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A Novel Method for Curvefitting the Stretched Exponential Function to Experimental Data

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Abstract

The stretched exponential function has many applications in modeling numerous types of experimental relaxation data. However, problems arise when using standard algorithms to fit this function: we have observed that different initializations result in distinct fitted parameters. To avoid this problem, we developed a novel algorithm for fitting the stretched exponential model to relaxation data. This method is advantageous both because it requires only a single adjustable parameter and because it does not require initialization in the solution space. We tested this method on simulated data and experimental stress-relaxation data from bone and cartilage and found favorable results compared to a commonly-used Quasi-Newton method. For the simulated data, strong correlations were found between the simulated and fitted parameters suggesting that this method can accurately determine stretched exponential parameters. When this method was tested on experimental data, high quality fits were observed for both bone and cartilage stress-relaxation data that were significantly better than those determined with the Quasi-Newton algorithm.

Keywords

Cartilage Biomechanics; Osteoarthritis; Curvefitting; Optimization; Polymer Dynamics

I. INTRODUCTION

Relaxation experiments are widely used across multiple scientific and engineering disciplines. Examples include dielectric relaxation, for characterization of dielectric responses to external electric fields [1], nuclear magnetic relaxation, used to characterize the molecular mobility of paramagnetic atomic nuclei [2], and stress-relaxation, which characterizes the response of a viscoelastic material to a rapid deformation [3].

Interpreting relaxation data involves fitting mathematical models to the experimental data to determine how the model parameters vary between experimental conditions. Therefore, curvefitting the model to the experimental data is of fundamental importance for deriving meaningful model parameters toward improved understanding of physical systems. Many different models have been used to fit relaxation data including various mono and multi-exponential functions [4–9]. The stretched exponential function (Equation 1) is a common

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model for describing relaxation data. This model has been used extensively to represent the stress-relaxation of biological tissues and other systems [10–14].

The stretched exponential model presents significant challenges for standard curve fitting methods, including redundant fits (Figure 1). The present study was motivated by these fitting difficulties which present a challenge to the interpretation of relaxation data modeled by the stretched exponential model. Our objective was to develop a method for stretched exponential fitting that improves the aforementioned difficulties.

II. METHODS

A. Stretched Exponential Model and Optimization Framework

The stretched exponential function is a model for relaxation of complex systems (Equation 1) [15, 16]:

$$\sigma = Ae^{-\left(\frac{t}{\tau}\right)^\beta} + B \quad (1)$$

σ represents the temporally-measured dependent variable (*e.g.* stress in a stress-relaxation experiment). This model has four parameters, A , B , τ , and β , which can be adjusted to fit relaxation data [17]. The parameters B and A represent the equilibrium and peak beyond equilibrium values, respectively. τ is the time constant of relaxation of the system. β ($0 < \beta < 1$) is the stretching parameter. For polymer systems, the time constant can be derived from polymer statistical physics, and the stretched exponential form represents polydisperse collections of polymers such as cartilage [15, 16, 18].

Ideally, we want to minimize the weighted and squared L^2 norm (wL^2) between the data and the model (Equation 2) with respect to the model parameters.

$$wL^2(A, \tau, \beta, B) = \sum_i w_i (d_i - \sigma_i)^2 \quad (2)$$

In Equation 2, w represents the weight, d the data, and σ the model. The index i represents each discrete timepoint at which data was sampled. First, we note that this function is not convex in the parameter set, and empirically we have found this parameter set to have multiple local minima [19]. Both of these facts complicate fitting substantially.

B. Data Transformation and Novel Fitting Algorithm

Toward addressing the fitting complications, the relaxation data were transformed by first subtracting the measured equilibrium value, σ_{eq} , followed by division by the difference between the peak and equilibrium values, $\sigma_{peak} - \sigma_{eq}$. Note that these values allow direct calculation of model parameters A and B . For testing, the equilibrium value was estimated from the average of the last 0.25 seconds of data and the peak value from the largest stress value. We call the transformed data \hat{d}_i (Figure 2, Equation 3):

$$\hat{d}_i = \frac{d_i - \sigma_{eq}}{\sigma_{peak} - \sigma_{eq}} \quad (3)$$

Our goal becomes to find the values of τ and β that best fit the transformed data using the transformed model ($\hat{\sigma}$, Equation 4):

$$\hat{\sigma} = e^{-\left(\frac{t}{\tau}\right)^\beta} \quad (4)$$

We applied an additional constraint that required the area under the curve of the transformed data to equal the area under the transformed model. Experimental noise is usually symmetrically-distributed (data not shown), so this is a logical constraint. We defined \hat{D} as the area under the transformed data and $\widehat{\Sigma}$ as the area under the transformed model. $\widehat{\Sigma}$ was calculated in closed form (Equation 5), as

$$\widehat{\Sigma} = \int \hat{\sigma}(t) dt = \frac{\tau}{\beta} \Gamma\left(\frac{1}{\beta}\right) \widehat{\Sigma} = \frac{\tau}{\beta} \Gamma\left(\frac{1}{\beta}\right) \quad (5)$$

where Γ is the gamma function. \hat{D} was estimated from the data using the trapezoidal rule [20]. Applying the area constraint, $\hat{D} = \widehat{\Sigma}$, allows us to solve Equation 5 for τ , substitution of which into Equation 6 yields a one-dimensional Γ optimization problem, with the single free parameter β .

$$\tau = \frac{\beta \hat{D}}{\Gamma\left(\frac{1}{\beta}\right)} \quad (6)$$

We minimized the squared L^2 norm of the weighted residuals between the transformed data and the model (Equation 7):

$$wL^2(\beta) = \sum_{i=1} w_i [\hat{d}_i - \hat{\sigma}_i(\beta)]^2 \quad (7)$$

Weights, w_i , were determined using a standard method by dividing the stress at each timepoint by the signal variance, which provided a method for minimizing the potentially dominant influence of datapoints collected during the equilibrium portion of the experiment which have a low signal to noise ratio (e.g. Figure 1a) [21]. Fitting the model required determining the free parameter β that minimizes Equation 7. Note that the domain of β is (0, 1). Hereafter, we refer to the method presented above as the Transform- β (T β) method.

We minimized Equation 7 for values of β between 0 and 1 using the Golden Section Search algorithm with custom-written MATLAB (Mathworks, Natick, MA, USA) software. Our convergence criterion was a change of less than 10^{-9} between successive values of either β or the cost function. The T β algorithm was compared to a commonly-used Quasi-Newton method implemented in MATLAB via the function `fminunc` with the same convergence criterion using initial values of $\tau = 5$ and $\beta = 0.5$. This Quasi-Newton method utilized a line search procedure [22], finite-differences to approximate the Hessian matrix via the BFGS method [23–26], and the same convergence criterion as the T β algorithm.

C. Data for Algorithm Testing

The optimization algorithms were tested on both simulated and experimental data. Simulated data were generated from a stretched exponential function with randomly-selected parameters of known values, which acted as gold standards for determining the accuracy of the fitting method. These data ($n = 100$) were simulated using τ and β values sampled from a uniform random distribution ($0 < \tau < 10$ and $0 < \beta < 1$) with normally distributed noise added with a variance $\sigma^2 = 2.68 \times 10^{-6}$.

The experimental data used to test the curvefitting algorithm consisted of stress-relaxation tests performed on cartilage and bone samples. For the tests, a rapid compression was applied and the relaxation of the stress, defined as measured force divided by the initial sample cross-sectional area, was observed. For cartilage tests, a 5% compression was applied followed by 300 s of relaxation for $n = 70$ samples [12]. For demineralized bone samples [27], a 50% strain was applied and relaxation data were sampled for 1000 s ($n = 6$).

D. Analysis

The quality of the fitting methods was assessed using both a nonlinear R^2 metric (Equation 8) [7, 28] and the value of the weighted L^2 norm for the converged fit parameters.

$$R^2 = 1 - \frac{\sum_{i=1}^N (\hat{d}_i - \hat{\sigma})^2}{\sum_{i=1}^N (\hat{d}_i - \hat{d}_{mean})^2} \quad (8)$$

For calculation of R^2 , \hat{d}_{mean} represents the average value of the transformed data and the subscript i represents the index of the discretely-sampled data. The wL^2 norm values were normalized by dividing by the number of datapoints to account for small variations in sampled datapoints between experiments. To compare the algorithms, Wilcoxon Signed-Rank tests [29] with an *a priori* significance level of $\alpha = 0.05$ were used to assess statistical differences between R^2 and wL^2 values. To assess the accuracy of the Transform- β method, linear correlation coefficients were calculated between the fitted τ and β values and the gold-standard parameters used to simulate the data. To examine the T β method, graphs of the wL^2 norm as a function of β were generated and examined for each stress-relaxation dataset. The relaxation data are downsampled for ease of visual display in all plots.

III. RESULTS

Data for the fit quality metrics of R^2 and wL^2 norm were found to be non-normally distributed using the Lilliefors test [30], necessitating the use of the non-parametric Wilcoxon test for statistical comparison between the novel Transform- β method and the Quasi-Newton algorithm. The Transform- β method resulted in significantly larger R^2 values ($p = 0.0003$) and smaller wL^2 norm values ($p = 0.003$) than the Quasi-Newton method when fitting the stress-relaxation data from bone and cartilage samples (Figure 3). Generally, the T β method resulted in qualitatively better or similar fits than the Quasi-Newton method for each experimental dataset. The worst fits for the T β method were better than the worst fits for the Quasi-Newton method (Figure 4). The T β method resulted in accurate fits for both stretched exponential parameters τ ($r = 0.97$) and β ($r = 0.99$). Graphs of wL^2 norm as a function of β demonstrated a single minimum for each dataset ($n = 76$) indicating that the T β method results in unimodal fits. When fitting the simulated data, each algorithm failed to converge on a meaningful solution for a small number of datasets (3 out of 100 for the T β method and 10 out of 100 for the Quasi-Newton method).

IV. DISCUSSION AND CONCLUSIONS

The algorithm presented herein fits the model using a single adjustable parameter in a transformed space which greatly simplifies computation. Using simulated data, we compared this algorithm to a common Quasi-Newton method and found both good performance and accurate fits over a wide parameter range for the novel method. Furthermore, we tested this method on experimental stress-relaxation data from bone and

cartilage, and found that it provided good fits that were better than the Quasi-Newton method.

The Transform- β method provided high-quality fits to the experimental data. Large R^2 and small wL^2 values (Figure 3) demonstrate the capacity to fit cartilage and bone stress-relaxation data (Figure 4). The $T\beta$ method resulted in better fits than the Quasi-Newton method as assessed by R^2 and wL^2 norm values indicating that it may be an improvement for determining stretched exponential parameters for relaxation data. The high-quality fits to the simulated data suggesting that the $T\beta$ algorithm may be generalizable to many different relaxation systems [1, 2, 6, 10].

The $T\beta$ method has advantages compared with the Quasi-Newton and other gradient-based methods. While gradient-based methods require an initialization in the solution space, because the $T\beta$ method requires searching for only a single parameter over a finite interval ($0 < \beta < 1$), no initialization is needed, and the entire solution space can be searched. Finally, in all of the datasets examined, the wL^2 norm appears to be a unimodal function of β indicating that unique solution can be obtained to avoid the problems of redundant fits (Figure 5).

While the $T\beta$ method can provide high-quality fits to experimental and simulated data, there are important limitations to this method. First, the relaxation data to be fit must be shaped as a stretched exponential. Non-stretched exponential relaxation data cannot be fit by this algorithm. Additionally, the $T\beta$ method is challenged by datasets containing small τ and large β values. In these cases our empirical results suggest that good estimation of the equilibrium values is essential toward using the $T\beta$ method to obtain high-quality fits.

In conclusion, we have developed a novel algorithm for determining the stretched exponential parameters for relaxation data. This method is advantageous compared with other fitting algorithms in that it does not require an initialization in solution space and requires searching in only one dimension.

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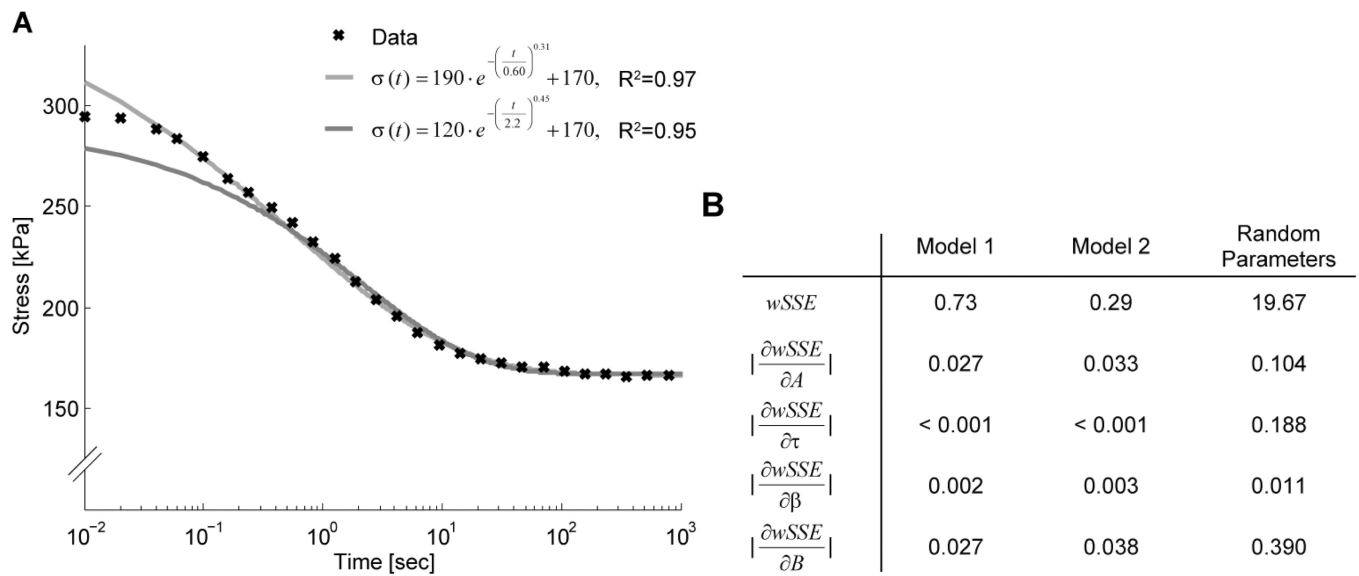
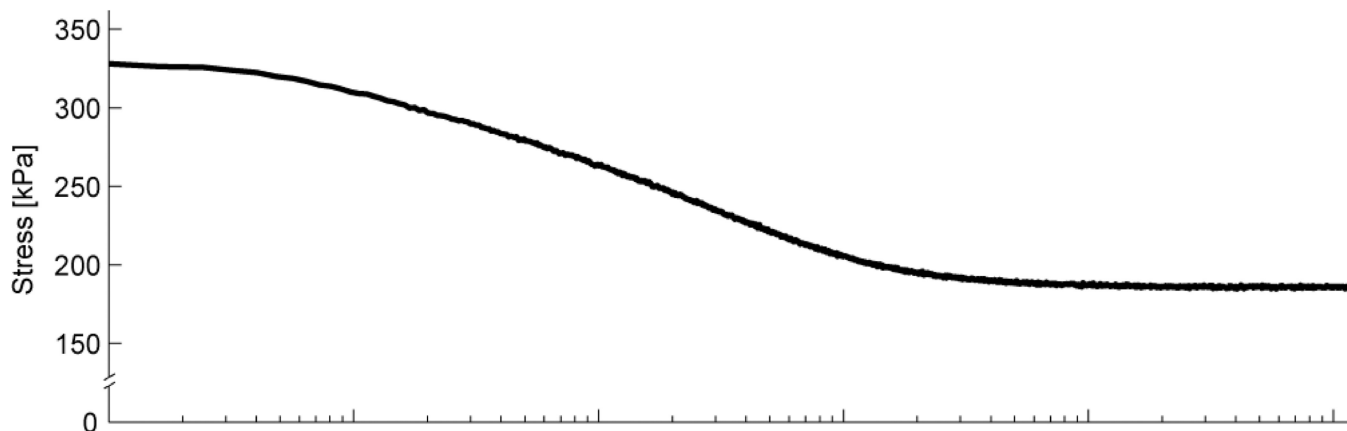
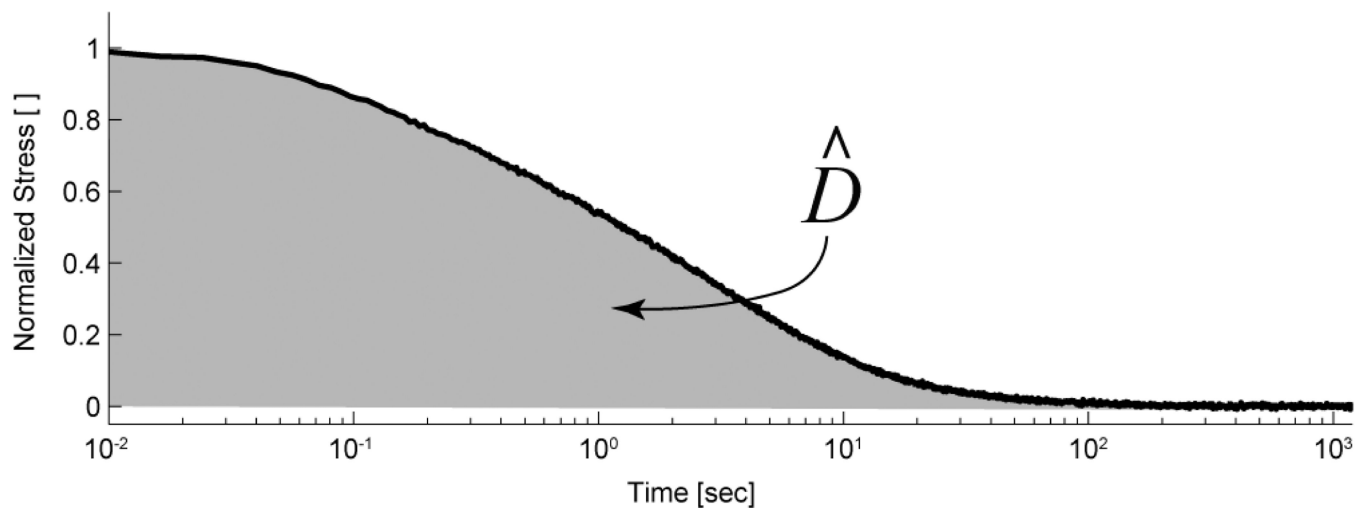


Fig. 1. Non-unique fitting of the stretched exponential function to relaxation data using a gradient-based steepest descent method. (A) Two distinct converged models for the same experimental dataset have substantially different τ and β values while having similar derivative values (B). These fits were obtained by minimizing the weighted sum of the squared error ($wSSE$) between the model and the data using a Quasi-Newton method. Similar quality fits resulted with disparate combinations of fitted parameters that depended on the initialization of the algorithm. These results demonstrate the challenges associated with using a descent-based fitting method to determine stretched exponential parameters from experimental data.

A Raw Data**B Transformed Data****Fig. 2.**

Transformation used in $T\beta$ method. This method utilizes the parameter \hat{D} to fit the data. \hat{D} is calculated by transforming the data as shown. Relaxation data (A) were transformed via Equation 3. The shaded region (B) represents \hat{D} which is calculated from the experimental data and used to calculate the final fitted parameters.

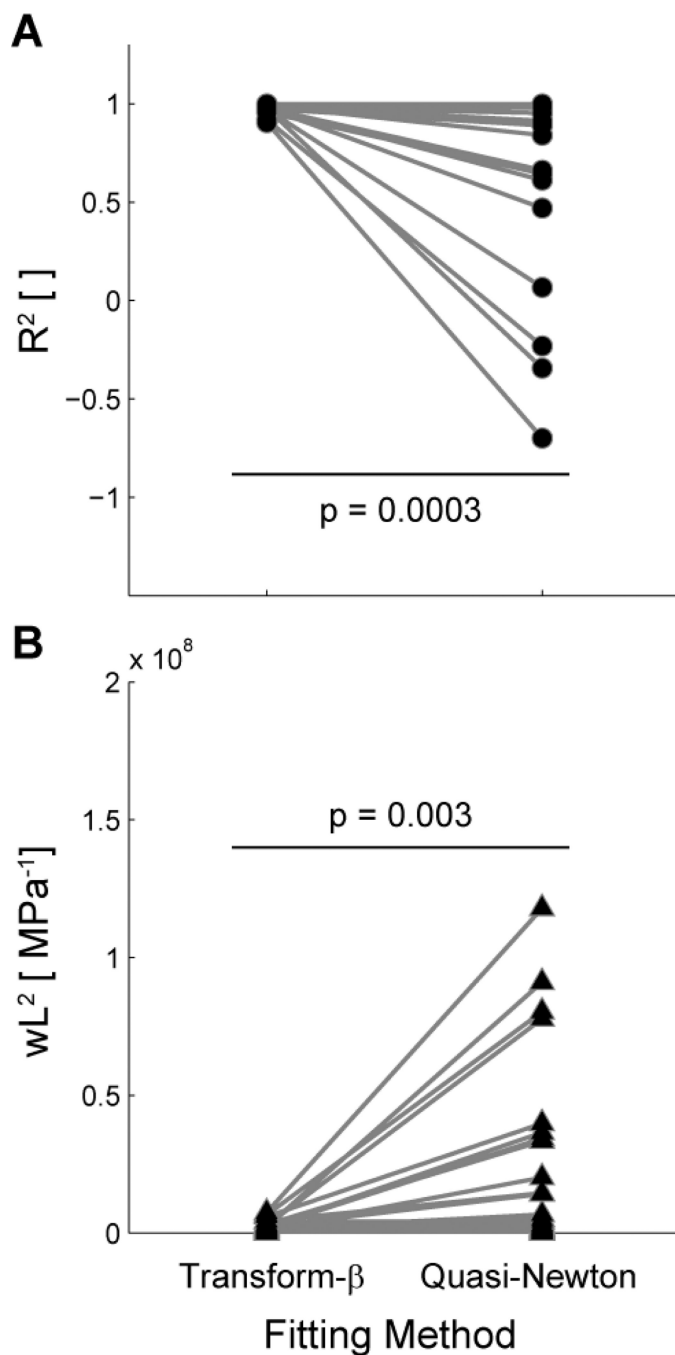
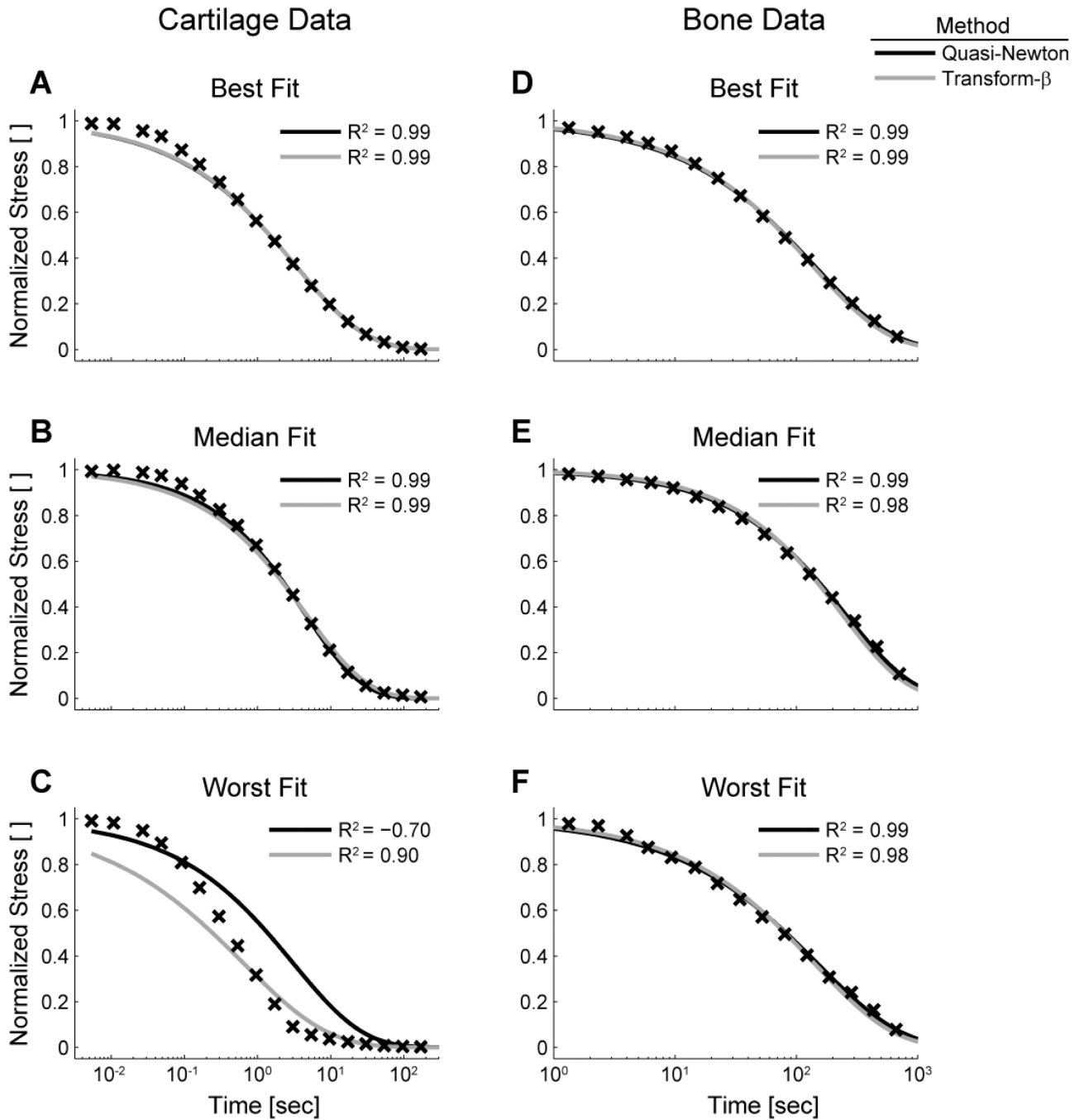


Fig. 3. The Transform- β method resulted in higher-quality fits than the commonly-used Quasi-Newton method. Each symbol represents results from an individual experimental dataset connected by gray lines between methods. (A) R^2 values were significantly larger ($p = 0.0003$) for fits resulting from the T β method than those resulting from the Quasi-Newton method. (B) wL^2 norm values were significantly smaller for the T β method than for the Quasi-Newton method ($p = 0.003$).

**Fig. 4.**

The Transform- β method yielded better fits to both bone and cartilage stress-relaxation data than the Quasi-Newton method. Panels A–C (left) show cartilage data and panels D–F (right) show bone data. Best (top), median (middle), and worst (bottom) fits were determined, respectively, by the smallest, median, and largest normalized wL^2 norm values obtained using the present method. For comparison, the Quasi-Newton results are shown in black.

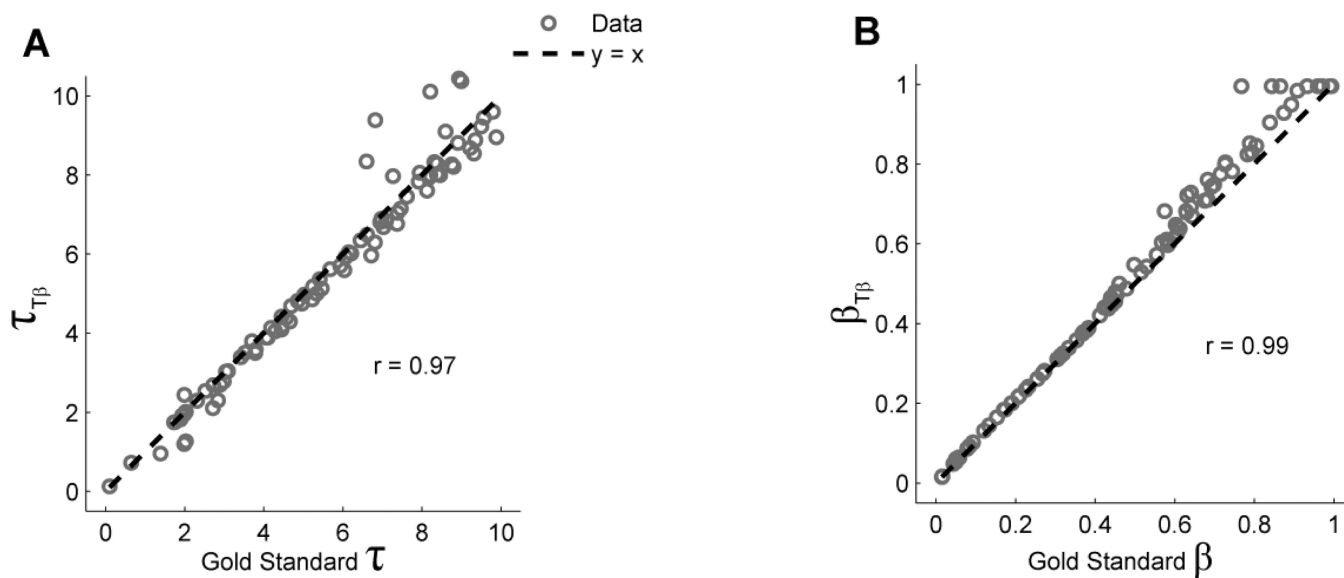


Fig. 5. Large correlation coefficient values between simulated data (gold standard) and converged model parameters for the present method. Gold standard data were simulated numerically including zero-mean noise. The proposed algorithm was used to determine τ and β from the simulated data. Large correlations > 0.96 were found between the gold standard values for both τ (A) and β (B).