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Ultrafast Diffusion Modeling via the Riemann–Liouville Nonlocal Structural Derivative and Its Application in Porous Media

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Abstract: Ultrafast diffusion disperses faster than super-diffusion, and this has been proven by several theoretical and experimental investigations. The mean square displacement of ultrafast diffusion grows exponentially, which provides a significant challenge for modeling. Due to the inhomogeneity, nonlinear interactions, and high porosity of cement materials, the motion of particles on their surfaces satisfies the conditions for ultrafast diffusion. The investigation of the diffusion behavior in cementitious materials is crucial for predicting the mechanical properties of cement. In this study, we first attempted to investigate the dynamic of ultrafast diffusion in cementitious materials underlying the Riemann–Liouville nonlocal structural derivative. We constructed a Riemann–Liouville nonlocal structural derivative ultrafast diffusion model with an exponential function and then extended the modeling strategy using the Mittag–Leffler function. The mean square displacement is analogous to the integral of the corresponding structural derivative, providing a reference standard for the selection of structural functions in practical applications. Based on experimental data on cement mortar, the accuracy of the Riemann–Liouville nonlocal structural derivative ultrafast diffusion model was verified. Compared to the power law diffusion and the exponential law diffusion, the mean square displacement with respect to the Mittag–Leffler law is closely tied to the actual data. The modeling approach based on the Riemann–Liouville nonlocal structural derivative provides an efficient tool for depicting ultrafast diffusion in porous media.

Keywords: ultrafast diffusion; nonlocal structural derivative; structural function; mean square displacement; porous media

Citation: Xu, W.; Liu, H.; Chen, L.; Zhou, Y. Ultrafast Diffusion Modeling via the Riemann–Liouville Nonlocal Structural Derivative and Its Application in Porous Media. *Fractal Fract.* **2024**, *8*, 110. https://doi.org/10.3390/fractalfract8020110

Academic Editors: Xiaoli Chen, Dongfang Li, Ting Gao and Ivanka Stamova

Received: 31 December 2023
Revised: 6 February 2024
Accepted: 9 February 2024
Published: 12 February 2024

1. Introduction

In the fields of natural science and engineering, the classification of diffusion phenomena primarily relies on the growth of the mean squared displacement (MSD) over time. The normal diffusion process is based on the classic Brownian motion theory, in which the MSD of a particle increases linearly with time [1]. A large number of experiments have shown that anomalous diffusion is ubiquitous in complex systems such as physical, chemical, and biological systems [2]. The main characteristic of anomalous diffusion is that the corresponding MSD grows as a power law: $\langle x^2(t) \rangle \sim t^{\eta}$ [3], with $\eta > 1$, is used for characterizing super-diffusion, and $\eta < 1$ can describe sub-diffusion. Ultrafast diffusion disperses faster than super-diffusion, which has been proven by several theoretical and experimental investigations [4–6]. Unlike anomalous diffusion, the MSD of ultrafast diffusion increases exponentially over time, i.e., $\langle x^2(t) \rangle \sim e^t$, posing a huge challenge to the existing anomalous diffusion models [5]. From the time perspective, ultrafast diffusion generally occurs quickly in the initial stage, characterizing the short-term evolution process of particles. As a typical nonlocal and non-Markov process, ultrafast diffusion is influenced by the external environment and internal structural characteristics including strong hetero-

geneity, the dominant channels of the medium structure, and the mechanical properties of particles [6].

Cement mortar plays an important role in the construction industry, in transportation, in water conservation, and in other fields [7,8]. In order to enhance the strength, durability, and hydration process of cementitious mortars, it is customary to introduce various additives into their matrices, such as a superplasticizer and magnesium phosphate [9,10]. The incorporation of these additives can lead to the formation of various pores and cracks in the internal microstructure [11]. Due to inhomogeneous and nonlinear interactions, the motion of particles in porous media usually exhibits anomalous diffusion, which does not satisfy the classic Fick law [12]. Even the dynamic behavior of certain particles in cement mortar conforms to ultrafast diffusion [13,14]. Although the diffusion process of suspended particulates in cement is difficult to observe, a microrheology analyzer can measure the MSD of cement particles based on laser scattering [15]. At present, this process is mainly performed using theoretical or empirical models. So, the investigation of the ultrafast diffusion behavior in cementitious materials is crucial for predicting the long-term performance of concrete structures.

The key to analyzing the dynamic behavior of a particle is to establish a reasonable mechanical model. The fractional derivative and fractal derivative have been successfully applied to capture sub-diffusion behaviors and super-diffusion behaviors [16–18]. Fomin et al. employed the fractional derivative diffusion equations to model the anomalous diffusion in cracked rock mass with altered crack regions [19]. Ref. [20] reported that the fractal derivative diffusion model is suitable for simulating anomalies of moisture transport in cement-based materials. Furthermore, continuous-time random walk (CTRW) models have been confirmed to be able to explain anomalous transport in three-dimensional porous media with different cementation degrees [21]. These models use fractal geometry to describe the irregularities in a porous medium. However, they cannot accurately explain the ultrafast diffusion process wherein the MSD does not conform to the power-law characteristics. To solve such problems, O'Malley et al. proposed a statistical model of fractional Brownian motion run with a nonlinear clock [22]. Xu et al. presented a spatial local structural derivative diffusion model to describe the motion of nanoparticles suspended in polymer solutions [23]. However, the understanding of nonlocality in ultrafast diffusion is not yet fully developed. The primary challenge in this regard is determining how to deduce the properties of the nonlinear functions based on dynamic processes.

The structural derivatives, including the local operator and the nonlocal operator, were proposed by Chen et al. [24]. The local structural derivative and the nonlocal structural derivative are extensions of the Hausdorff derivative and the fractional derivative, respectively. In comparison with the classical derivatives, the structural derivatives are a generalization of the space-time fractal power law scale. Usually, structural functions are used to determine the quantitative relationship between the time scale or the medium structure and some physical parameters of mathematical mechanical models [4]. At the same time, the structural function plays a leading role in determining the form and properties of the structural derivative and is suitable for describing dynamic processes with a memory effect [25]. Moreover, the fundamental solution of the corresponding structural derivative diffusion equation can reflect the statistical law of diffusion [23]. At present, theoretical models and numerical algorithms of the structural derivatives have been successfully used to describe various complex mechanical problems. The nonlocal structural derivative diffusion model with the inverse Mittag–Leffler (ML) function has been applied to simulate ultraslow diffusion in dense colloids over long time scales [25].

The geometric foundation of non-local structural derivatives is rooted in Euclidean distance. This framework can capture local features under non-uniform scales and with long-memory properties [4]. Additionally, by implementing structural functions (arbitrary functions of space time), the research scope extends to more intricate structural systems. Given the intricacies of the computational process and the challenging nature of modeling, the research on nonlocal structural derivatives primarily focuses on exploring the time-

dependent stochastic processes. At present, the Riemann–Liouville nonlocal structural derivative has gained attention for modeling the ultraslow creep of non-Newtonian fluids [26,27]. But the application of cementitious materials is in its nascent stages. This study concentrates on investigating ultrafast diffusion in a cement system and further analyzing the influential mechanism underlying the Riemann–Liouville nonlocal structural derivative. As in the generation of the Riemann–Liouville fractional derivative, the Riemann–Liouville nonlocal structural derivative retains the convolution operator for describing the memory and widens the prediction and simulation range through its kernel function or structural function [24]. In this study, we constructed a Riemann–Liouville nonlocal structural derivative ultrafast diffusion model with an exponential function and extended the modeling strategy through the ML function to analyze the mechanical properties of cement. The accuracy of the model is substantiated via a test conducted on magnesium phosphate cement mortar [28].

The subsequent sections of this study are structured as follows. In Section 2, we provide the Riemann–Liouville nonlocal structural derivative and propose a Riemann–Liouville nonlocal structural derivative ultrafast diffusion model with an exponential function and an ML function. In Section 3, the dynamics processes of several particles in cement materials are simulated using the proposed model. Some discussion is provided in Section 4. Section 5 summarizes the conclusions.

2. Theory

2.1. The Riemann–Liouville Nonlocal Structural Derivative

Traditional integer-order derivatives are limited to characterizing the local diffusion effects of particles. Non-integer-order derivatives can capture non-local properties, long-range memory, and the non-stationary evolution of statistical characteristics over time [16]. The most common non-integer-order derivatives are fractal derivatives and fractional derivatives; the Riemann–Liouville nonlocal structural derivative can be regarded as an extension of the Riemann–Liouville fractional derivative. If $P(x,t)$ is a continuous derivable function in $(0,+\infty)$, the Riemann–Liouville nonlocal structural derivative can be defined as follows [25]:

$$ {}_0^{RL}D_t^{1-K}P(x,t) = \frac{\partial}{\partial t}\int_0^t K(t-\tau)\cdot P(x,\tau)\mathrm{d}\tau. \tag{1}$$

Here, ${}_0^{RL}D_t^{1-K}$ is the time Riemann–Liouville nonlocal structural derivative operator; the time structural function $K(t)$ is arbitrary. When $K(t)$ is a power-law function,

$$K(t) = \frac{t^{\alpha-1}}{\Gamma(\alpha)}, \tag{2}$$

Equation (1) can be transformed into the classical Riemann–Liouville fractional derivative [10]:

$$ {}_0^{RL}D_t^{1-K}P(x,t) = {}_0^{RL}D_t^{1-\alpha}P(x,t) = \frac{1}{\Gamma(\alpha)}\frac{\partial}{\partial t}\int_0^t (t-\tau)^{\alpha-1}\cdot P(x,\tau)\mathrm{d}\tau, \tag{3}$$

where ${}_0^{RL}D_t^{1-\alpha}$ is the Riemann–Liouville fractional derivative operator. It is worth noting that the dynamic behavior of different media structures can be determined by transforming the structural function. For complex systems with extremely irregular structures, the structural metric can be used to effectively describe diffusion mechanisms and media complexity, which can be defined as

$$K_s = |K(t_2-t_0) - K(t_1-t_0)|, \tag{4}$$

where K_s denotes structural time. The time local structural derivative can be derived from the velocity of a moving particle in structural time:

$$\frac{\partial P(x,t)}{\partial K(t)} = \lim_{t_1 \to t} \frac{P(x,t_1) - P(x,t)}{K(t_1) - K(t)}. \tag{5}$$

Compared with the Riemann–Liouville nonlocal structural derivative, the local structural derivative of Equation (5) is based on non-Euclidean distances, and the operator itself does not have memory.

2.2. The Ultrafast Diffusion Model via Riemann–Liouville Nonlocal Structural Derivative

As an effective method for simulating non-Markovian processes in complex systems, the Riemann–Liouville fractional derivative anomalous diffusion equation over time can be directly derived using a CTRW model [18]. Its mathematical expression consists of replacing the traditional first-order derivative with the fractional derivative, as follows [3]:

$$\frac{\partial P(x,t)}{\partial t} = {}_0^{RL}D_t^{1-\alpha} D_s \frac{\partial^2 P(x,t)}{\partial x^2}. \tag{6}$$

Here, D_s is known as a generalized diffusion coefficient and its dimensions depend on the structural function. ${}_0^{RL}D_t^{1-\alpha}$ is the Riemann–Liouville fractional derivative in Equation (3). $P(x,t)$ denotes the probability density function (PDF) of concentration in x at time t. Using the integral operation $\int_{-\infty}^{+\infty} x^2 \mathrm{d}x$, Equation (6) can be transformed into

$$\frac{\partial}{\partial t}\int_{-\infty}^{+\infty} P(x,t) \cdot x^2 \mathrm{d}x = {}_0^{RL}D_t^{1-\alpha} D_s \int_{-\infty}^{+\infty} \frac{\partial^2 P(x,t)}{\partial x^2} \cdot x^2 \mathrm{d}x. \tag{7}$$

Through derivation, Equation (7) can be transformed into [18]

$$\frac{\mathrm{d}}{\mathrm{d}t}\left\langle x^2(t) \right\rangle = 2{}_0^{RL}D_t^{1-\alpha} D_s. \tag{8}$$

Then, substituting Equation (3) into Equation (8), we can obtain

$$\frac{\mathrm{d}}{\mathrm{d}t}\left\langle x^2(t) \right\rangle = \frac{2D_s}{\Gamma(\alpha)} t^{\alpha-1}. \tag{9}$$

The corresponding MSD can be acquired through the integration of the variable t, which increases the power law.

Considering the propensity of the MSD to undergo ultrafast diffusion in cement materials, we suppose the time structural function is an exponential function:

$$K(t) = \lambda e^{\lambda t},\ \lambda > 0. \tag{10}$$

The corresponding Riemann–Liouville nonlocal structural derivative is

$${}_0^{RL}D_t^{1-K} U(x,t) = \frac{\partial}{\partial t}\int_0^t \lambda e^{\lambda(t-\tau)} \cdot U(x,\tau)\mathrm{d}\tau. \tag{11}$$

Similar to the properties of the fractional derivative, Equation (10) obeys the following relationship: ${}_0^{RL}D_t^{1-K} = \frac{\partial}{\partial t}{}_0^{RL}D_t^{-K}$. According to the theoretical analysis of the Riemann–Liouville fractional derivative diffusion model, the time Riemann–Liouville nonlocal structural derivative ultrafast diffusion model in Equation (11) is

$$\frac{\partial U(x,t)}{\partial t} = {}_0^{RL}D_t^{1-K} D_s \frac{\partial^2 U(x,t)}{\partial x^2}, \tag{12}$$

where $U(x,t)$ denotes the PDF of the particles in cement materials at x at time t. After applying the integration operator $\int_{-\infty}^{+\infty} x^2 \mathrm{d}x$ to both sides of Equation (12), we obtain

$$\int_{-\infty}^{+\infty} x^2 \cdot \frac{\partial U(x,t)}{\partial t} \mathrm{d}x = \int_{-\infty}^{+\infty} x^2 \cdot {}_0^{RL}D_t^{1-K} D_s \frac{\partial^2 U(x,t)}{\partial x^2} \mathrm{d}x. \tag{13}$$

Based on the properties of calculus, Equation (13) can be rewritten as follows:

$$\frac{\partial}{\partial t} \int_{-\infty}^{+\infty} x^2 \cdot U(x,t) \mathrm{d}x = \int_{-\infty}^{+\infty} x^2 \cdot {}_0^{RL}D_t^{1-K} D_s \frac{\partial^2 U(x,t)}{\partial x^2} \mathrm{d}x. \tag{14}$$

Because the definition of the MSD is

$$\left\langle x^2(t) \right\rangle = \int_{-\infty}^{+\infty} x^2 \cdot U(x,t) \mathrm{d}x, \tag{15}$$

Equation (14) becomes

$$\frac{\mathrm{d}\left\langle x^2(t) \right\rangle}{\mathrm{d}t} = {}_0^{RL}D_t^{1-K} 2D_s. \tag{16}$$

Due to integration with respect to t the value of the MSD at $t = 0$ needs to be 0. So, the MSD can be derived based on Equation (15):

$$\left\langle x^2(t) \right\rangle = 2D_s \int_0^t \lambda e^{\lambda(t-\tau)} \mathrm{d}\tau = e^{\lambda t} - 1. \tag{17}$$

Indeed, the relationship between $\left\langle x^2(t) \right\rangle$ and $K(t)$ can also be inferred from Equation (15), leading to

$$\left\langle x^2(t) \right\rangle = 2D_s \int_0^t K(\tau)\, \mathrm{d}\tau. \tag{18}$$

The ML function is also commonly used to analyze the properties of fractional integral equations and fractional derivative equations [29]. Recent studies indicate that the ML function and its inverse function have great application potential in physics, biology, and engineering, especially in regard to anomalous diffusion for complex systems [29,30]. Ref. [30] analyzed ultrafast diffusion using the fractional Brownian motion run with the ML clock; the MSD was calculated as:

$$\left\langle x^2(t) \right\rangle = E_\alpha(t) = \sum_{k=0}^{+\infty} \frac{t^k}{\Gamma(\alpha k + 1)}. \tag{19}$$

In order to increase the applicability of the model in complex practical systems, we assume that the structural function is mainly composed of an ML function

$$K(t) = \eta [E_\alpha(t)]^{\eta - 1} E_\alpha{}'(t). \tag{20}$$

where $E_\alpha{}'(t)$ is the first-order derivative of the ML function. Then, the Riemann–Liouville nonlocal structural derivative ultrafast diffusion model with an ML function can be established. Combining Equations (14) and (16), the MSD is an ML function that satisfies the following form:

$$\left\langle x^2(t) \right\rangle = [E_\alpha(t)]^\eta, \eta > 0. \tag{21}$$

Overall, the structural function can be seen as a memory kernel that is related to complex medium structures, and it mainly depends on the characteristics of dynamic behavior. Equation (17) directly indicates the equivalence of the MSD and the integration of a structural function over t. This law will provide a reference standard for the selection of structural functions in practical applications.

3. Applications and Results

In the work above, Riemann–Liouville nonlocal structural derivative ultrafast diffusion models were established theoretically, but further research is needed to validate their effectiveness in capturing the intricacies of particle transport. This section focuses on exploring the accuracy of the models based on experimental data on magnesium phosphate cement mortar [28]. The corresponding parameters and fitting errors of the models were estimated through nonlinear least squares fitting, and the MATLAB simulation analysis tool was used to achieve this.

Magnesium phosphate cement (MPC) mortar, also known as phosphate chemical ceramic bonding material, is one of the hotspots in the research on new cementitious materials [31]. It has very good application prospects regarding building repair engineering materials, the solidification of heavy metals and pollutants, refractory materials, and biomedical engineering materials [32,33]. To investigate the mechanical properties and water resistance of MPC mortar, Hou et al. prepared magnesium sodium phosphate cement and magnesium potassium phosphate cement and then analyzed the microstructure of their hydration product, struvite-K [28]. The experimental results indicated that the compressive strength of MPC mortar can be evaluated by referring to the mobility of different atoms in struvite-K. Expressed with dual logarithm coordinates, the MSD of oxygen atoms in water and K ions for the struvite-K near-water solution does not follow a linear relationship with time but shows an exponential growth trend. This result could potentially provide insights into the pronounced cracking observed for the MPC paste immersed in the water solution. Further elaboration on the experiments and the corresponding experimental data can be found in Ref. [28].

In order to test the feasibility of the Riemann–Liouville nonlocal structural derivative ultrafast diffusion model, the experimental data characterizing the MSD of particles in cement mortar were simulated using the power law diffusion with $\eta > 1$, the exponential law diffusion in Equation (16), and the Mittag–Leffler law diffusion in Equation (19). Figures 1 and 2 describe the fitting results for different models when simulating the experimental data of the MSD for the oxygen atoms and the K ions on MPC, respectively. The black filled circles are the experimental data measured in cement mortar; the red solid line is the fitting result of the Riemann–Liouville nonlocal structural derivative diffusion model with the ML function in Equation (19), $\langle x^2(t) \rangle \sim [E_\alpha(t)]^\eta$; the blue line is the fitting result of the exponential law diffusion in Equation (16), $\langle x^2(t) \rangle \sim e^{\lambda t} - 1$; and the pink dashed line is the fitting result of the power law model, $\langle x^2(t) \rangle \sim t^\eta$. Tables 1 and 2 provide the corresponding parameters and the fitting errors of the models.

The observed data indicate the validity of all the diffusion laws as temporal progression occurs. Particularly within a limited time range, the curves of the ML diffusion law adeptly capture the experimental data in Figure 1. The maximum absolute and mean squared errors of the Riemann–Liouville nonlocal structural derivative ultrafast diffusion model with ML diffusion in Table 1 are less than those of the other diffusion laws. At the same time, there is little difference in the parameters of the three models. It can be observed from Figure 2 that the overall trend of the experimental data is more consistent with ultrafast diffusion, but the accuracy of the models is difficult to evaluate in the initial stage. With the passage of time, the Riemann–Liouville nonlocal structural derivative diffusion model with the ML function is gradually highlighted. The errors in Table 2 can also reflect this point very well. In fact, the phenomenon of ultrafast diffusion occurs quickly and unstably, so it is difficult to capture the relevant experimental data. This is perhaps why the experimental data cannot match the model perfectly in Figure 2.

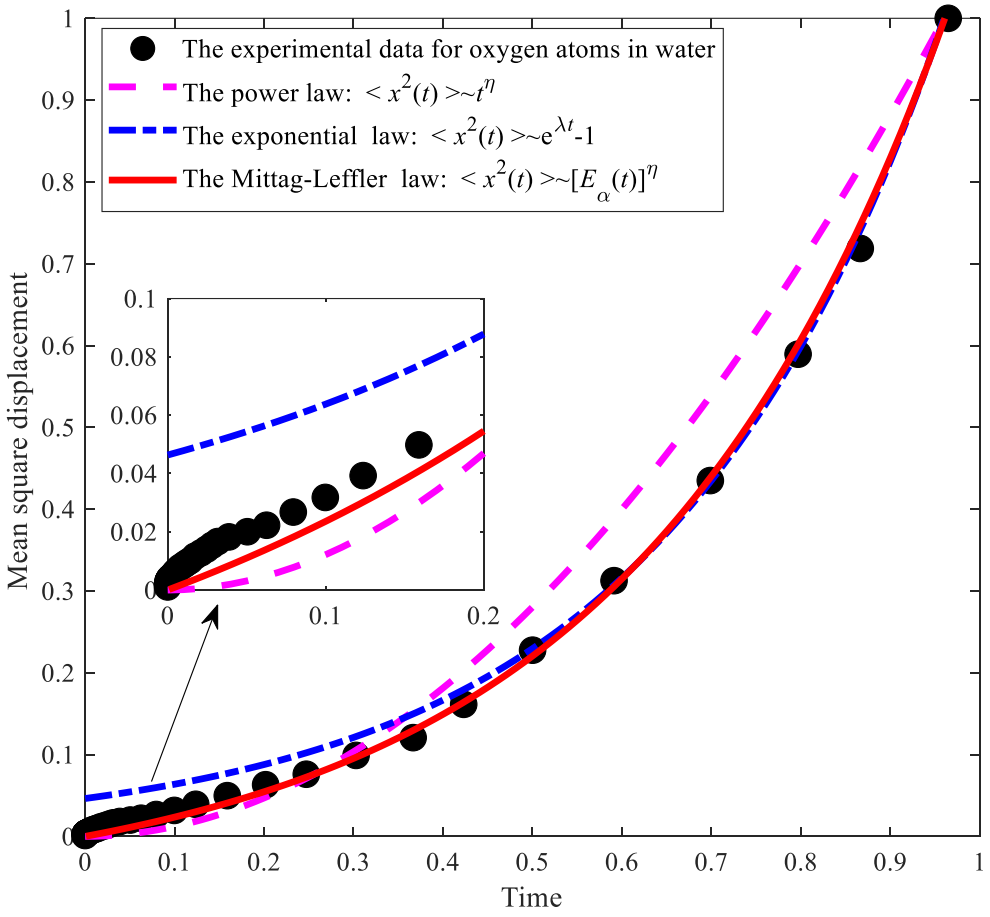

Figure 1. Fitting results regarding power law diffusion ($\langle x^2(t)\rangle \sim t^{\eta}$, $\eta = 1.95$), exponential law diffusion ($\langle x^2(t)\rangle \sim e^{\lambda t} - 1$, $\lambda = 2.2$), and Mittag–Leffler law diffusion ($\langle x^2(t)\rangle \sim [E_{\alpha}(t)]^{\eta}$, $\alpha = 0.95$, $\eta = 2$) for the MSD of oxygen atoms in water. The experimental data were sourced from [28].

Table 1. The fitting results for oxygen atoms in water for different models and their errors (the diffusion coefficient is dimensionless).

Models	Power Law Diffusion	Exponential Law Diffusion	Mittag–Leffler Law Diffusion
η	$\eta = 1.95$		$\eta = 2$
λ		$\lambda = 2.2$	
α			$\alpha = 0.95$
Maximum absolute error	0.0995	0.0446	0.0201
Mean square error	0.0314	0.0341	0.0017

Table 2. The fitting results for K ions from different models and their errors (the diffusion coefficient is dimensionless).

Models	Power Law Diffusion	Exponential Law Diffusion	Mittag–Leffler Law Diffusion
η	$\eta = 1.8$		$\eta = 2.5$
λ		$\lambda = 2.7$	
α			$\alpha = 0.98$
Maximum absolute error	0.0671	0.0567	0.0418
Mean square error	0.0390	0.0371	0.0252

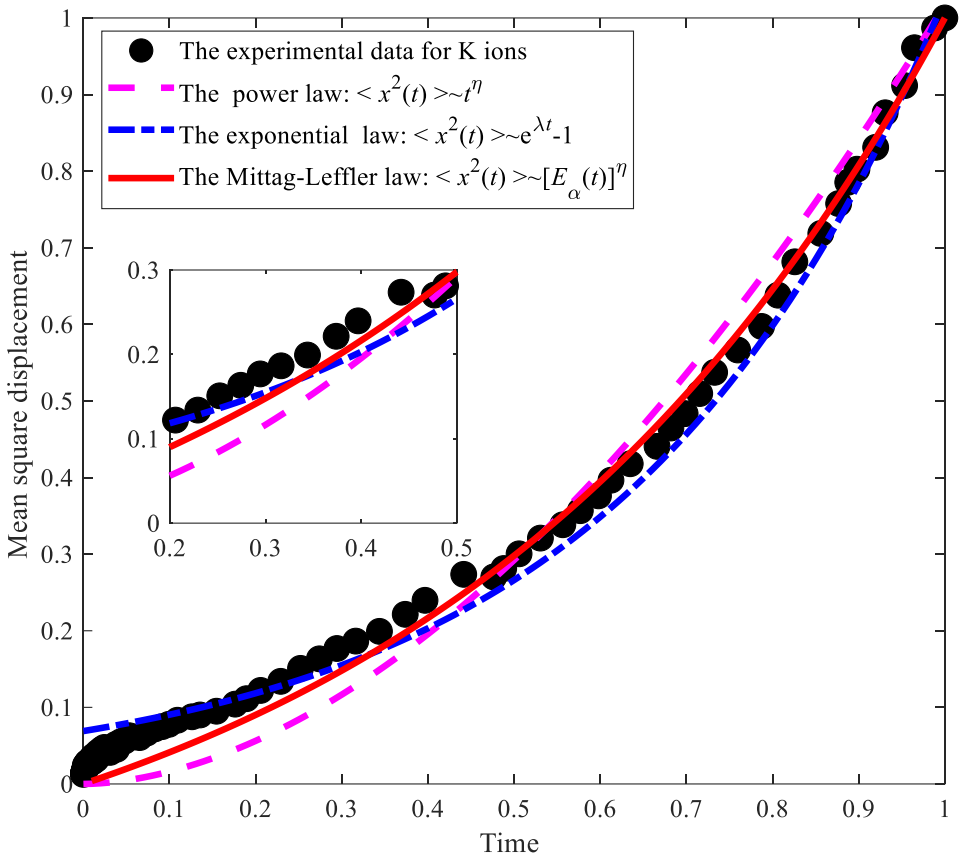

Figure 2. Fitting results regarding super-diffusion power law diffusion ($\langle x^2(t)\rangle \sim t^{\eta}$, $\eta = 1.8$), exponential diffusion law diffusion ($\langle x^2(t)\rangle \sim e^{\lambda t} - 1$, $\lambda = 2.7$), and Mittag–Leffler law diffusion ($\langle x^2(t)\rangle \sim [E_{\alpha}(t)]^{\eta}$, $\alpha = 0.98$, $\eta = 2.5$) for the MSD of K ions. The experimental data were sourced from [28].

4. Discussion

In this study, the dynamics of ultrafast diffusion in cementitious materials underlying the Riemann–Liouville nonlocal structural derivative diffusion model were investigated. Similar to the fractional-order derivatives, the analytical solution of the Riemann–Liouville nonlocal structural derivative diffusion model is difficult to deduce due to the convolution operators. The numerical solutions of the fractional differential equations are still important topics [34]. For the same reason, there is little theoretical research on spatial Riemann–Liouville nonlocal structural derivatives. Even if the analytical solution can be obtained, the form of the solution is often a complex special function or an infinite series, which are difficult to directly apply in practical applications. Therefore, finding an effective and high-precision numerical solution method has a strong effect on the practical application of Riemann–Liouville nonlocal structural derivative equations.

Significantly, the macroscopic constitutive model based on a differential equation is empirical, which is helpful for understanding and predicting the dynamic processes of complex systems. From a statistical mechanics point of view, the microscopic statistical models can accurately describe the transitions, collisions, and other behaviors of particles between different energy levels based on their behavior and thus reveal the corresponding laws and properties of motion [30]. The continuous-time random walk model provides a framework for describing the anomalous diffusion of particles and establishes a strong relationship between the microscopic mechanism and macroscopic equations [18]. The focus of our future work will be to analyze the statistical characteristics of the waiting time and jump length of ultrafast diffusion and then explain the microscopic mechanism of particle diffusion.

This study verifies the effectiveness of the Riemann–Liouville nonlocal structural derivative ultrafast diffusion model on the basis of analyzing the theories and experimental results. The model is in a simple form and is convenient to program without considering the effects of convection and adsorption. However, the physical significance of the parameters and the relationship between the structural function and the medium structure are still undetermined. Experimental data are direct evidence that can be used to determine the physical meaning of parameters. The results of the comparison between experiments and models will provide important clues for the accuracy evaluation and parameter optimization of the model. In future studies, the physical meanings of parameters and the applications for the Riemann–Liouville nonlocal structural derivative diffusion model will be explored with more practical applications.

5. Conclusions

In this study, we investigated the macroscopic mechanical constitutive models for the ultrafast diffusion of particles in cement paste and qualitatively analyzed the influence mechanism. The following conclusions can be drawn from the preceding results and discussions:

1. The Riemann–Liouville nonlocal structural derivative diffusion equation has the advantages of strong application potential, high flexibility, and a mean square displacement that is analogous to the integral form of the corresponding structural function.
2. The physical mechanism of the Riemann–Liouville nonlocal structural derivative diffusion model is obvious, and the law of the ultrafast diffusion in cement mortar over a short time scale satisfies the inverse Mittag–Leffler function.
3. The verification of the experimental results indicated that the Riemann–Liouville nonlocal structural derivative diffusion model is more effective at depicting ultrafast diffusion behavior in cement mortar, as evidenced by the fitting curves and errors.

Author Contributions: Conceptualization, W.X. and L.C.; methodology, W.X., H.L., and Y.Z.; software, W.X. and H.L.; validation, W.X. and H.L.; formal analysis, W.X., H.L., L.C., and Y.Z.; investigation, W.X., H.L., L.C., and Y.Z.; resources, W.X. and Y.Z.; data curation, W.X., H.L., L.C., and Y.Z.; writing—original draft preparation, W.X., H.L., and Y.Z.; writing—review and editing, W.X., H.L., L.C., and Y.Z.; visualization, W.X. and H.L.; supervision, W.X. and L.C.; project administration, W.X.; funding acquisition, W.X. and Y.Z. All authors have read and agreed to the published version of the manuscript.

Funding: The work described in this paper was supported by the Natural Science Foundation of Shandong Province (No. ZR2021QA042), part of the National Natural Science Foundation of China (Nos. 12202225, 12101037).

Data Availability Statement: The data presented in this study are not available due to privacy concerns.

Conflicts of Interest: The authors declare no conflicts of interest.

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