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# Viscometric characterization of cobalt nanoparticle-based magnetorheological fluids using genetic algorithms

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**Abstract**

The rheological flow curves (shear stress vs. shear rate) of a nanoparticle cobalt-based magnetorheological fluid can be modeled using Bingham-plastic and Herschel–Bulkley constitutive models. Steady-state rheological flow curves were measured using a parallel disk rheometer for constant shear rates as a function of applied magnetic field. Genetic algorithms were used to identify constitutive model parameters from the flow curve data.


*Keywords:* Magnetorheological fluids; Nanoparticles; Cobalt; Bingham-plastic model; Herschel–Bulkley model; Genetic algorithms; Rheology

## 1. Introduction

Magnetorheological (MR) fluids are a class of smart materials whose rheological properties may be varied by application of a magnetic field. These fluids are suspensions of soft magnetic particles (such as iron or cobalt) in a carrier fluid. Each particle has a dipole moment, the strength of which is roughly proportional to its diameter [1]. Upon application of a magnetic field, these dipoles align parallel to the magnetic field and form chains. A finite stress must develop in the fluid to yield these structures. The field-dependent yield stress of these fluids is continuously controllable and this controllability has been the primary reason for their use in numerous smart actuation systems [2,3].

MR fluids have been produced with different types and sizes of magnetic carrier particles. The majority of existing MR fluids are composed of micron-scale Fe particles suspended in a nonmagnetic carrier fluid [4–10]. These MR fluids have high yield stress (20–100 kPa) due to the strength of the dipoles created by the particles. The comparatively higher yield stress over electrorheological (ER) fluids (2–5 kPa) is one major

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advantage of MR fluids. However, the density of the particles makes them susceptible to settling in the absence of frequent mixing due to predominant gravity forces. Once sedimented, the residual magnetic attraction between particles makes redispersion difficult. The large particles also lead to unwanted abrasion of the components in contact with the fluid. Ferrofluids, which are suspensions of iron particles of less than 10 nm in diameter, have also been reported [11,12]. However, these ferrofluids do not present formation of elongated chain-like microstructures under the application of a magnetic field and they are unable to provide significant magnetoviscous effect, especially significant yield stress. Nanometer-sized particles (10–100 nm) have been introduced [13,14] with an attempt to reduce settling while maintaining useful yield stress levels. The mixture is seen to overcome the settling problem due to the predominance of thermodynamic forces [15,16] but the yield stresses obtained for the same shear rates and magnetic field levels are drastically reduced in comparison to fluids containing micron-sized particles. The yield shear stresses in these fluids are comparable with those achieved in electrorheological fluids.

We use the Bingham-plastic (BP) and Herschel–Bulkley (HB) constitutive models to characterize the rheological behavior of MR fluid. In both fluid models, it is assumed that the preyield behavior is rigid, and that the fluid flows if and only if the local shear stress is greater than the yield stress. For a BP model, once the local shear stress exceeds the yield stress, the postyield behavior is linear in that the shear stress increases linearly with shear rate (linear postyield model). The BP model employs two parameters: yield stress, $\tau_y$, and viscosity, $\mu$. For the HB model, once the local shear stress exceeds the yield stress, the postyield behavior is nonlinear in that the shear stress increases as a power law of shear rate (nonlinear postyield model). The HB model employs three parameters: yield stress, $\tau_y$, consistency, $K$, and flow index, $n$. The flow index $n$ can be used to classify the fluid; $n>1$ indicates a shear-thickening fluid and $n<1$ indicates a shear-thinning flow. The HB model has been used to characterize the flow of MR fluids, especially where shear thickening or thinning is seen [17–19]. A key issue is to identify model parameters from flow curve measurements, that is, the shear stress as a function of shear rate for varying levels of applied magnetic field.

Genetic algorithms (GA) have been widely used in applications where a globally optimal solution is required. In conventional estimation methods, a model structure is chosen and the parameters of that model are calculated by minimizing an objective function. Gradient descent techniques are usually used for the minimization, but these are very susceptible to initial guesses and the obtained parameters may only be locally optimal. On the other hand, GA uses a probabilistically guided search procedure that simulates genetic evolution [20,21]. Populations with stronger fitness are identified and retained, while weaker ones are discarded. The process ensures that successive generations are fitter. The algorithm cannot be trapped in local minima since it employs random mutation procedures. The overall search procedure is stable and robust and can identify globally optimal parameters of a system.

The rheological parameters of MR fluid constitutive models can be identified from flow curves measured using a parallel disk rheometer. Flow curves were measured as a function of applied field for MR fluid with a solids loading of 40 weight percent (wt%) nanometer scale cobalt particles. The particles were aspherical in shape with an equivalent diameter of 100 nm (Fig. 1). Both the BP and HB models were fitted using a simple GA to estimate model parameters. Constraints were applied within the algorithm to ensure a monotonically increasing trend of the yield stress with increase in applied magnetic field. The identified rheological parameters provided a good model fit to measured flow curves. The parameter variations with change in applied magnetic field were smooth. Comparison of estimation errors suggest that the HB model is more accurate over the entire range of measured shear rates, but the BP model is equally good for the high shear rate regime.

## 2. Synthesis and testing of MR fluids

A chemical precipitation technique was employed to produce cobalt nanopowders. In this

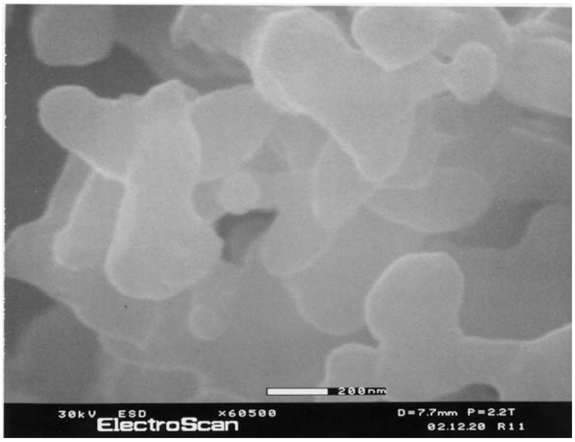


Fig. 1. SEM picture of cobalt nanoparticles.

process, ammonium hydroxide was used to precipitate an aqueous suspension of cobalt nitrate at a pH of 11. The resulting precipitate (cobalt hydroxide) was then centrifuged and washed several times with distilled water and iso-propanol to remove excess hydroxide. This was followed by overnight drying at 150 °C to produce cobalt oxide nanopowders. Cobalt oxide nanopowders were further reduced under hydrogen at 450 °C for 3 hours to obtain pure elemental cobalt nanoparticles.

To prepare stable MR fluids, hydraulic oil was chosen as a carrier fluid. Mobil DTE20 series is used extensively in high-pressure systems including industrial, marine and mobile services because of its excellent anti-wear properties, multi-metal compatibility and corrosion resistance. Air Products AD01 EnviroGem was used as a surfactant in the magnetorheological suspensions. The fluid was mixed in hydraulic oil using a high-speed emulsifier at nearly 11,000 rpm. Nanopowders obtained from the microwave plasma synthesis were added to the oil and the mixing continued. Concentration of cobalt was 40% (by weight) in the fluid.

## 3. Rheological models

MR fluids demonstrate nonlinear behavior when subjected to external magnetic fields. The rheological behavior of these materials can be separated into distinct preyield and postyield regimes. MR fluids are known to exhibit a strong field-dependent shear modulus and a yield stress that resists flow until the shear stress reaches a critical value. Both the BP and HB models present such a behavior, and both models present rigid preyield behavior. The BP model has been used as a constitutive model for MR fluids [10]. The simplicity of this two-parameter model, with yield stress, $\tau_y$, and postyield viscosity, $\mu$, has led to its wide use for representation of field-controllable fluids. This model assumes that the fluid exhibits shear stress proportional to shear rate in the postyield region and is described by the following equation:

$$\tau = \tau_y + \mu\dot{\gamma}. \tag{1}$$

However, in cases where the fluid experiences postyield shear thickening or shear thinning, the behavior becomes nonlinear and the assumption of constant plastic viscosity is invalid. The HB model is more suitable as a constitutive model for MR fluids [14] and has been applied to analysis of MR-based devices [18]. The HB model can be represented as

$$\tau = \tau_y + K\dot{\gamma}^n, \tag{2}$$

where yield stress, $\tau_y$, consistency, $K$ and flow index, $n$, are the model parameters. The model assumes that below the yield stress, $\tau_y$, the suspension behaves as a rigid solid, similar to the BP model. The flow index characterizes the postyield behavior; $n > 1$ indicates shear thickening and $n < 1$ indicates shear thinning. The BP model is a special case ($n = 1$) of the HB model.

## 4. Genetic algorithms for model parameter estimation

The cumulative squared error between measured and fitted shear stress values as a function of shear rate was used as the objective function, which leads to a quadratic minimization problem. Most algorithms perform a minimization of the cost function starting at a user-provided initial guess and use additional information, usually function gradients, to approach the minima. However, such techniques only yield a local minimum in the proximity of the initial guess. In the case of nonlinear estimation, there may exist multiple optimal solutions that are equally significant, but identification of all solution sets is not possible unless the optimizer is run with different initial guesses. Function gradients at all points are also not always available or are computationally costly.

GAs are a tool for finding global solutions in large parameter spaces by simultaneously evaluating and refining different solution sets to identify near-optimal solutions rather than employing methods to converge to a single solution. GAs are different from usual optimization and search procedures [20,21] in that they search from a population of points and use probabilistic transition rules, not deterministic rules. An important consideration is that a GA uses only objective/fitness function information, and not derivative or any other auxiliary knowledge. The evolution scheme selects "fitter" individuals to be members of the new generation and adds to this generation the individuals that result from crossover and mutation of the selected individuals. Convergence is guaranteed by the selection that makes the best solution of the new generation better or equal to the best solution of the previous generation. The overall search procedure is stable and robust and can identify globally optimal parameters of a system. GAs have the ability to solve highly nonlinear functions where other techniques fail.

We used a simple genetic algorithm (SGA) [22,23] for identifying the parameters of the rheological model. A population of $P$ individuals was generated, each member composed of a binary string of length $N$. Each individual was mapped to a rheological parameter using maximum and minimum bounds as follows:

$$\text{Parameter value} = (\text{decoded value of binary string}) \times \frac{(\text{upper bound} - \text{lower bound})}{(2^N - 1)}. \quad (3)$$

The initial population was generated randomly and spread over the entire space of possible parameter values. Model equation (2) was then used to determine the error and corresponding fitness value of each population member. For each iteration, we applied the three genetic operators (reproduction, crossover and mutation) to create a new generation. Reproduction was simulated by a simple roulette wheel-based selection scheme. Crossover was carried out at a single point for each parameter of the model. To prevent the algorithm from getting stuck in local minima, new genetic information was periodically injected via mutation. The objective function used was the sum of squared errors and the corresponding fitness was evaluated as the reciprocal of the objective function. For each individual,

$$\text{squared error} \quad E_j = \sum_{i=1}^{N_e} w_i \times (\tau_i - \hat{\tau}_i)^2, \quad (4)$$

$$\text{fitness value} \quad F_j = 1/E_j, \quad (5)$$

where $N_e$ is the number of experimentally obtained points for each value of magnetic field, $\hat{\tau}_i$ is the measured shear stress for a particular strain rate and $\tau_i$ is the shear stress at the same strain rate for a particular set of rheological parameters. $w_i$ was used as a weighing factor. To compare the fitted model with the experimental data, we defined errors as functions of magnetic field, $\phi$, as follows:

$$\text{Abs error} \quad e(\phi) = \sqrt{\frac{1}{N_e} \sum_{i=1}^{N_e} (\tau_i - \hat{\tau}_i)^2}, \quad (6)$$

$$\text{Percentage error} \quad P(\phi) = \frac{e(\phi)}{\tau_{\phi\ \max}} \times 100, \quad (7)$$

where $\tau_{\phi\ \max}$ is the maximum measured shear stress at a particular value of magnetic field, $\phi$. The search procedure was carried out for each value of magnetic field. The flow index, $n$, was allowed to vary from 0 to 1, as the measured flow curves strongly indicated that the cobalt nanoparticle MR fluid exhibits shear thinning. In case of the yield stress, $\tau_y$, for each level of applied magnetic field, we selected the highest measured shear stress as the upper bound and the estimated yield stress at the next lowest magnetic field as the lower bound. This procedure resulted in a monotonically increasing yield stress as a function of magnetic field. This constraint was included because a yield stress that increases monotonically with applied magnetic field was physically meaningful.

Several termination criteria for a genetic search have been proposed. One simple criterion is to stop the search when almost all individuals in the population are nearly identical; another criterion is to test the improvement in the best fitness score over successive generations. However, the first criterion can lead to extensive search times while the second one is not good for functions characterized by ''plateau-type'' regions [24]. In our work, we stopped the GA after a fixed number of generations, which was chosen as a compromise between constraints of convergence, computing time and accuracy.

HB model GA parameter identification results were also compared to a least mean squares (LMS) error minimization technique employing the same constraints as discussed above for the GA technique.

## 5. Results

The parameters of the constitutive models were obtained after carrying out global optimization using a simple genetic algorithm. The algorithm was run with a population size $P = 2000$ and using $N = 50$ bits for encoding. The probability of crossover was chosen as $P_c = 0.85$, and the probability of mutation was chosen as $P_m = 0.025$. Constraints were applied to the search scheme to ensure that a monotonically increasing characteristic was obtained for the yield stress with increasing magnetization. The cost function was evaluated Eq. (4). The weight, $w_i$, is adjusted so that the algorithm would provide a better fit to the flow curve data at the higher shear rates.

The parameters estimated for the BP model are shown in Fig. 2. The yield stress increases monotonically from 50 Pa at 0.03 T to 1750 Pa at 0.30 T. The variation in yield stress is smooth and almost linear, while the viscosity changes smoothly in a nonlinear manner as magnetic field increases.

The identified model parameters for the HB model are shown in Fig. 3. The parameters change smoothly over the range of applied magnetic fields. Yield stress, $\tau_y$, presents a monotonically increasing trend with values ranging from 10 to 1450 Pa, and are lower than those obtained from the BP model. The estimated consistency parameter ($K$) and flow index ($n$) show considerable scatter and

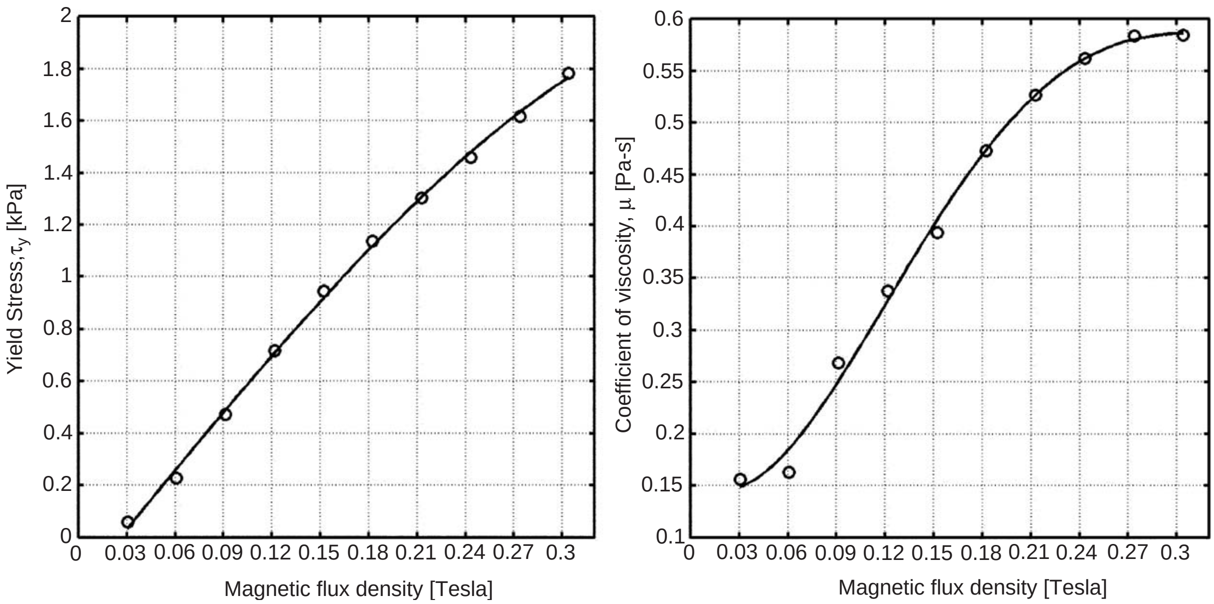


Fig. 2. Identified parameters for Bingham-plastic model.

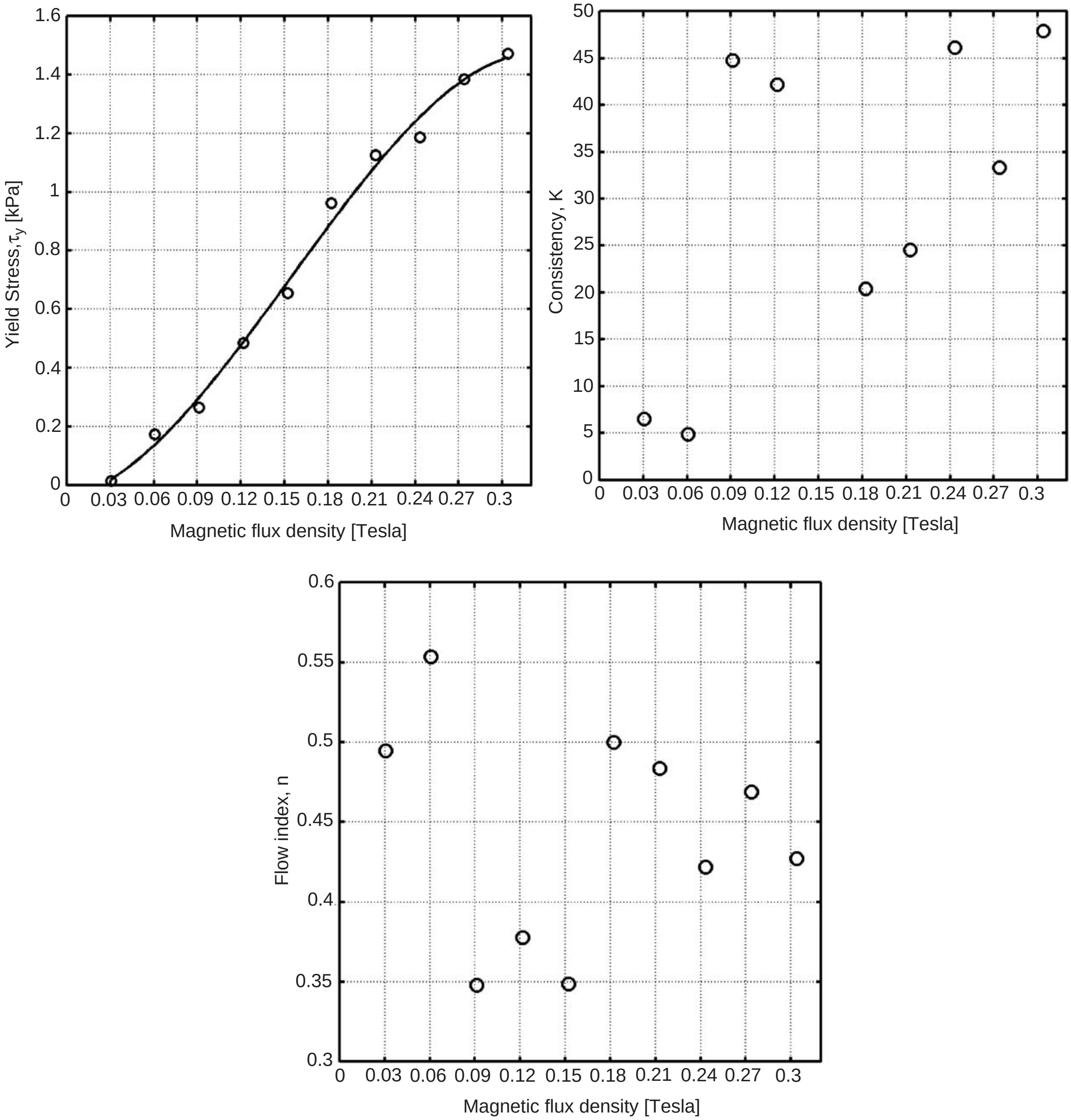

Fig. 3. Identified parameters for Herschel–Bulkley model.

their variations do not carry physical significance. To improve the estimation results, we ran the optimizer with a fixed flow index value of $n = 0.45$. These results are shown in Fig. 4. Both yield stress and consistency are seen to be monotonically increasing functions of magnetic field in this case.

Using the identified parameters, we fitted models to the measured flow curve data (Fig. 5).

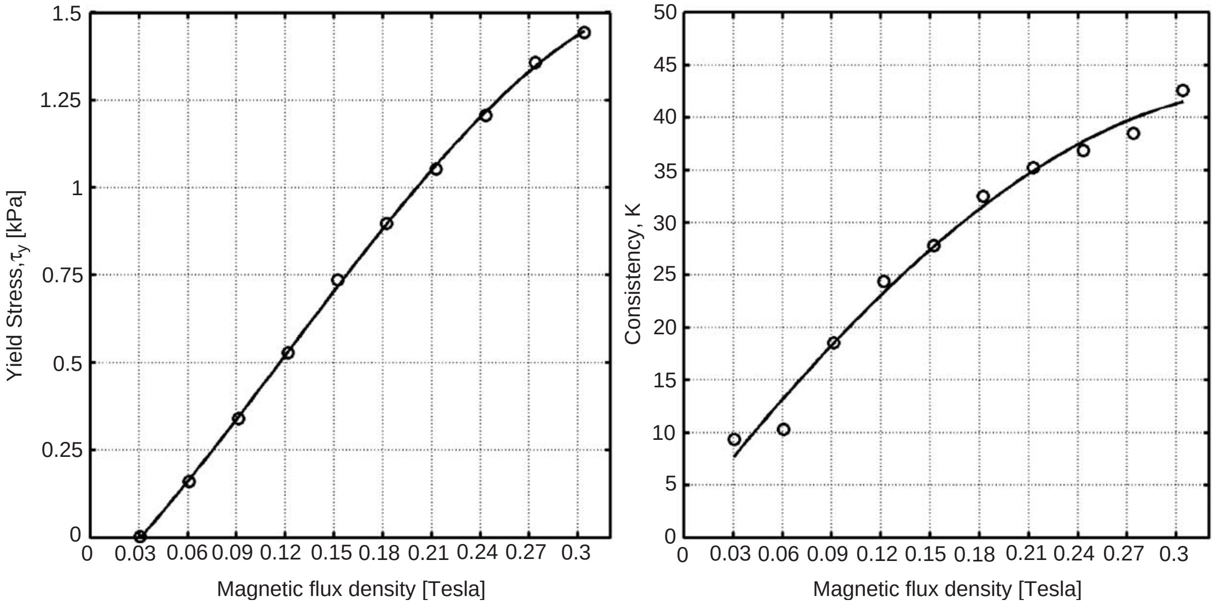


Fig. 4. Identified parameters for Herschel–Bulkley model with $n = 0.45$.

Both the BP and HB models fit the high shear rate data equally well, but the low shear rate regions are better represented by the HB model. Finally, we compare the fitting errors, obtained using Eqs. (6) and (7), for different models (BP and HB) and schemes (LMS and GA). The plots are shown in Fig. 6. The HB model, with errors limited to 2% of the maximum stress, was more accurate than the BP model. In addition, the HB model parameters identified via the GA technique were improved over those identified via the LMS technique in that the modeling error was lower. The HB model with fixed $n$ also had the lowest modeling errors and is more suitable as a constitutive equation for the fluid flow since it results in smoother variation of all the identified parameters.

## 6. Conclusion

In this study, we identified the rheological parameters of two constitutive models, Bingham-plastic (BP) and Herschel–Bulkley (HB), that we used to characterize the rheological properties of nanocobalt based magnetorheological fluid. Flow curves were measured for a suspension of cobalt particles, having equivalent diameter of 100 nm, at different levels of magnetic field. The flow curves were measured for discrete values of applied magnetic field ranging from 0.03 to 0.30 T. A simple genetic algorithm was used to estimate the parameters of the constitutive model. Constraints were imposed on the minimization process to ensure that the yield stress was a monotonically increasing function of applied magnetic field. The following points are noted:

(i) The maximum yield stress measured in the cobalt nanoparticle-based MR fluid at 0.3 T was 1.75 kPa (BP) or 1.45 kPa (HB), which is much lower than yield stresses reported for MR fluids with micron-sized particles at comparable solids loading and applied magnetic field.
(ii) The dynamic yield stress is seen to vary from about 10 Pa at 0.03 T to almost 1450 Pa at 0.30 T for the HB model, and from 50 to 1750 Pa for the BP model.
(iii) For the BP model, the yield stress and postyield viscosity were both monotonically increasing functions of magnetic field.

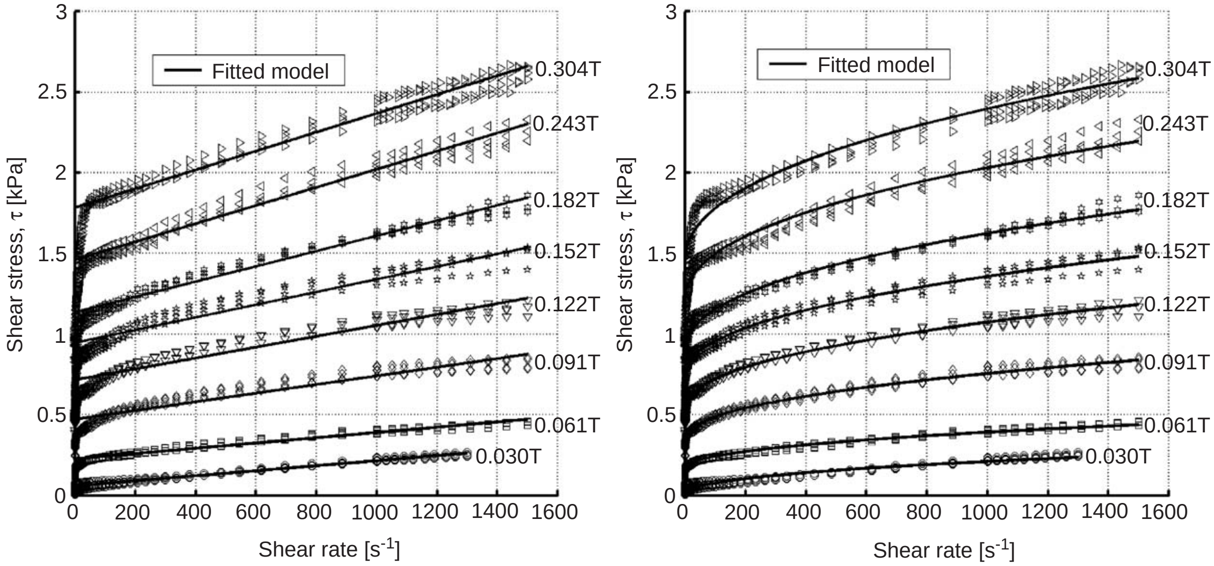


Fig. 5. Reconstruction of rheological flow curves using (a) Bingham-plastic model, and (b) Herschel–Bulkley model.

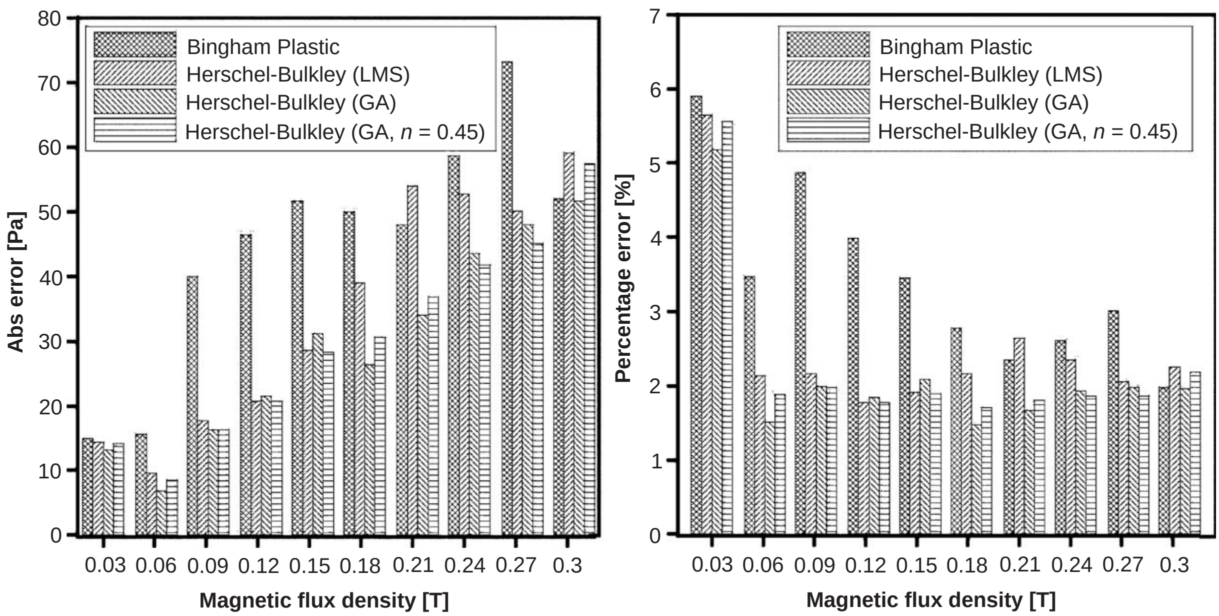


Fig. 6. Errors between reconstructed and experimentally obtained rheological flow curves: (a) absolute error, (b) percentage error.

(iv) For the HB model, choosing a constant value of $n = 0.45$ (indicating shear thinning) produced yield stress and postyield consistency that were both monotonically increasing functions of magnetic field. Also, a constant flow index, that is, a flow index that is not dependent on field strength, resulted in the lowest modeling error.

(v) The percentage errors in model fitting were less than 2% of the peak shear stress when GA was used to identify the HB model parameters. The LMS technique had higher errors than the GA technique.
(vi) Both the BP and HB models can model the fluid effectively. The HB model had errors averaging 2%, which is about 1% lower than the errors observed for the BP model.

We can conclude that both the Bingham-plastic and Herschel–Bulkley model can be used as constitutive equations to describe the behavior of nanocobalt-based MR fluids, especially in the high shear rate region. Genetic algorithms can be efficiently applied to such rheological parameter estimation problems and the results are comparable to those obtained from gradient-based schemes.

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