



Synthesis, Kinetics and Mathematical Modelling of Environment Friendly Acrylate-Based Binder

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ABSTRACT

The copolymers consisting of acrylic and methacrylic esters have achieved prime importance due to their versatile applications. Copolymerization of Methyl meth acrylate (MMA) with butyl acrylate has been carried out for modifying the properties of the polymer. Dynamic swelling kinetics were conducted at room temperatures to investigate the synthesized binder's swelling properties for paint industries. The experimental swelling curves were analyzed using three different models: Peleg's model, the first-order absorption kinetic model, and the exponential association equation model. All of these models demonstrated excellent agreement with the experimental data, as indicated by high R-Square values and low values for Chi Square, Sum of Squared Errors (SSE), and Root Mean Square Error (RMSE). Comparing the determination coefficients for these models, it was concluded that the Peleg model provides a better representation of the swelling characteristics across various concentrations of the crosslinker in the polymer. Specifically, the Peleg model exhibited high R-Square values of 0.98121, 0.9869, and 0.97605 for 0%, 5%, and 10% PPGDA concentrations, respectively. Furthermore, it yielded reduced chi-square values of 1.44546, 0.74895, and 0.86587 and root mean square error values of 1.202, 0.8654, and 0.9305 for the same respective concentrations. These results establish the Peleg model as the most favorable choice for characterizing the swelling behavior with different cross linker concentrations in the polymer. The prepared latexes are used as binder for environment friendly coatings.

Keywords: Acrylate; Swelling; Kinetics; Polymer; Peleg model; First order absorption model; Exponential association model.

1. INTRODUCTION

In recent years, due to higher concerns on environmental protection, the emulsion polymerization technique is selected, as a green approach for the synthesis of polymers (Kane *et al.* 2008; Pladis *et al.* 2014; Kang *et al.* 2015). During the last few decades, polymer industry has been the fastest growing sector of chemical industry. Polymers are quite often used in the field of agriculture, medicine, automobile parts, packing materials, adhesives, surface coatings, matrix for composites, and elastomers, etc. The surface coatings is an important application of polymers for protecting a wide variety of substrates against the influences of different parameters like mechanical, chemical, and atmospheric, and at the same time, serve to decorate the substrate. A number of homopolymers and co-polymers are used as film-forming materials. The prime requirement for a polymer to form a film is its capacity to form bonds with different surfaces. The solvent evaporation technique has been used for the preparation of films on various substrates for the last four decades. Due to environmental hazards related to solvent evaporation mechanism and its higher cost, the water borne cured films are emerging as better option for achieving superior properties in coating applications. Day to day increase of solvents cost is major attraction

for the development of water borne coatings. Among all polymeric binders the acrylate-based binder is major in use for coating-based applications. Acrylic resins may be used alone or as blend with other resins to form the suitable binder system for coatings. The literature with the free radical polymerization of the acrylic resin has been reported by number of researchers (McKenna *et al.* 1995). The relative ease of polymerization and the wide range of properties among the acrylic resins have led to the commercial production of many different resins suitable for broad variety of applications (Zhu *et al.* 1990). For example, these resins play a prominent role in the paint industry due to the unique combination of their properties (Zhou *et al.* 2018). In this study, polyacrylate latex was synthesized by emulsion polymerization technique. The swelling characteristics like swelling ratio, swelling rate, diffusion kinetics, and activation energy of polymer latex were investigated based on the swelling values obtained at different time intervals. In the literature, few research and models were conducted about the swelling kinetics of acrylate latex. Our unique approach entails the application of mathematical modelling, leveraging the Peleg model, exponential association equation model, and first-order absorption kinetic model, to elucidate the swelling kinetics of acrylate latex. This endeavor aims to bridge existing gaps in the literature, where limited research and models have been developed concerning the

swelling kinetics of acrylate latex. Through a combination of empirical data and mathematical frameworks, our study aspires to offer fresh insights into this crucial facet of polymer behavior.

2. MATERIALS AND METHOD

2.1 Experimental Materials

Monomers Methyl methacrylate (MMA; Aldrich, New Delhi) and Butyl Acrylate (BA; Aldrich, New Delhi) were used after the purification by alkali method. Free radical initiator potassium persulfate (KPS; Thomas Baker, Mumbai) and emulsifier sodium lauryl sulphate (SLS; Thomas Baker) were used as received. Crosslinker polypropylene diacrylate (PPGDA) has been prepared in laboratory (Shukla *et al.* 2013) by taking polypropylene glycol 400 (PPG400; Thomas baker) and acrylic acid (AA; Thomas baker). Deionized water was used throughout the experimental work.

2.2 Polymerization

The emulsion polymerization was carried out in a three-necked round-bottom flask of capacity 500 mL, equipped with stirrer, reflux condenser, dropping funnel, and a thermometer. Polymer latex samples were prepared by thermally initiated free radical polymerization (KPS as initiator) of MMA and BA in the presence and absence of crosslinker as monomer PPGDA. The polymerization reaction was carried out at temperature $70 \pm 1^\circ\text{C}$, using emulsifier SLS for the course of 3 hours. The percentage conversion of synthesized latex is varied with the feedratio of monomer in the previous study (Bajpai *et al.* 2009). The selected synthesized latex was coated on clean and smooth substrates as glass. The films were dried for 1 hour in a hot air oven to remove most of the water present as solvent. The thickness of the dried films in case of all the polymer ratio synthesized, namely SA₅, SA₅₁, and SA₅₂, was maintained as 0.1 ± 0.05 mm. The actual feed compositions and designations of polymers are given in Table 1.

Table 1. Monomer feed compositions (MMA/BA/PPGDA)

Polymer code	Mole ratio of monomers		Weight percentage of monomers			Amount of MMA (g)	Amount of BA (g)	Amount of PPGDA (g)
	MMA	BA	MMA	BA	PPGDA			
SA ₅	5	5	50	50	0	21.9	28.1	0
SA ₅₁	5	5	41.6	53.3	5	20.8	26.6	2.5
SA ₅₂	5	5	39.4	50.5	10	19.7	19.7	5

2.3 Swelling kinetics of Latex Films

Gravimetric analysis is used to determine the degree of swelling. In every experiment, a pre-weighed cured sample was suspended in a beaker of distilled water at room temperature. The film was taken out from the water at different time intervals, quickly blotted free of surface water by using filter paper, weighed on analytical balance, and return again to the swelling medium. The swelling ratio and degree of swelling were calculated from the equation 1 (Frisch *et al.* 1969).

$$\text{Swelling ratio} = \frac{W_s}{W_d} \quad (1)$$

$$\text{Degree of swelling (\%)} = \frac{W_s - W_d}{W_d} \times 100$$

Where, W_d and W_s were the weight of dry and swollen films respectively.

3. MATHEMATICAL MODELLING OF SWELLING CONTENT OF POLYMER

This study focuses on mathematical modeling of the swelling behavior of polymers. Several well-established methods have been utilized in our study, which includes the application of Peleg's model (2), the first-order absorption kinetic model (3), and the

exponential association equation (4) (Vasudeva *et al.* 2010; Noshad *et al.* 2012; Zeinali Kalkhoran *et al.* 2018). Peleg introduced a two-parameter model (equation 2) for characterizing the swelling content of polymers (Vasudeva *et al.* 2010):

$$C = C_0 \pm \frac{t}{k_1 + k_2} \quad (2)$$

Where, C represents the swelling content at any time t (g/g d.b.), C_0 is the initial swelling measurement at $t = 0$ (g/g d.b.), k_1 is the kinetic constant of the model (h(g d.b.)/g), k_2 is a characteristic constant of the model (g d.b.)/g).

In equation (2), the sign “ \pm ” changes to “ $+$ ” if the process pertains to absorption or adsorption, and to “ $-$ ” if it pertains to drying or desorption. The first-order absorption kinetic model employs the following equation (Grnicki *et al.* 2013):

$$C = C_e + (C_0 - C_e) \exp(-k_{r1}t) \quad (3)$$

Where, C_e represents the equilibrium swelling content, k_{r1} is the swelling kinetic constant (h^{-1}). The exponential association equation is expressed by equation (4):

$$C = C_e [1 - \exp(-k_{r2}t)] \quad (4)$$

Where, k_{r2} is the kinetic constant (h^{-1}).

These equations help in quantifying and understanding the swelling behavior of polymers under different conditions, providing valuable insights into absorption, adsorption, drying, and desorption processes. This study carries significant importance in the field of polymer research for several reasons:

1. **Understanding Polymer Expansion in Various Conditions:** These models provide a useful set of tools for studying how polymers expand in different situations. By using these models, scientists can better grasp how polymers react to processes like soaking up liquids, sticking to surfaces, drying out, and releasing absorbed substances. This flexibility is important because polymers are used in many different ways, and their swelling behavior can change based on the situation.
2. **Parameter Estimation for Comprehensive Polymer Swelling Analysis:** In these models, several parameters come into play, including kinetic constants (k_1 , k_2 , k_{r1} , and k_{r2}) and initial swelling content (C_0). When researchers match real-world data with these models, they can determine these parameters. This process offers valuable insights into the fundamental physical and chemical processes that control polymer swelling. This parameter estimation plays a crucial role in quantitatively characterizing polymer behavior.
3. **Parameter Estimation for Comprehensive Polymer Swelling Characterization:** In the study of polymer swelling, various models include parameters like kinetic constants (k_1 , k_2 , k_{r1} , and k_{r2}) and initial swelling content (C_0). By matching experimental data to these models, scientists can determine these parameters, offering valuable insights into the fundamental physical and chemical processes that control polymer swelling. This parameter estimation process contributes to a quantitative understanding of polymer behavior.
4. **Comparative Analysis for Model Selection:** By employing multiple models, researchers can conduct a comparative analysis to determine which model aligns best with their experimental data. This method aids in choosing the most suitable model for a specific polymer or process. This comparative approach improves the precision and trustworthiness of the research outcomes.
5. **Predictive Abilities:** After successfully fitting these models to real-world data, they can be used to make predictions. Researchers can use them to anticipate how polymers will behave in different situations or over time. This valuable information can help in making choices about which materials to use and how to optimize processes.
6. **Scientific Progress:** Above mentioned models help to advance our scientific knowledge in the field of polymer science. Researchers can use these models

as a foundation to create more detailed and accurate descriptions of how polymers behave. This deeper understanding of polymer properties and behavior contributes to the overall growth of our knowledge in the field.

Hence, using these models in polymer research allows researchers to thoroughly study and grasp polymer swelling behavior. It empowers them to make predictions and addition to the extensive pool of scientific knowledge in this field. This knowledge proves invaluable in designing materials and processes across diverse industries such as pharmaceuticals, agriculture, and materials science.

4. STATISTICAL ANALYSIS

Statistical analysis of the experimental data was conducted utilizing both MATLAB and Origin Software. The assessment of the fit between the experimental data and all three models was based on several key statistical criteria within a 95% confidence interval. These criteria included Chi-square, Sum of Squared Errors (SSE), R-Square, and Root Mean Square Error (RMSE) (Kaleta *et al.* 2010). The criterion for selecting the best model, favoring the one with the highest R-Square and the least Chi-Square, SSE, and RMSE, is a sound approach. A model with a high R-Square value explains a significant portion of the data's variability, while low values for Chi Square, SSE, and RMSE indicate that the model closely matches the observed data points. This rigorous statistical analysis ensures that the chosen model accurately represents the swelling characteristics of the polymer under investigation, providing confidence in the study's findings and conclusions.

5. EXPERIMENTAL FINDINGS AND DISCUSSION

5.1 Swelling Curves

Swelling plays a crucial role in the characterization of polymers. Figure 1 illustrates the variations in swelling content over different time intervals. This three-dimensional (3D) chart was generated to represent the combined impact of both time and PPGDA concentration on polymer swelling. The chart provides insights into how the swelling content is influenced by these two input variables. Notably, it is evident from the absorption curves that the rate of swelling diminishes as the concentration of PPGDA increases. This chart serves as a valuable tool for understanding the interplay between these two input variables and their impact on swelling behavior. By examining the chart, it becomes evident that the absorption curves demonstrate a specific trend: as the concentration of PPGDA increases, the rate of swelling decreases. This observation is significant as it indicates that PPGDA concentration plays a pivotal role in modulating the swelling behavior of the polymer. As the

concentration rises, the polymer exhibits reduced swelling, suggesting a potential application for controlling and fine-tuning the swelling characteristics of the material. Understanding these relationships between variables is fundamental in tailoring polymer properties for various applications, ranging from drug delivery systems to materials science. Figure 1, with its visual representation of these dynamics, contributes valuable insights to the field of polymer research and engineering.

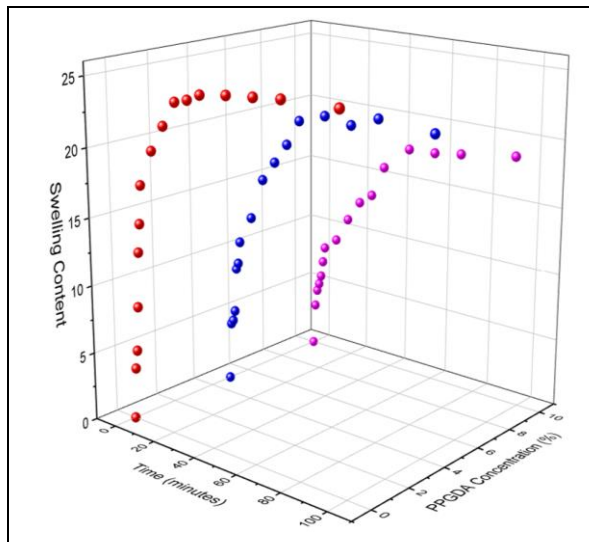


Fig. 1: Swelling content at different PPGDA concentrations 0% (Red), 5% (Blue), 10% (Pink)

5.2 Evaluation of Peleg Model

Fig. 2 presents the outcomes of the swelling tests conducted at different time intervals for varying PPGDA concentrations, specifically 0%, 5%, and 10%. These experimental results have been fitted to Peleg's

model, which is a powerful tool for characterizing swelling behavior in polymers. The fitted model provides valuable insights into how the polymer's swelling properties change with time and PPGDA concentration.

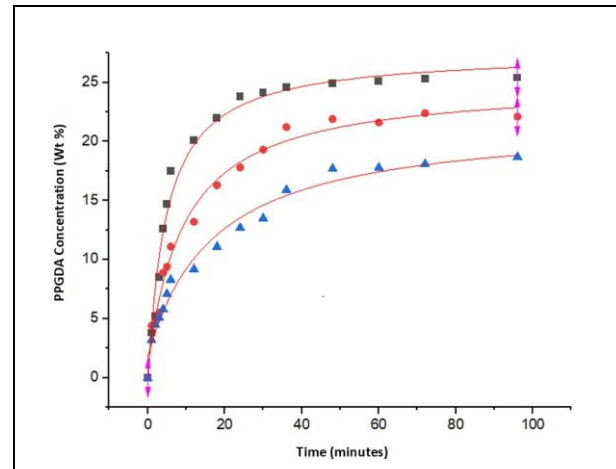


Fig. 2: Peleg model at 0% (Brown), 5% (Red), 10% (Blue) PPGDA Concentration

In Table 2, we can find the constants k_1 and k_2 for each of the PPGDA concentrations tested. These constants are fundamental to the Peleg model and help quantify the kinetics and characteristics of the swelling process. Additionally, the table includes statistical values associated with the Peleg model, which further validate the model's goodness of fit to the experimental data.

Based on the above information of Peleg model, we have generated various parameters and use these values to generalize the concepts that is to formulate for each parameter regarding other PPGDA concentration, which is represented in Fig. 3.

Table 2. Peleg Model Coefficients and Statistical Parameters for Swelling Content at Different PPGDA Concentrations

PPGDA Concentration (Wt%)	C_0	k_1	k_2	Chi-Square	SSE	R-Square	RMSE
0	-1.2293	0.15611	0.03475	1.44548	18.79	0.98121	1.202
5	0.7067	0.37994	0.04116	0.74895	9.736	0.9869	0.8654
10	1.71881	0.83812	0.0497	0.86587	11.26	0.97605	0.9305

These figures likely illustrate how each of the model parameters changes in relation to varying PPGDA concentrations. The figures provide a visual representation of how the constants k_1 and k_2 and other model-related parameters vary as a function of PPGDA concentration. This data enables the generalization of concepts, helping to formulate a better understanding of how PPGDA concentration influences the swelling behavior of the polymer.

In essence, this comprehensive analysis, including experimental data, model fitting, parameter determination, and subsequent generalization through Figure 3, contributes significantly to our understanding of how PPGDA concentration impacts the swelling characteristics of the polymer. Such insights are invaluable in designing and tailoring polymers for specific applications in fields such as materials science and pharmaceuticals.

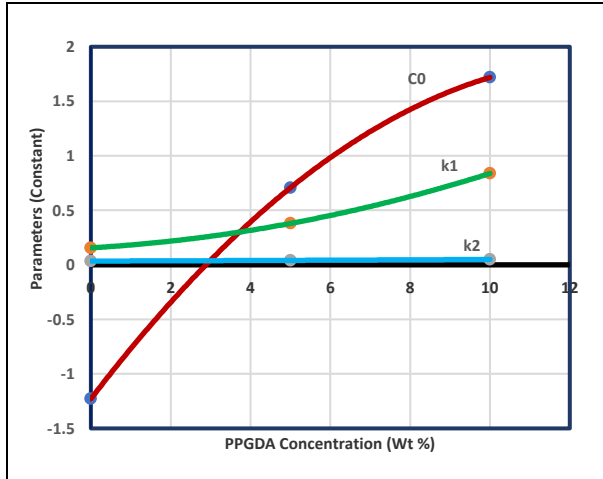


Fig. 3: Impact of PPGDA Concentration on various parameters of Peleg Model and their modeling

5.3 Evaluation of First-order Absorption Kineticmodel

Fig. 4 displays the experimental results of the swelling tests conducted at various time intervals for different PPGDA concentrations (0%, 5%, and 10%). The data in Figure 4 have been fitted to the first-order absorption kinetic model, demonstrating how well the model aligns with the actual experimental observations. This fitting process helps assess the model's ability to describe the swelling behavior under different conditions.

Table 3 presents the coefficients associated with the first-order absorption kinetic model for each PPGDA concentration. These coefficients are essential for characterizing the kinetics of the swelling process. Table 3 also includes statistical values that provide insights into the goodness of fit between the model and the

experimental data. These statistics help determine the model's reliability and accuracy in representing the swelling behavior.

Overall, this evaluation serves as a critical step in assessing the applicability and performance of the first-order absorption kinetic model in describing polymer swelling under varying PPGDA concentrations. It helps validate the model's effectiveness in capturing the underlying kinetics of the swelling process and provides valuable data for further analysis and application in fields such as materials science and polymer engineering. Based on above information of first-order absorption kinetic model, we have generated various parameters and use these values to generalize the concepts that is to formulate for each parameter regarding other PPGDA concentration, which is represented in Fig. 5.

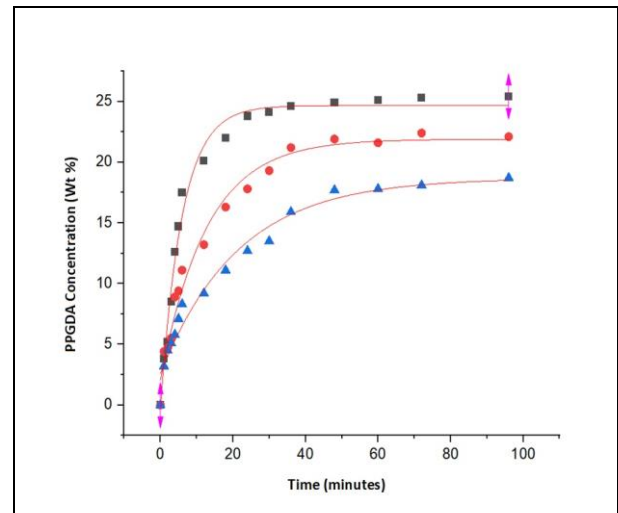


Fig. 4: First-order absorption kinetic model at 0% (Brown), 5% (Red), 10% (Blue) PPGDA Concentration

Table 3. The first-order absorption kinetic model Coefficients and Statistical Parameters for Swelling Content at Different PPGDA Concentrations

PPGDA Concentration (Wt%)	C_0	C_e	k_{r1}	Chi-Square	SSE	R-Square	RMSE
0	-0.36407	24.66111	0.16843	1.08042	14.05	0.98596	1.039
5	2.02568	21.89761	0.07864	1.16365	15.13	0.97966	1.079
10	2.55896	18.68602	0.04681	1.15208	14.98	0.96814	1.073

Table 4. The coefficients of the exponential association equation and Statistical Parameters for Swelling Content at Different PPGDA Concentrations

PPGDA Concentration (Wt%)	C_e	k_{r2}	Chi-Square	SSE	R-Square	RMSE
0	24.6903	0.16419	1.01841	14.25775	0.9816	1.047
5	21.36377	0.10099	1.73097	24.23355	0.9603	1.365
10	17.44128	0.07438	2.4622	34.47062	0.9124	1.628

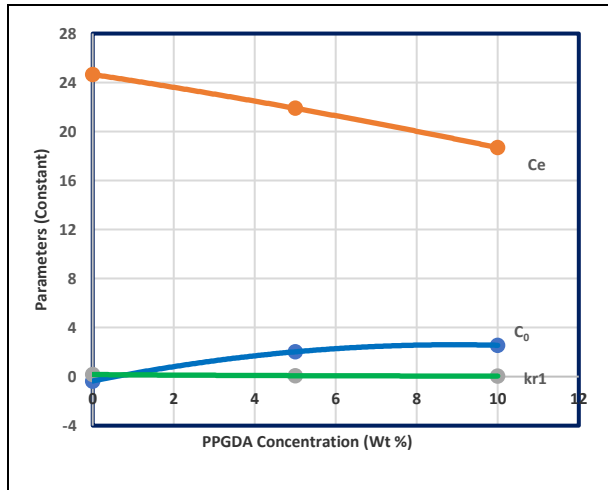


Fig. 5: Impact of PPGDA Concentration on various parameters of first-order absorption kinetic model and their modeling

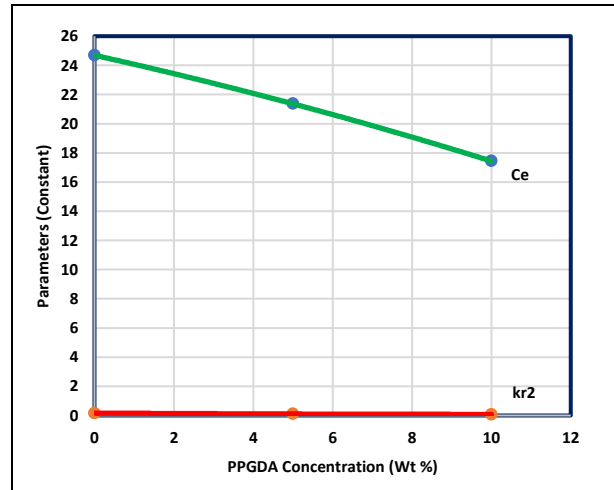


Fig. 7: Impact of PPGDA Concentration on various parameters of exponential association equation and their modeling

5.4 Evaluation of Exponential Association Equation

Fig. 6 shows the results of swelling test at different times for the various PPGDA concentrations like 0%, 5% and 10% concentration, fitted on exponential association equation.

Table 4 shows the coefficients of exponential association equation at various PPGDA concentrations and also represents the statistical values of this model.

Based on above information of exponential association equation, we have generated various parameters and use these values to generalize the concepts that is to formulate for each parameter regarding other PPGDA concentrations, which is represented in Fig. 7.

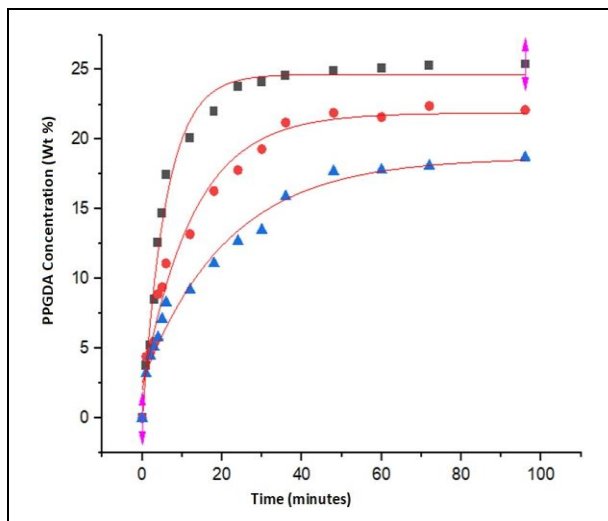


Fig. 6: Exponential association equation at at 0% (Brown), 5% (Red), 10% (Blue) PPGDA Concentration

6. CONCLUSION

The prepared polymer latexes are waterborne coating due environmental hazards related to solvent evaporation and its cost; the waterborne systems are emerging as one of the better options for achieving superior properties in high performance applications. In conclusion, our study has yielded valuable insights into the swelling kinetics of the polymer, particularly concerning varying concentrations of acrylates:

- **Model Performance:** All three models employed in our study (Peleg's model, first-order absorption kinetic model, and exponential association model) have demonstrated satisfactory predictive capabilities when characterizing the polymer's swelling content across different acrylate concentrations. This underscores the versatility of these models for analyzing polymer swelling.
- **Swelling Kinetics:** Throughout the immersion process, we observed an initial rapid increase in swelling content. However, as time progressed, the rate of swelling gradually decreased until it reached a saturation point. This observation highlights the dynamic nature of the swelling process and the significance of considering time-dependent behavior.
- **Peleg model's rate constants, k_1 and k_2 ,** exhibited an increase with higher concentrations of PPGDA. This implies that as PPGDA concentration rises, the swelling kinetics of the polymer become more definite.
- **First-Order Absorption Model:** Conversely, the kinetic model constant K_{r1} from the first-order absorption model showed a decrease with increasing PPGDA concentration. This suggests a potentially

complex relationship between PPGDA concentration and the first-order kinetics of swelling.

- Exponential Association Model: Similarly, the exponential constant K_{12} of the exponential association model decreased with higher PPGDA concentrations, indicating that as PPGDA concentration increases, the exponential association between time and swelling content becomes less prominent.
- Model Selection: After conducting statistical comparisons, we determined that the Peleg model provided the best fit for modeling the experimental data. This finding underscores the Peleg model's suitability for describing the polymer's swelling behavior under the specific conditions investigated in our study.

In summary, these results offer a comprehensive understanding of how PPGDA concentration affects the swelling characteristics of the polymer. The study highlights the importance of considering time-dependent kinetics and provides valuable data for modeling and optimizing polymer swelling in various practical applications. The selection of the Peleg model as the best fit further validates its applicability in characterizing polymer swelling behavior in this context.

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CONFLICTS OF INTEREST

The authors declare that there is no conflict of interest.

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