Modeling chemical kinetics of geopolymers using physics informed neural network

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Article Info

ABSTRACT

Using a physics informed neural network for the analysis of geopolymers as an alternate material for cement can be a viable approach, as neural networks are capable of modeling complex, nonlinear relationships in data, which can be beneficial for representing the dynamics of chemical properties. If you have a substantial amount of theoretical data, a neural network can learn patterns and relationships in the data, even when the underlying system dynamics are not well-defined or are difficult to model analytically. A well-trained neural network can generalize from the training data to make predictions for unseen scenarios, which can be useful for real-time analysis of the material.

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1. INTRODUCTION

"Geopolymer" was originally used by Joseph Davidovits in 1978. He suggested producing the blocks by pouring water, clay, lime, and crushed limestone into moulds. When this material formed into an artificial stone, he called it geopolymer. On the basis of the same idea, scientists have created a brand-new concrete that has zero cement in it! They use a variety of waste materials, industrial residues, and minimally processed natural resources in place of Portland Cement. These waste materials are difficult to get rid of and might harm the environment if left unchecked. Therefore, using them could significantly reduce the carbon footprint of concrete [1]. They are fly ash, sometimes referred to as PFA (pulverized fuel ash), which is a fine, powdery byproduct of burning coal in power plants. Its main constituents are silicon dioxide, aluminium oxide, and calcium oxide. Meta-kaolin is produced by heating kaolin or China clay to 1,500 degrees Fahrenheit or 800 degrees Celsius. It can be used to make tiles and conventional concrete. Granulated blast furnace slag that has been ground is a byproduct of making steel. Its high calcium silicate hydrate (CSH) content improves the appearance, durability, and strength of concrete. Palm oil fuel ash, or POFA, is created by burning the husks and shells of palm oil. Even though it can be used to make cement, this dangerous substance is usually disposed of in landfills. Each of these raw materials has a unique performance and yields a distinct kind of geopolymer cement. The majority of waste products contain silicon dioxide, which is the straightforward general response. They are used with an alkaline activator such as potassium or sodium hydroxide, together with potassium or sodium silicate. Aluminosilicate gels are created when the silicon oxygen silicon bonds disintegrate and allow aluminium atoms to pass through. These gels solidify into a geopolymer cement when more alkali is added. The mixture of cement, aggregate, and water creates geopolymer concrete. Given the complexity of geopolymers and the environmental implications of their use,

modeling and analyzing their properties and performance become crucial. Physics-informed neural networks (PINNs), which have shown promise in various domains for solving differential equations and modeling complex systems [2], [3], offer an innovative approach to understanding the behavior and optimizing the composition of geopolymers. This method, blending domain knowledge with computational modeling, could lead to more efficient use of waste materials and further advancements in sustainable construction methods.

2. PINN VS ANN

Partial differential equations (PDEs) are fundamental in characterizing PINNs, serving as a universal function approximator capable of incorporating any physical rules governing a data set into the learning process [4]. This capability is especially valuable in fields where data may be sparse, rendering many advanced machine-learning algorithms less effective. In such contexts, prior knowledge of physical principles acts as a regulatory agent within neural network (NN) training, narrowing the possible solution space and enhancing accuracy in function approximation [5]. The rising popularity of PINNs across various engineering fields is largely due to their proficiency in addressing practical problems characterized by noisy inputs and frequently missing physics [6]. Automatic differentiation enables PINNs to evaluate differential operators accurately without the need for discretization, creating a multitask learning scenario that aims to fit observed data while adhering to physical laws [7]. Such applications have been explored in heat transfer challenges among others, showcasing the method's effectiveness where traditional computational approaches may falter [8]. For example, investigations into convection on heated surfaces utilizing sparse temperature observations aim to determine temperature and velocity fields, including boundary conditions, illustrating PINNs' potential in deriving comprehensive solutions from limited data [9]. The Stefan problem, addressing two-phase flow with the goal of inferring moving interfaces and distinct phase properties from minimal temperature measurements, further exemplifies PINNs utility [10]. A wide array of industrial applications, particularly in power electronics, underscores the practical benefits of PINNs in bridging the divide between computational and experimental approaches to heat transfer and solving problems considered intractable by conventional methods [11], [12]. This attribute of PINNs to interpret physical laws through partial differential equations (e.g., Navier Stokes equations from fluid mechanics) reinforces their significance in quantifying system dynamics under defined conditions [13], [14]. Understanding PINNs also requires a grasp of basic neural network functions, which approximate a mapping function from input vectors to output, utilizing weights and biases [15], [16]. Through multiple layers and activation functions, neural networks achieve complex function approximations, essential for modeling physical phenomena [17], [18]. Back-propagation, crucial for learning from data, and the availability of gradients, enabling the application of physics to neural networks, are foundational elements of PINNs [19], [20]. The development of custom loss functions incorporating differential equation residuals, alongside initial and boundary conditions, facilitates the resolution of differential equations [21], [22]. PINNs stand out when data is scarce but the underlying physical principles are well understood. They apply the mathematical framework of differential equations to impose physical constraints on neural networks, leveraging universal approximation capabilities and readily available derivatives [23]. Neural networks thus become powerful tools for optimizing solutions to complex differential equations, supported by custom loss functions that integrate physical laws directly into the learning process [24], [25].

3. MATHEMATICAL ANALYSIS OF GEOPOLYMERS

The mathematical analysis of geopolymers, particularly through the lens of differential equations to describe chemical reactions, is pivotal in understanding the kinetics and mechanistic pathways of geopolymerization reaction seen in Figure 1. The differential equation representing the rate of chemical reactions can express the change in concentration of reactants and products over time. For a simple reaction where reactant A converts to product B with a rate constant K1, the equations can be stated as:

$$\delta(A)/\delta(t) = -K1[A] \tag{1}$$

Here, [A] denotes the concentration of reactant A. Similarly, for product B, the rate of formation is proportional to its concentration.

$$\delta(B)/\delta(t) = -K1[B] \tag{2}$$

In more complex scenarios, such as those involving multiple reactants, intermediates, and reverse reactions, similar rate-concentration models can be constructed to describe the system dynamics. Geopolymerization, characterized by its complexity, involves the transformation of aluminosilicate

precursors into a structured network through such chemical reactions. This process demands a multidisciplinary approach combining experimental techniques like infrared spectroscopy and high-energy X-ray diffraction to probe the molecular and nano structural changes, alongside thermogravimetry and rheology to assess physical properties. The mathematical modeling of geopolymers serves as a complementary tool to these experimental techniques, offering insights into the reaction kinetics across different scales. Through the application of differential equations, informed by data such as reactant concentrations and rate constants under specific initial and boundary conditions, a quantitative framework can be developed to simulate the geopolymerization process. This modeling capability is crucial for designing geopolymer mixtures tailored for specific applications, enhancing quality control during manufacturing, and optimizing the material properties. Comparing the solutions of these mathematical models with experimental data, such as calorimetric measurements, validates the accuracy of the models and their predictive power. Additionally, modeling the heat evolution during geopolymerization can inform strategies to accelerate the setting and hardening processes, crucial for industrial production. PINNs offer a forward-looking approach to tackle the mathematical modeling challenges associated with geopolymers. By integrating the governing physical laws, such as the differential equations describing chemical reactions and heat transfer, into the architecture of neural networks, PINNs can learn the complex relationships inherent in the geopolymerization process. This approach not only enhances the predictive accuracy of the models but also opens new avenues for discovering novel geopolymer formulations and optimizing manufacturing processes through computational simulations. In conclusion, the mathematical analysis of geopolymers, supported by both traditional and PINN-based modeling approaches, is essential for advancing our understanding of geopolymer chemistry, improving material properties, and fostering innovation in sustainable construction materials.
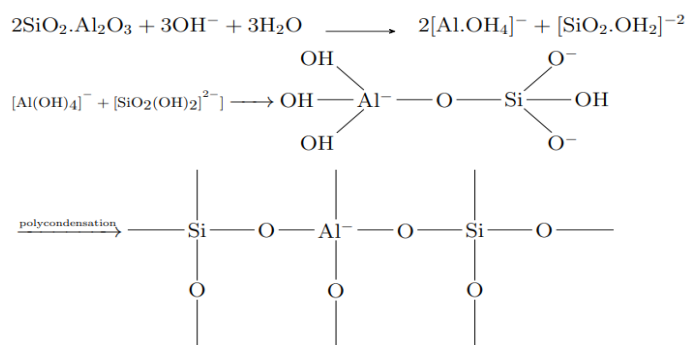

Figure 1. Geopolymerization chemical reaction

4. RESULTS AND DISCUSSION

The results, represented by a series of figures, provide an in-depth examination of the geopolymerization process, employing a dual approach of mathematical observation and PINN modeling to understand the kinetic and dynamic aspects of this complex reaction. Figure 2 temperature approximation is a three-dimensional (3D) scatter plot that elegantly demonstrates the relationship between temperature, time, and the change in concentration of reactants during the geopolymerization reaction. The plot reveals a significant correlation between temperature ($x1$ axis, ranging from -1 to 1) and the change in concentration (z-axis), overshadowing the time factor ($x2$ axis, from 0 to 1). This pattern indicates an inverse parabolic relationship, suggesting that at elevated temperatures, the concentration of reactants diminishes more swiftly. This behavior is indicative of an endothermic reaction, wherein reactant consumption intensifies with rising temperatures a phenomenon characteristic of many chemical processes but particularly relevant in the context of geopolymerization. Such temperature-driven kinetics, especially under the constraint of low reactant concentrations, point to a scenario where thermal conditions exert a dominant influence over the reaction rate, a consequence of the significant role played by the activation energy in such processes. The Arrhenius equation, which defines the rate of a chemical reaction as a function of temperature, underscores the critical nature of this relationship. Small fluctuations in temperature can profoundly affect the reaction rate, particularly when the activation energy of the process is substantial. Understanding the intricacies of this relationship is paramount for optimizing geopolymer synthesis. The data suggest a means to predict the progression of the reaction across different temporal and thermal conditions, facilitating the industrial scaling

of geopolymer production where control over material properties is crucial for quality assurance. As we advance to Figure 3, the complexity of the model is enhanced by integrating time-dependent kinetics alongside thermal influences, portraying a comprehensive view of the reaction's evolution. Here, the dependency on both temporal and thermal variables heralds a more complex mechanism, reflective of the reaction's actual progression and the interplay between the consumption of reactants and the formation of new products.
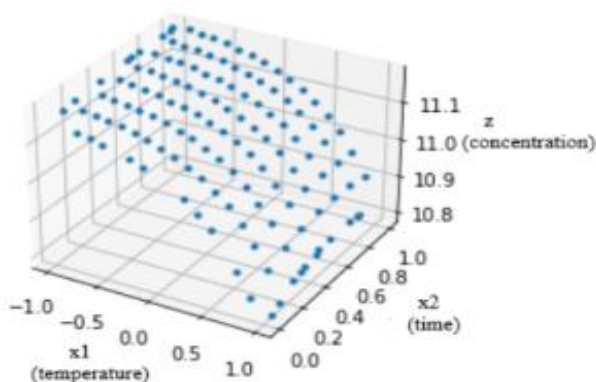
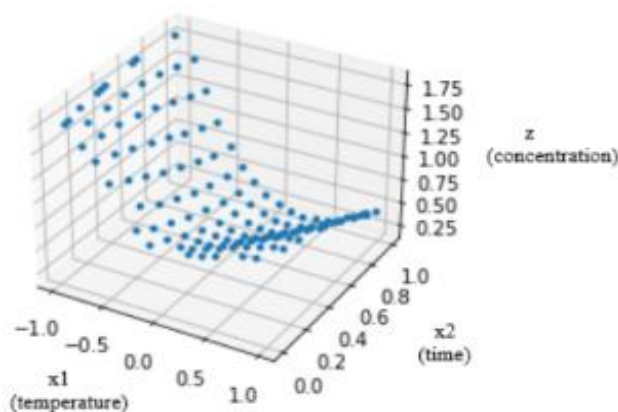Figure 2. Temperature approximation

Figure 3. Temperature and first order approximation

Figure 3 shown temperature and first order approximation contrasts markedly with the initial observations, offering a deeper dive into the interplay between temperature, time, and concentration changes. This graph delineates a more pronounced temperature influence on concentration changes, showcasing a steeper, more direct response of the reaction rate to temperature variations. The depicted dynamics underscore the critical impact of both the first-order kinetic equation and the temperature-dependent Arrhenius equation on the reaction's progression. The enhanced temperature sensitivity observed here aligns with common chemical reaction behavior, where increased temperature accelerates reaction rates through heightened molecular activity and the increased likelihood of collision events surpassing the activation energy barrier. Notably, the time-dependent variation in concentration changes suggests a gradual evolution of the reaction, with the initial stages showing less pronounced concentration changes even at higher temperatures. This pattern evolves as the reaction progresses, highlighting the diminishing concentrations of reactants over time due to ongoing consumption. The boundary and initial conditions established for this reaction model provide a foundational framework for understanding these dynamics. At the initial moment (time=0 $t=0$), irrespective of the temperature, the reactant concentration retains its original value, indicating the absence of reaction progress. At the minimum temperature ($x1=-1$), the reaction remains uninitiated, preserving the original concentration of reactants, reflective of an environment too cold to activate the reaction. Conversely, at the maximum temperature ($x1=1$), the reaction proceeds instantaneously, consuming

all reactants and reducing their concentration to zero, demonstrating the reaction's completion at elevated temperatures. This detailed analysis provided by Figure 3 enhances our comprehension of geopolymerization by elucidating the nuanced effects of temperature and time on reaction kinetics. The model unveils a more intricate picture of how geopolymer synthesis is influenced by these factors, offering valuable insights for optimizing production parameters. The identification of steeper temperature gradients within the reaction's progression not only allows for pinpointing optimal synthesis conditions but also highlights opportunities for energy optimization, underscoring the potential for more sustainable and efficient geopolymer production practices.

Progression towards optimizing the PINN for our geopolymerization reaction model is vividly illustrated in Figures 4 and 5, where we observe the training and testing loss metrics over numerous optimization steps. Figures 4 and 5 encapsulates the essence of a learning process reaching fruition. The consistent and linear reduction in both training and testing losses is a hallmark of the PINN's ability to bridge the gap between theoretical predictions and actual experimental data. This decline is emblematic of the network's increasing acumen in capturing the intricacies of geopolymerization kinetics, adjusting its internal parameters weights and biases to refine its predictive prowess. Figure 4 captures the journey of learning from the training data set. The decline in training loss signifies the model's growing competence in internalizing the kinetics it's being trained on, suggesting a robust learning curve not marred by stagnation or regression. Figure 5 extends this narrative to unseen data, showcasing the model's aptitude to apply learned principles beyond the confines of its training data. The reduction in testing loss corroborates the model's ability to generalize well, mitigating concerns of over-fitting and underscoring its adaptability. The convergence observed in these plots signals more than just a successful optimization it denotes the PINN's adherence to the fundamental physical laws that govern the geopolymerization process. The chosen optimization algorithms stand validated as "good optimizers," adept at navigating the complex parameter space to enhance both the accuracy and generalizability of the model's predictions. Crucially, the consistent alignment of the model with the predefined physical conditions reaffirms the PINN's capacity to not merely fit data but to do so in a manner that respects the scientific principles underlying geopolymer chemistry. This congruence between data-driven learning and physical law compliance reinforces the PINN's role as a reliable and insightful tool for dissecting and forecasting the dynamics of geopolymers, opening up new avenues for optimizing material compositions and reaction conditions in the pursuit of sustainable construction materials.
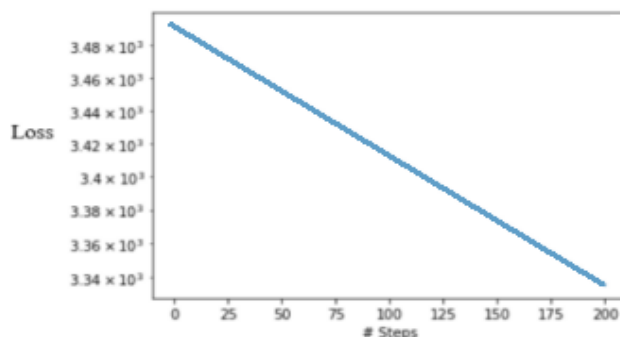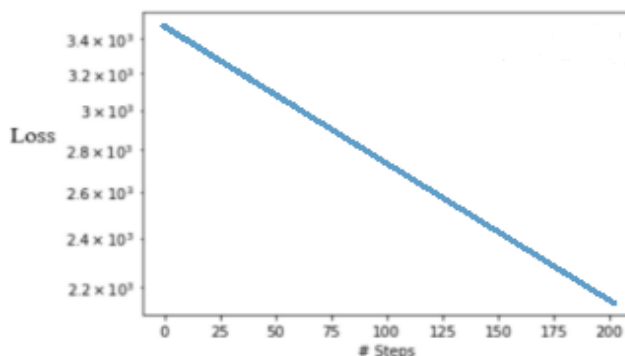

Figure 4. Training loss

Figure 5. Testing loss

Figure 6 introduces a 3D scatter plot that explores the interplay between temperature, time, and concentration change during geopolymerization. Unlike earlier stages focused on temperature's immediate effects, this figure suggests a transition to a phase where time becomes the dominant factor influencing reactant concentration. This shift indicates the reaction's progression into advanced stages, likely corresponding to curing or setting processes critical for the material's final properties. The plot implies that after the initial, temperature-sensitive phase, the reaction's kinetics are increasingly governed by time-dependent processes, such as diffusion or slow reactions among remaining reactants. If attributed to PINN modeling, the data distribution reflects an improved understanding of the geopolymerization dynamics, highlighting the model's enhanced predictive accuracy over time and across temperatures. This advancement is crucial for fine-tuning geopolymer synthesis, allowing for precise control over curing conditions and mix design to achieve desired material characteristics.

Figure 7 unveils a time-series plot that spans from 0 to 100 on the x-axis, with y-axis values ranging from -20 to 10, depicting oscillatory behavior potentially indicative of key reaction phenomena or model learning metrics in the geopolymerization process. The plot's initial high amplitude fluctuations, gradually tapering to more consistent, periodic oscillations, offer insight into the reactions or model's evolution over time. Should these oscillations correspond to reaction kinetics, they might reflect variable fluctuations such as temperature, pressure, or viscosity characteristic of transitioning from the highly reactive initial mixing phase to a more stable state as the reaction nears completion. This transition is critical for pinpointing optimal stages for intervention, adjustments, or curing in the geopolymer production process. Alternatively, if representing the PINN model's performance metrics, the decreasing amplitude in fluctuations could signify the model's progressive adaptation and refinement, achieving a stabilized understanding and prediction of the geopolymerization dynamics. This learning curve underscores the PINN's growing proficiency and its implications for optimizing the reaction conditions to enhance material properties and production efficiency. Figures thus encapsulate the complexity and progression of the geopolymerization process, whether through direct experimental observation or as a reflection of the computational model's optimization journey. This analysis is instrumental in advancing our understanding of geopolymer synthesis and the application of cutting-edge modeling techniques to drive innovations in sustainable construction materials.
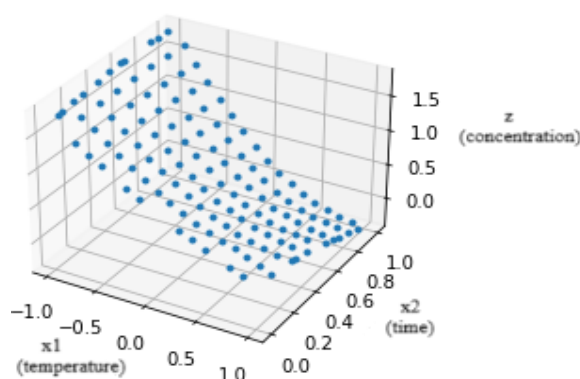

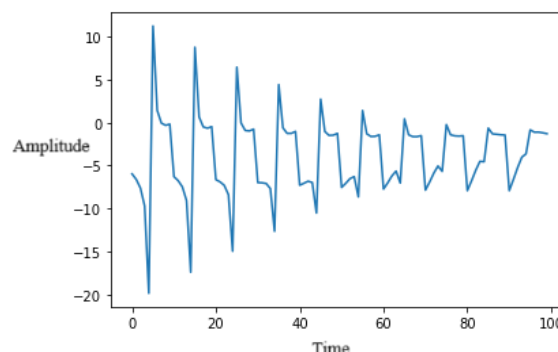Figure 6. Reaction kinetics in geopolymerization

Figure 7. Time-series analysis of geopolymerization

5. CONCLUSION

The comprehensive analysis of geopolymerization through both experimental investigation and advanced computational modeling has yielded significant insights into the reaction kinetics of this sustainable construction material. The experimental data has provided robust rate constants and Arrhenius parameters, which, when input into the PINN, allow for precise modeling of the reaction dynamics at various temperatures and over numerous time steps. The use of a PINN for this application goes beyond conventional optimization; it offers a predictive tool that utilizes the fundamentals of machine learning while incorporating the underlying physical laws that govern geopolymerization. By feeding the network accurate experimental data on concentration changes over time, the PINN becomes finely tuned to predict the reaction's behavior under different conditions. With these optimized models, the consumption of valuable and rare materials can be minimized by fine-tuning the reaction parameters. The ability to adjust temperatures to influence the reaction kinetics opens the door to more efficient use of resources and a reduction in the costs associated with geopolymer production. Furthermore, the neural network model can assist in identifying the optimal temperature range for geopolymer synthesis, along with suitable catalysts, by leveraging its understanding of

the activation energy involved in the process. Such advancements facilitated by PINN could significantly enhance the production scale and quality of geopolymers. By providing a clearer understanding of the reaction mechanisms, PINNs can help in designing geopolymer mixtures that are not only cost-effective but also possess the desired mechanical and durability properties. The integration of machine learning with traditional experimental techniques marks a leap forward in the field of materials science, offering a powerful approach to tackling the challenges associated with the production of geopolymers and, more broadly, advancing the technology of sustainable materials.

AUTHOR CONTRIBUTIONS STATEMENT

Name of Author	C	M	So	Va	Fo	I	R	D	O	E	Vi	Su	P	Fu
Blesso Abraham	✓	✓	✓	✓	✓	✓		✓	✓	✓	✓			
Thirumalaivasal Devanathan Sudhakar						✓				✓		✓	✓	

C	:	**C**onceptualization	I	:	**I**nvestigation	
M	:	**M**ethodology	R	:	**R**esources	
So	:	**So**ftware	D	:	**D**ata Curation	
Va	:	**Va**lidation	O	:	Writing - **O**riginal Draft	
Fo	:	**Fo**rmal analysis	E	:	Writing - Review & **E**diting	

Vi : **Vi**sualization
Su : **Su**pervision
P　: **P**roject administration
Fu : **Fu**nding acquisition

CONFLICT OF INTEREST STATEMENT
Authors state no conflict of interest.

DATA AVAILABILITY
The input data is taken from the boundary conditions of the differential equations. The output data of six graphs from Figure 2 to Figure 7 was created.

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