L⁻¹. This work also evaluated the influence of pH in the PAA decomposition kinetics. Therefore, two sets of PAA solutions were prepared with the same concentration range but the pH of the solutions were raised from its original pH (3.11) to 5.0 or 7.0 using a 0.1 M sodium hydroxide solution. The solutions were then maintained in 500 mL Erlenmeyers at constant temperatures (25.0, 35.0, 40.0 and 45.0 °C) in a water bath in static condition. The Erlenmeyers had been previously rinsed with a diluted HCl solution and then rinsed with de-ionized water before use. The concentrations of PAA were measured at regular intervals using the iodometric methodology as proposed by Greenspan and MacKellar (1948) and used by Yuan and Heiningen (1997a,b); Musante *et al.* (2000); Veschetti *et al.* (2003); Falsanisi *et al.* (2006) and Zhao *et al.* (2008) in their works. All chemicals used in the experiments (potassium perman-

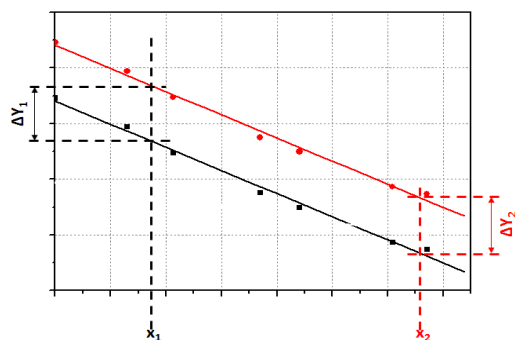


Figure 1 – Analyze done to verify if two lines are parallel.

ganate, potassium iodide, sodium thiosulfate, sulfuric acid) were analytically pure. No chelators were used in any of the PAA solution. Each datum was the average of three to five measurements. As described by Davies (1967), to compare regression lines, the following statistical analyses were done: variance about the regression; standard error of regression coefficient; difference between regression coefficients; and parallel line assays. Figure 1 shows the analyses done. Two equations can be considered parallel when there is not a significant difference between ΔY_1 and ΔY_2 within a ΔX value. The kinetic constants were fitted according to the experimental data using Origin 6.0 software.

III. RESULTS AND DISCUSSION

Kunigk *et al.* (2001) and Zhao *et al.* (2007) reported that PAA decomposition occurs by hydrolysis and decomposition kinetics can be expressed by a first-order reaction in an acid environment and below 50 °C with respect to PAA concentration. Zhao *et al.* (2008) and Kitis (2004) showed that at pH between 5.5 and 10.2, the spontaneous decomposition is the major mechanism of decomposition. The results obtained in this work for all temperatures and pH values can also be expressed by a pseudo first-order reaction as it can be seen in Figs. 2 to 4. Mathematical equation (1) was used to represent the decomposition kinetic.

$$\ln[PAA] = \ln[PAA]_0 - k \cdot t \quad (1)$$

where $[PAA]$ is the PAA concentration; $[PAA]_0$ is the initial PAA concentration ($\text{mg} \cdot \text{L}^{-1}$); t is the storage time (h) e k is the reaction rate constant (h^{-1}). Tables 1 to 8 show the equations parameters for pH 3.11 and 5.0. Doing a comparison of several regression lines as described by Davies (1967) it can be shown that equations for each table can be considered parallels ($p \leq 0.05$) unless for equations (12), (17), (22), (26), (30), (34) and (37). This behavior could be explained by the fact that under the experimental condition in which these equations were obtained, the dominant decomposition mechanism should be changing to the spontaneous decomposition which follows a second-order reaction. These changes in the mechanisms due temperature and pH variations were observed by Yuan and Heinigen (1997a,b), Zhao *et al.* (2008). Figure 3 shows the PAA decomposition and the Tables 9 to 12, their mathematical equations for a solution with pH corrected to 7.0. The equations in Tables 9 to 12 cannot be considered

Table 1 – Mathematical equations that represent PAA decomposition kinetics in solutions with pH 3.11 and stored at 25 °C. The regressions lines were shown in Fig. 1A.

$[PAA]_0$ (mg/L)	Linear co- efficient	Eq. slope $k \cdot 10^3$	r^2	Eq. N°
250	5.52	2.13	0.997	(2)
491	6.23	2.14	0.982	(3)
664	6.53	2.16	0.992	(4)
759	6.59	2.03	0.990	(5)
866	6.77	1.86	0.996	(6)

Table 2 – Mathematical equations that represent PAA decomposition kinetics in solutions with pH 3.11 and stored at 35 °C. The regressions lines were shown in Fig. 1B.

$[PAA]_0$ (mg/L)	Linear co- efficient	Eq. slope $(k \cdot 10^3)$	r^2	Eq. N°
250	5.50	4.30	0.998	(7)
491	6.21	4.25	0.992	(8)
664	6.52	4.46	0.998	(9)
759	6.58	4.10	0.980	(10)
866	6.77	4.81	0.999	(11)

Table 3 – Mathematical equations that represent Peracetic acid decomposition kinetics in solutions with pH 3.11 and stored at 40 °C. The regressions lines were shown in Figure 1C.

$[PAA]_0$ (mg/L)	Linear co- efficient	Eq. slope $(k \cdot 10^3)$	r^2	Eq. N°
250	5.52	5.05	0.998	(12)*
491	6.18	5.58	0.998	(13)
664	6.51	5.79	0.994	(14)
759	6.58	6.06	0.996	(15)
866	6.82	5.71	0.999	(16)

*equation not parallel with the others.

Table 4 – Mathematical equations that represent Peracetic acid decomposition kinetics in solutions with pH 3.11 and stored at 45 °C. The regressions lines were shown in Figure 1D.

$[PAA]_0$ (mg/L)	Linear co- efficient	Eq. slope $(k \cdot 10^3)$	r^2	Eq. N°
250	5.52	8.29	0.994	(17)*
491	6.18	9.55	0.983	(18)
664	6.58	9.14	0.994	(19)
759	6.64	9.38	0.996	(20)
866	6.79	9.51	0.998	(21)

*equation not parallel with the others.

parallel ($p \leq 0.05$) when compared with the others equations in the same table when the analyses described by Davies (1967) were done.

Figure 5 shows the slopes of the equations shown in Tables 1 to 8 as a function of PAA initial concentration. Tables 9 to 12 were not used due the equations in a same table were not parallel between them. The parameters of equations obtained from Fig. 5 are showed in Tables 13 and 14. These equations are parallel to x-axis ($p \leq 0.05$) but they are not coincident lines. Therefore the PAA initial concentration does not affect the PAA decomposition kinetics but storage temperature affects its decomposition. Therefore, the linear coefficient shown in Tables 13 and 14 was used as the rate constants of the PAA decomposition. These figures and tables show that increasing the temperature and the pH, the decomposition of PAA also increase.

From Table 13 (PAA solutions with pH 3.11) it can be seen that increasing the temperature from 25 to 45 °C, there was an increase of the rate constant of PAA

decomposition (obtained from linear coefficient) of 4.5 times going from $2.08 \cdot 10^{-3}$ to $9.44 \cdot 10^{-3} \text{ h}^{-1}$, respectively. For PAA solutions with pH 5.0, it can be seen from Table 14 that the rate constant of PAA decomposition has increased 6.4 times. Therefore, temperature has an important role on rate constants of PAA decomposition for all studied pH conditions. Kunigk *et al.* (2001) found a variation in rate constant of 5.6 by increasing the temperature from 25 to 45 °C. The difference between the variations of the observed rate constant in these two studies can be explained by the fact that it was used a different commercial PAA formulations (unpublished data). Therefore the formulation composition of sanitizers can affect their decomposition. The smaller decomposition rate constant obtained in this work could mean that this product is less affected by temperature

Table 5 – Mathematical equations that represent PAA decomposition in solutions with pH 5.0 and stored at 25 °C. The regressions lines were shown in Fig. 2A.

[PAA] ₀ (mg/L)	Linear coefficient	Eq. slope ($k \cdot 10^3$)	r^2	Eq. N°
246	5.46	2.27	0.951	(22)*
487	6.19	2.55	0.998	(23)
752	6.63	2.51	0.996	(24)
880	6.72	2.50	0.996	(25)

*equation not parallel with the others.

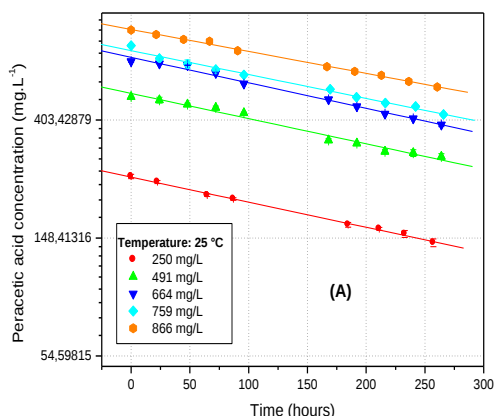


Table 6 – Mathematical equations that represent PAA decomposition in solutions with pH 5.0 and stored at 35 °C. The regressions lines were shown in Fig. 2B.

[PAA] ₀ (mg/L)	Linear coefficient	Eq. slope ($k \cdot 10^3$)	r^2	Eq. N°
246	5.50	4.87	0.998	(26)*
487	6.19	6.49	0.999	(27)
752	6.64	6.48	0.999	(28)
880	6.80	6.22	0.998	(29)

*equation not parallel with the others.

Table 7 – Mathematical equations that represent PAA decomposition in solutions with pH 5.0 and stored at 40 °C. The regressions lines were shown in Figure 2C.

[PAA] ₀ (mg/L)	Linear coefficient	Eq. slope ($k \cdot 10^3$)	r^2	Eq. N°
246	5.46	5.78	0.996	(30)*
487	6.20	10.57	0.996	(31)
752	6.68	10.47	0.997	(32)
880	6.79	10.57	0.994	(33)

*equation not parallel with the others.

Table 8 – Mathematical equations that represent PAA decomposition in solutions with pH 5.0 and stored at 45 °C. The regressions lines were shown in Figure 2D.

[PAA] ₀ (mg/L)	Linear coefficient	Eq. slope ($k \cdot 10^3$)	r^2	Eq. N°
246	5.45	8.63	0.990	(34)*
487	6.31	15.99	0.998	(35)
752	6.64	17.39	0.999	(36)
880	6.75	22.44	0.992	(37)*

*equations not parallel with the others.

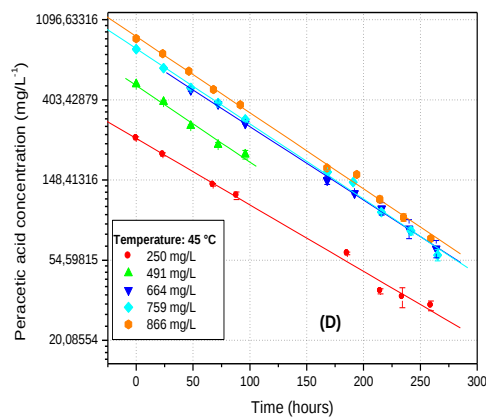
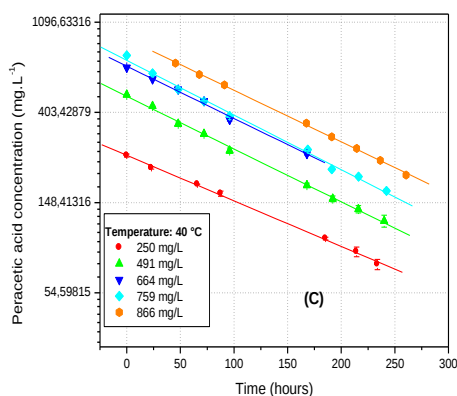
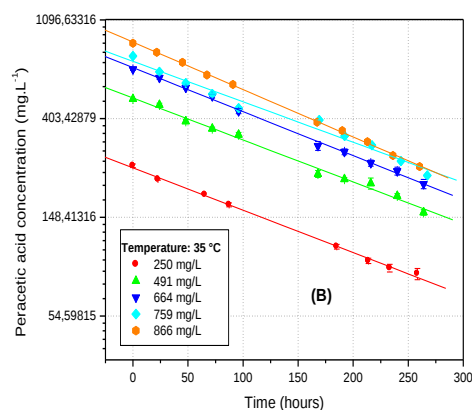


Figure 2 – Peracetic acid concentration in solutions with pH 3.11. The regressions' parameters were shown in Tables 1 to 4.

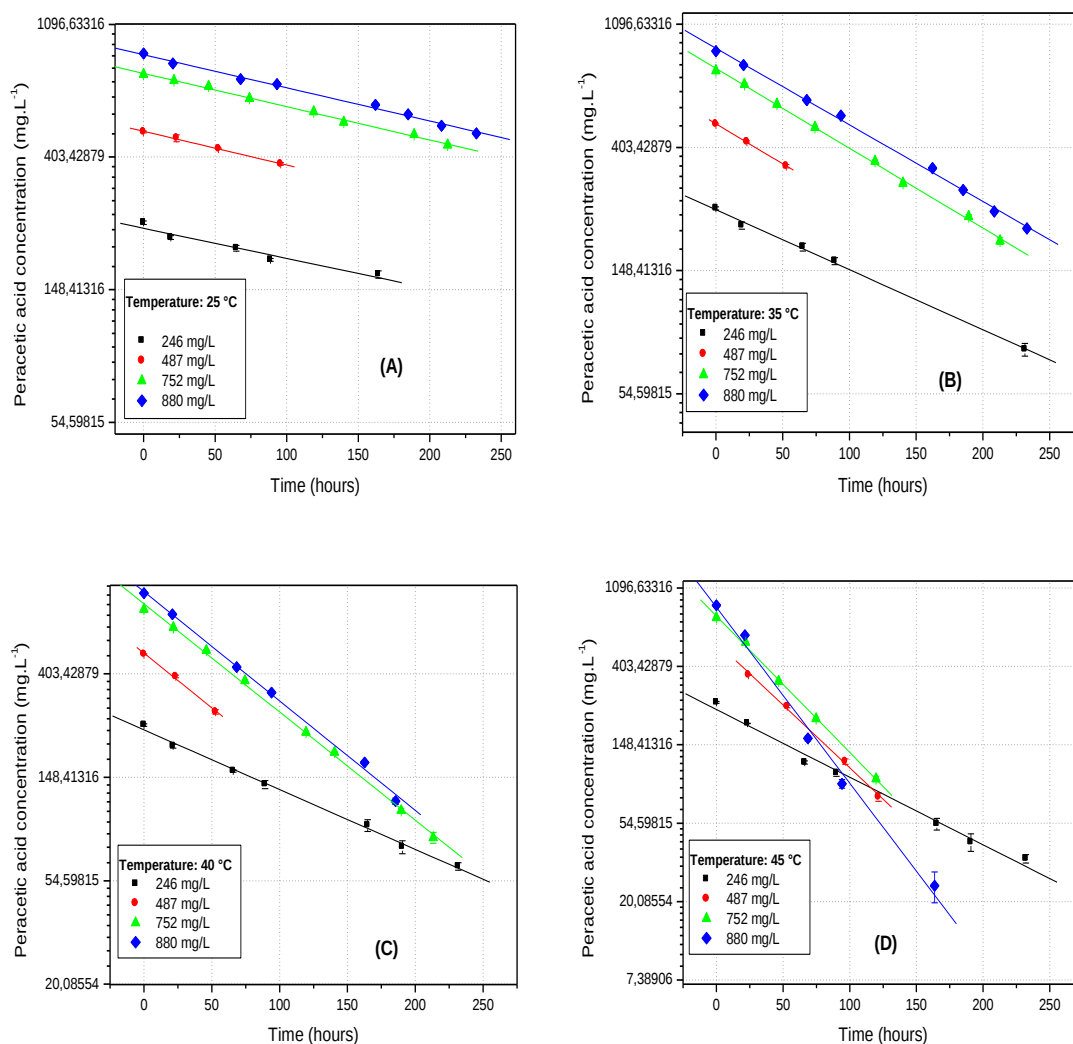


Figure 3 – Peracetic acid concentration in solutions with pH 5.0. The regressions' parameters were shown in Tables 5 to 8.

than the product used by Kunigk *et al.* (2001). Rucker and Cates (1988) found an observed first order constant for decomposition of PAA solutions at pH 7.1 ranging from $4.572 \cdot 10^{-2} \text{ h}^{-1}$ at 20°C to $4.836 \cdot 10^{-1} \text{ h}^{-1}$ at 40°C that means a variation of 10.6. At this pH we found a variation of 9.1 by increasing the temperature from 25 to 45°C . Despite higher temperatures, the lower variation in this work can be explained by the use of distilled water solutions which has less contamination to promote PAA decomposition.

The PAA half-life, $t_{1/2}$, was calculated using equation (1) and values of linear coefficient from Table 13 and 14. It was admitted that $[\text{PAA}]_0$ is initial PAA concentration and $[\text{PAA}]_0/2$ is final PAA concentration. The PAA half-life, $t_{1/2}$, was calculated by:

$$t_{1/2} = \frac{0.6931}{k} \quad (76)$$

Equation (76) is a rate constant function only. Therefore, for PAA solutions with pH 3.11, half-life went from 73.4 to 333.2 hours when the solutions were stored at 45°C and 25°C , respectively. The variation obtained was 78%. Kunigk *et al.* (2001) found a variation of 68%

when storage temperature increased from 25 to 40°C and a variation of 82% when storage temperature increased from 25 to 45°C . For PAA solutions with pH 5.0, half-life went from 41.5 to 265.6 hours when the solutions were stored at 45°C and 25°C so the variation was 84%. PAA solutions with pH 7.0, half-life went from 14.6 to 92.4 hours for the same temperature range and a variation of 84% was also observed. Yuan *et al.* (1997b) have shown that at pH 7.0 the half-life at 25, 50 and 70°C were 672, 144 and 48 hours, respectively. These results show that increasing the temperature and the pH there are a faster decomposition of PAA.

Another important parameter to consider in kinetics studies is the activation energy, calculated by Arrhenius equation:

$$k = k_0 \cdot e^{-\frac{E_a}{R \cdot T}} \quad (77)$$

Figure 6 shows the Arrhenius plots of the experimental data of this work. Table 15 show the frequency or pre-exponential factor, k_0 (h^{-1}) and the activation energies, E_a ($\text{J} \cdot \text{mol}^{-1}$), from PAA decomposition.

Table 9 – Mathematical equations that represent PAA decomposition in solutions with pH 7.0 and stored at 25 °C. The regressions lines were shown in Fig. 3A.

[PAA] ₀ (mg/L)	Linear coef.	Eq. slope ($k \cdot 10^3$)	r^2	Eq. N°
239	5.47	6.99	0.986	(38)
240	5.46	6.63	0.984	(39)
487	6.16	4.22	0.982	(40)
507	6.20	20.78	0.992	(41)
522	6.23	14.50	0.998	(42)
733	6.57	6.46	0.978	(43)
887	6.80	10.49	0.978	(44)
889	6.76	4.18	0.974	(45)

Table 10 – Mathematical equations that represent PAA decomposition in solutions with pH 7.0 and stored at 35 °C. The regressions lines were shown in Fig. 3B.

[PAA] ₀ (mg/L)	Linear coef.	Eq. slope ($k \cdot 10^3$)	r^2	Eq. N°
239	5.45	16.14	0.996	(46)
240	5.47	13.69	0.994	(47)
487	6.15	28.50	0.994	(48)
507	6.20	14.55	0.994	(49)
522	6.24	15.90	0.992	(50)
733	6.58	20.34	0.988	(51)
887	6.83	31.73	0.999	(52)
889	6.83	17.05	0.992	(53)

Table 11 – Mathematical equations that represent PAA decomposition in solutions with pH 7.0 and stored at 40 °C. The regressions lines were shown in Fig. 3C.

[PAA] ₀ (mg/L)	Linear coef.	Eq. slope ($k \cdot 10^3$)	r^2	Eq. N°
239	5.53	37.61	0.996	(54)
240	5.47	19.51	0.999	(55)
487	6.31	49.49	0.994	(56)
507	6.28	42.50	0.998	(57)
522	6.23	25.58	0.999	(58)
733	6.61	32.17	0.988	(59)
887	6.83	35.33	0.972	(60)
889	6.83	26.74	0.990	(61)

Table 12 – Mathematical equations that represent PAA decomposition in solutions with pH 7.0 and stored at 45 °C. The regressions lines were shown in Fig. 3D.

[PAA] ₀ (mg/L)	Linear coef.	Eq. slope ($k \cdot 10^3$)	r^2	Eq. N°
239	5.54	43.29	0.992	(62)
240	5.42	33.39	0.990	(63)
487	6.33	42.00	0.978	(64)
507	6.15	57.18	0.986	(65)
733	6.55	47.42	0.998	(66)
887	6.29	57.31	0.908	(67)
889	6.83	30.36	0.984	(68)

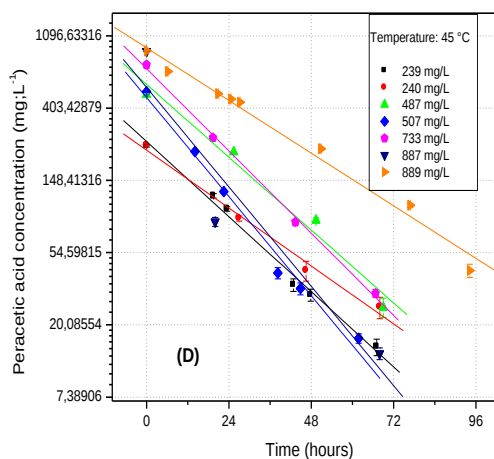
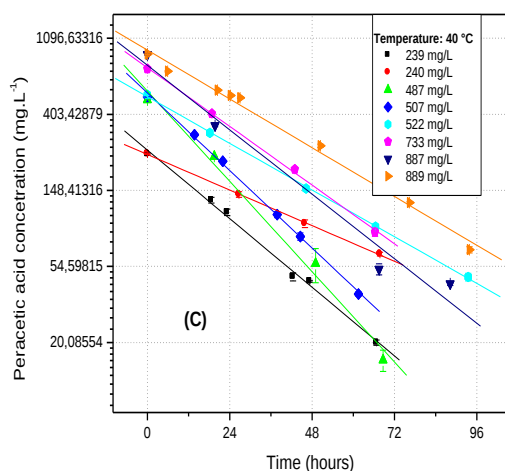
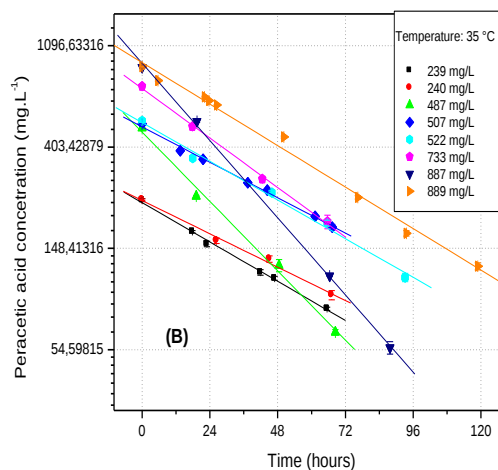
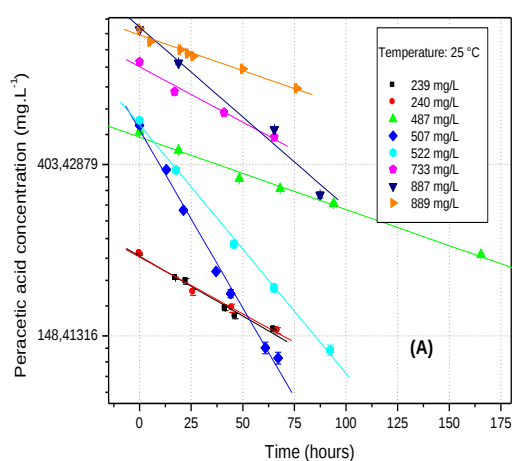


Figure 4 – Peracetic acid concentration in solutions with pH 7.0. The regressions' parameters were shown in Tables 9 to 12.

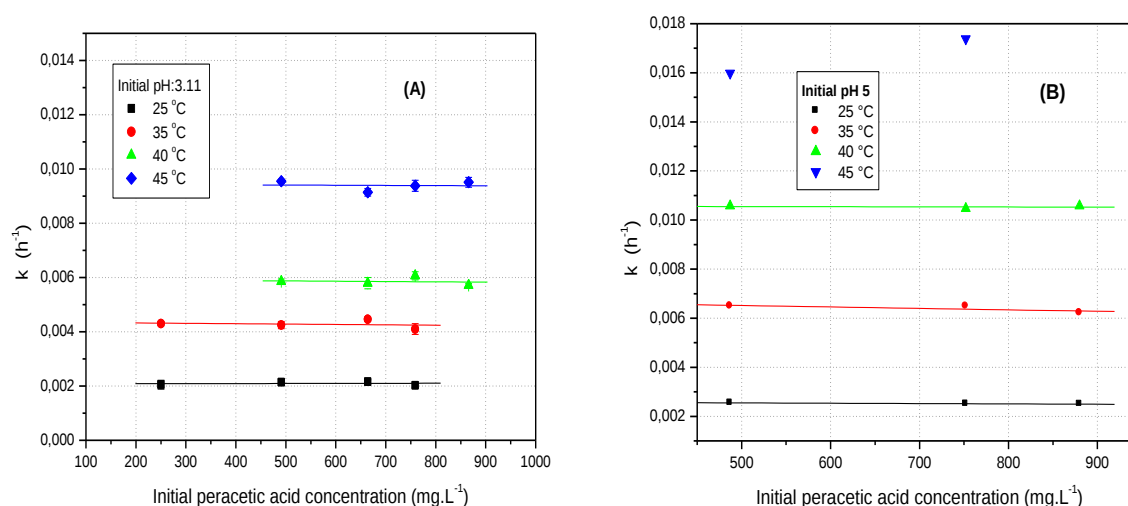


Figure 5 – Peracetic Acid decomposition rate constant as a function of the initial Peracetic Acid concentration $[PAA]_0$ for the four temperatures studied. The regressions' parameters were shown in Tables 13 and 14.

Table 13 – Mathematical equations that represent the influence of $[PAA]_0$ over the rate constants of PAA decomposition with pH 3.11. The regressions lines were shown in Figure 4A.

Temp. (°C)	Linear coefficient ($\times 10^3$)	Eq. slope ($k \cdot 10^8$)	r^2	Eq. N°
25*	2.08	3.12	0.012	(69)
35*	4.35	14.30	0.046	(70)
40**	5.93	11.41	0.015	(71)
45**	9.44	6.60	0.003	(72)

*the equations for $[PAA]_0 = 866 \text{ mg} \cdot \text{L}^{-1}$ were not parallel. Slopes were not used.

** the equations for $[PAA]_0 = 250 \text{ mg} \cdot \text{L}^{-1}$ were not parallel. Slopes were not used.

Table 14 – Mathematical equations that represent the influence of $[PAA]_0$ over the rate constants of PAA decomposition with pH 5.0. The regressions lines were shown in Figure 4B.

Temp. (°C)	Linear coefficient ($\times 10^3$)	Eq. slope ($k \cdot 10^8$)	r^2	Eq. N°
25	2.61	1.31	0.098	(73)
35	6.81	5.89	0.060	(74)
40	10.58	5.68	0.039	(75)
45	16.69*	---	---	---

*rate constants are the average of the equations slopes #35 and #36.

Table 15 – Pre-exponential factor and activation energies for PAA decomposition kinetics.

pH	$\ln(k_0)$	Ea (kcal·mol ⁻¹)	Ea (kJ·mol ⁻¹)	Eq. N°
3.11	7.50	13.94	58.36	(78)
5.0	18.87	17.41	72.89	(79)

Table 15 shows that activation energies of PAA decomposition, in solutions with pH 3.11, is close to the values obtained by Kunigk *et al* (2001) and Zhao *et al* (2007), 66.2 and 60.4 kJ·mol⁻¹, respectively. Zhao *et al* (2007) have shown that activation energies of PAA de-

composition, in solutions with pH 8.2 was 94.4 kJ·mol⁻¹. All these values are lower than the 99.65 kJ·mol⁻¹ reported by Rucker and Cates (1988) who have studied the PAA decomposition at a pH of 7.1. These data suggests that, the higher the pH of solutions, more temperature-sensitive is the reaction; reactions with low activation energy are relatively temperature-insensitive (Levenspiel, 1999). So the higher the activation energy, the lower must be the temperature variation to double the rate constant of PAA decomposition. Therefore, it is important the prevention of contaminations with alkalis during the utilization of PAA solutions.

IV. CONCLUSION

We can see that by increasing temperature and pH, the rate constant of PAA decomposition increases. The decomposition of PAA, can be represented by a first-order reaction and was not affected by initial PAA concentration for all temperature and pH values studied in this work. It was also shown that the rate constant is affected by temperature according to Arrhenius equation. The activation energy of peracetic acid decomposition in aqueous solutions prepared from a commercial formulation used in this work is 58.36 kJ·mol⁻¹ when pH was 3.11 and 72.89 kJ·mol⁻¹ when pH was 5.0.

Peracetic acid is an instable molecule. This instability can be harmful to sanitation processes where the peracetic acid concentration plays a significant role in microorganism destruction. Destroying fewer microorganisms, more bacteria can be present either in food processing surfaces as in the food products so consumers could be put at risk.

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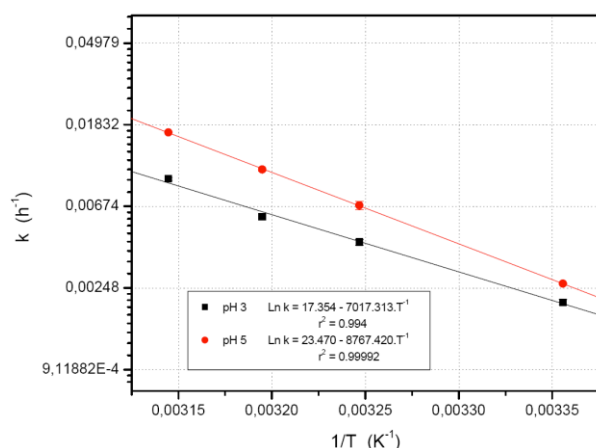


Figure 6 – The influence of temperature on the rate constant for decomposition of PAA for all pH studied.

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