ORIGINAL RESEARCH ARTICLE

Optimizing of chemical admixtures for 3D printable cementitious materials by central composite design

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Abstract

Printability of 3D printable cementitious materials is related to material rheological properties, and is affected and controlled by modern concrete chemical admixtures. In this work, the influence of several chemical admixtures including superplasticizer, retarder, and accelerator on the rheological characteristics of printable materials was investigated using central composite design (CCD). Twenty test points with varying dosages of chemical admixtures were performed to evaluate the primary effects of chemical admixtures and their combined interactive effects on the rheological properties. The results indicate that with the increase of retarder or superplasticizer dosage, all rheological parameters decrease while accelerator possesses an opposite impact. The rheological properties are negatively proportional to the combined interactive effect of retarder and accelerator. The combined interactive effect of retarder and superplasticizer positively affects dynamic yield stress, plastic viscosity, and thixotropy, while it negatively impacts static yield stress. The combined interactive effect of accelerator and retarder positively affects the yield stress, whereas it negatively influences the plastic viscosity and thixotropy. The results indicate that the CCD is an efficient method to find the desirable formulation within a given boundary.

Keywords: 3D concrete printing; Central composites design; Rheological properties; Statistical models

1. Introduction

The 3D concrete printing (3DCP) technique, an extrusion-based additive manufacturing process¹-³, has attracted much attention in recent years due to its advantages, such as automated process, formwork-free construction, and improved productivity⁴-⁶. These benefits are mainly due to the automated layer-by-layer construction process based on a 3D model. In the printing process, material rheological performance is a critical factor determining the printability of printed structures⁷-⁹.

Printability is characterized by buildability and pumpability, which are related to material rheological properties (static/dynamic yield stress, plastic viscosity, and thixotropy)¹⁰,¹¹. These parameters are affected by various factors, including material
constituents and chemical admixtures\textsuperscript{[12-26]}. Weng \textit{et al.}\textsuperscript{[8]} explored the impact of material constituents on rheological properties of 3D printable materials and proposed statistical models to predict rheological properties. Zhang \textit{et al.}\textsuperscript{[17]} presented that the buildability could increase by 150% with the addition of a small quantity of nano-clay. Apart from material constituents, chemical admixtures also serve vital roles on rheology of concrete\textsuperscript{[12,13]}. Dressler \textit{et al.}\textsuperscript{[18]} studied the effect of accelerator on the material properties in shotcrete 3D printing. Tao \textit{et al.}\textsuperscript{[19]} investigated the stiffening control of material using an inline mixing process with chemical admixtures. Yu \textit{et al.}\textsuperscript{[20,21]} studied the influence of mortar composition on the aggregate bed process by adjusting the sand/cement ratio and the water/cement ratio in aggregate-bed 3D concrete printing.

Many research works have been conducted to study the impact of chemical admixtures on rheological properties\textsuperscript{[22-24]}. However, there are still certain limitations. First, conclusions from the previous works are mainly qualitative. Few quantitative results have been established to explain the impact of chemical admixtures on the rheological properties. Furthermore, research needs to be carried out to explore the impact of chemical admixtures on thixotropy, which measures the structural rebuilding rate of materials. Therefore, more attention should be paid to explore the impact of chemical admixtures on rheological properties and construct models to predict rheological properties. More specifically, an efficient approach should be adopted for experimental design, and quantitative models should be built empirically through a series of experiments\textsuperscript{[25]}

Design of experiments (DoE) is a class of scientific methodology for experimental design and data analysis to improve research efficiency based on fundamental mathematical statistics\textsuperscript{[26]}. It has been successfully used in various research fields as a powerful approach to exploring the relationship between factors and responses\textsuperscript{[27-29]}. One of the useful DoE methods is called central composite design (CCD), quantifying the impact of variables on responses through constructing statistical models. Using CCD, the experimental process can be simplified, and the experimental runs can be reduced, while the sufficient information can be extracted from the experiment for data analysis. In summary, the CCD method is more efficient than traditional one-factor at one-time experiment design.

In this study, CCD was adopted to efficiently construct statistical models, expressing the rheological characteristics as functions of different factors, that is, various chemical admixtures. The constructed statistical models are not universally applicable\textsuperscript{[30]}, while the results indicate that the CCD is efficient to find the desirable formulation within a given boundary.

2. Methodology

2.1. Response surface method and central composite design

As one of the most reliable statistical methodologies in DoE, response surface methodology (RSM) includes optimization procedures for the settings of factorial variables, such that the response reaches a desired maximum or minimum value\textsuperscript{[26]}. The RSM includes various design structures, such as CCD and Box-Behnken\textsuperscript{[26]}. CCD design structure was used in this work as it explores a larger process space and provides a higher prediction quality over the entire space than that of Box-Behnken.

The structure of CCD design includes corner points, axial points, and center points (corresponding to ±1, ±1.68, and 0 as shown in \textbf{Figure 1}). Corner points are the parameters with boundary values. The axial points can make the model in quadratic terms, considering the curvature effect. The experiments of center points were replicated for several times to provide information on process reproducibility.

2.2. Rheology and time-dependent effect

2.2.1. Rheology of cementitious materials

Rheological properties of cementitious materials are described by Bingham plastic model and characterized by the static/dynamic yield stress and plastic viscosity. The correlation between shear stress $\tau$ (Pa) and shear rate $\gamma$ (1/s) in the Bingham model is described in Equation 1:

$$\tau = \tau_0 + k\gamma$$  \hspace{1cm} (I)

Where $\tau_0$ is yield stress, which includes static yield stress $\tau_s$ (Pa) and dynamic yield stress $\tau_d$ (Pa). $\tau_s$ and $\tau_d$ are the minimum shear stress to initiate and maintain the flow of materials, respectively. Plastic viscosity $k$ (Pa·s) describes the resistance of fluid to flow when it is agitated. All the rheological parameters can be obtained from the

\begin{figure}[h]
\centering
\includegraphics[width=0.5\textwidth]{Design_structure_of_CCD.png}
\caption{Design structure of CCD.}
\end{figure}
rheological test\cite{11}, in which Bingham model can also be expressed as the following formula (Equation II) for convenience of experiment design and data analysis:

\[ T = G + hN \]  

(II)

Equation II describes the correlation between the measured torque \( T \) (N·m) and rotational speed \( N \) (rpm). The parameter \( G \) (N·m) is flow resistance, representing the minimum torque required to initiate or maintain the flow of a material. The parameter \( h \) (N·m·min) is torque viscosity. Similar to the \( k \) in Equation I, the parameter \( h \) describes the change of applied torque with altering rotational speed.

Buildability and pumpability can be characterized by a build-up model and a pumping pressure model, respectively. The build-up model can be adopted to predict the printed height of structures with static yield stress of material, and the model is expressed in Equation III\cite{31}:

\[ H = \frac{\alpha}{\rho g} \cdot \tau_s(t) \]  

(III)

Where \( H \) (m) and \( \alpha \) are the printed height (buildability) and the geometric factor of printed structures, respectively; \( \rho \) (kg/m\(^3\)) and \( g \) (m/s\(^2\)) are the density of materials and gravitational constant, respectively. Equation III implies that the printed height is positively proportional to the static yield stress for a given material and structure. Pumpability, generally characterized by pumping pressure, is positively related to dynamic viscosity that measures a fluid’s resistance to flow when an external force is applied\cite{32}.

### 2.2.2. Time-dependent effect of rheological properties

Rheological properties evolve with time due to hydration process. A theoretical model proposed by Roussel et al.\cite{33} correlates yield stress with resting time. The model is expressed in Equation IV:

\[ \tau_s(t) = \tau_s(0) + A_{thix} \cdot t \]  

(IV)

Where \( t \) (s) is time at rest; \( A_{thix} \) (Pa/s) is thixotropy parameter, a constant value for a given material; \( \tau_s(0) \) is the static yield stress as a function of resting time \( t = 0 \). A high \( A_{thix} \) is required for the 3D printable cementitious material in printing process to accelerate the increase of static yield stress to make certain that materials possess appropriate buildability.

The time effect on the evolution of dynamic viscosity is expressed in Equation V:

\[ \mu(t) = \mu_0 + (1000 - \mu_0) \cdot \frac{t}{t_r} \]  

(V)

Where \( t \) (s) is time at rest; \( t_r \) is 1000 Pa·s, \( \mu_0 \) is initial dynamic viscosity. Generally, dynamic viscosity changes slightly in 30 min after mixing\cite{34}.

### 3. Materials, mixture design, and properties characterization

#### 3.1. Materials and mixture design

Material mixture in this study consists of ordinary Portland cement (OPC, ASTM type I, Grade 42.5), silica fume (SF, undensified, Grade 940, Elkem company), fine sand, fly ash (FA, Class F), water, superplasticizer (MasterPozzolith-R168, BASF Pte. Ltd.), accelerator (MasterRoc SA160, BASF Pte. Ltd.), and retarder (MasterReobuild1000, BASF Pte. Ltd.). Particle size distribution is illustrated in Figure 2, and the chemical composition of all the raw ingredients used is shown in Table 1. The mixtures used in this study follow the same mixture proportion, as shown in Table 2.

The dosage of chemical admixtures was designed by the CCD, and the coded and actual values used in the experiment are presented in Table 3. The relationship between coded and actual values is expressed in Equation VI:\cite{8}

\[ \text{Coded value} = \frac{(\text{Actual value} - \text{Factor mean})}{\left(\text{Range of factorial value} / 2\right)} \]  

(VI)

#### 3.2. Mixing process and properties characterization

A Hobart mixer X200L was used for mixing. The rheological properties of cement slurries are influenced by several factors, such as speed, time, and temperature. Thus, mixing procedures in this study were fixed to minimize the difference among batches. First, the powders of all solid ingredients were dry mixed for 1 min in stir speed (33 rpm). Water, superplasticizer, and retarder were then added, and the mixing process continued for 1 min in stir speed (33 rpm) followed by 1 min in speed I (61 rpm);
the accelerator was then added, and the mixing process continued for 1 min in speed II (113 rpm).
Rheological properties of mixed materials, including static/dynamic yield stress and plastic viscosity, were characterized via Viskomat XL. The six-blade vane probe and cage are used for rheological test to avoid slippage of cement paste. Both the diameter and the height of this vane probe are 69 mm, and the gap between probe and cage, and the bottoms of probe and barrel are both 40 mm. During the rheological test, the speed of rheometer increased linearly from 0 rpm to 60 rpm in 5 min. Afterward, the speed decreased linearly to 0 rpm in another 5 min as shown in Figure 3. The typical test result of rheological properties is plotted in Figure 4. Then the static/dynamic yield stress and plastic viscosity can be computed by Equation VII [11]:

$$\Gamma = \frac{4\pi R_1^2 R_2^2 l k}{R_2^3 - R_1^3} \omega_2 - \frac{4\pi R_1^2 R_2^2 l_2}{R_2^3 - R_1^3} \ln \frac{R_1}{R_2}$$  \hspace{1cm} (VII)

Where $\Gamma$ (N·m) is the torque, $\omega_2$ (rad/s) is the rotational speed of outer barrel, $l$ (m) and $R_1$ (m) are the length and radius of the probe, respectively, and $R_2$ (m) is the radius of the outer barrel. Thixotropy value is measured by the area of hysteresis loop [8,15,35] in Figure 4.

3.3. Printability test
Finally, a printing test was conducted to investigate the printability of designed mixture. As shown in Figure 5, a gantry printer with a 1.2 m × 1.2 m × 1.0 m (L × W × H) printing volume was used to print specimens. The dimension of nozzle opening was 30 mm × 10 mm. Nozzle travel speed and pumping speed are 2,000 mm/min and 650 rpm, respectively. The standoff distance was 30 mm. 3D model used in the printing test is shown in Figure 6.

Table 1. Chemical composition of FA and OPC

<table>
<thead>
<tr>
<th>Formula</th>
<th>FA (%)</th>
<th>OPC (%)</th>
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<tbody>
<tr>
<td>SiO$_2$</td>
<td>58.59</td>
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<tr>
<td>Al$_2$O$_3$</td>
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<td>Fe$_2$O$_3$</td>
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<td>TiO$_2$</td>
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<td>P$_2$O$_5$</td>
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<tr>
<td>Na$_2$O</td>
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<td>ZrO$_2$</td>
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<tr>
<td>MnO</td>
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<td>Cr$_2$O$_3$</td>
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<td>-</td>
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<tr>
<td>CuO</td>
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<tr>
<td>ZnO</td>
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<td>-</td>
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OPC: Ordinary Portland cement, FA: Fly ash

Table 2. Mixture proportion

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<tr>
<th>OPC (g)</th>
<th>Sand (g)</th>
<th>W (g)</th>
<th>FA (g)</th>
<th>SF (g)</th>
<th>Retarder (g)</th>
<th>Accelerator (g)</th>
<th>Superplasticizer (g)</th>
</tr>
</thead>
</table>

Figure 3. Rheological testing programs.

Figure 4. The typical test results of rheology.
Table 3. Coded values via CCD

<table>
<thead>
<tr>
<th>Test run</th>
<th>Retarder Coded value</th>
<th>Retarder Actual value (g)</th>
<th>Accelerator Coded value (g)</th>
<th>Accelerator Actual value (g)</th>
<th>Superplasticizer Coded value</th>
<th>Superplasticizer Actual value (g)</th>
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<tr>
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<tr>
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<td>0</td>
<td>5.08</td>
</tr>
</tbody>
</table>

Figure 5. Gantry printer.

4. Results and discussion

4.1. ANOVA analysis

The rheological testing results are presented in Table 4, including static torque, flow resistance, and torque viscosity. These results are converted into static/dynamic yield stress and plastic viscosity. The ANOVA results, which are shown in Table 5, indicate the significance of the influence of individual and interaction (second-order effect) of factors on the rheological properties. A, B, and C stand for the coded factors of superplasticizer, retarder, and accelerator, respectively. AB, BC, and AC represent the interactive effect of superplasticizer and retarder, retarder and accelerator, and superplasticizer and accelerator, respectively.
In ANOVA, P-value and F-value serve as critical parameters to evaluate the significance of the proposed model and individual parameters. The P-value is the probability of achieving the F-value. A small P-value reveals that the effects of the parameters are statistically significant, while the high F-value indicates that the variation reported by the model is significantly larger than that inherent in the process \[27\]. For example, the factor AC has a F-value of 16.77 and a P-value of 0.0022 associated with static yield stress, and the results indicate that there was only a 0.22% possibility that the F-value of 16.77 occurs because of noise. As can be seen from ANOVA results, except for some cases in AB, AC, and BC, the P-value is generally smaller than 0.05, which implies that the model used for predicting the response is statistically significant.

Figure 7 shows the normability tests. As can be seen from Figure 7, all the points are scattering around a straight line. The result indicates that residuals followed a normal distribution, and static/dynamic yield stress, plastic
viscosity, and thixotropy cannot be further improved by adopting other curve fitting methods.

To exclude the run order effects from various statistical rheological models, the relation between the residual and run order should be confirmed \cite{27}. Figure 8 shows that the points scatter randomly (no grouping) with the run order, which indicates that run order has no significant impact on the responses.

4.2. Prediction models

The coefficients of prediction models are presented in Table 6. A positive value indicates a positive effect on the response, while a negative value implies a negative effect on the response. The magnitude of the absolute value represents how significant the factor is and can be used to rank the importance of each factor. Results in Table 6 suggest that with an increase in the dosage of retarder and superplasticizer, all these rheological parameters decrease while accelerator possesses the opposite influence on the rheological properties. The interaction of retarder and accelerator (AB) has a negative influence on all the rheological properties. The interaction of retarder and superplasticizer (AC) possesses a positive influence on the dynamic yield stress, plastic viscosity, and thixotropy, while it has a negative influence on the static yield stress. The interaction of accelerator and superplasticizer (BC) has a positive influence on the yield stress, whereas it possesses a negative influence on the plastic viscosity and thixotropy.

The derived models for static/dynamic yield stress, plastic viscosity, and thixotropy with respect to the three main factors and their second-order effects can be written as follows:

Static yield stress = $829.1 - 71.89A + 80.27B - 125.5C - 87.19AB - 48.89AC + 17.24BC$ \hspace{1cm} (VIII)

Dynamic yield stress = $166.8 - 18.56A + 8.640B - 10.35C - 11.83AB + 1.400AC + 3.560BC$ \hspace{1cm} (IX)

Plastic viscosity = $20.91 - 1.650A + 1.010B - 0.3685C - 0.0360AB + 1.700AC - 2.230BC$ \hspace{1cm} (X)

Thixotropy = $17369 - 2550A + 1357B - 725.9C - 262.1AB + 1219AC - 244.5BC$ \hspace{1cm} (XI)

The statistical analysis, as shown in Table 6, was also conducted to estimate the statistical accuracy of established models. High R-squared value (larger than 0.8) and high adequate precision (larger than 4) indicate...

Figure 7. Normal probability plots of residuals for different responses: (A) Static yield stress; (B) dynamic yield stress; (C) plastic viscosity; and (D) thixotropy.
that the obtained model is statistically significant. The adequate precision is a signal-to-noise ratio. It compares the range of the predicted values at the design points to the average prediction error\(^2\). Ratios greater than 4 indicate adequate model discrimination. Figure 9 shows the predicted values (calculated from the model) versus the actual values (obtained from experiments). It is clear that the models were successful in capturing the correlation between the dosages of chemical admixtures and material rheological properties with high R-squared values in Table 6.

Figure 10 shows the 3D response surface, in which the coded value of superplasticizer is set as 0. As can be seen from Figure 10, the dosages required for accelerators or retarders to achieve different rheological properties can be found from the contour figures, for example, maximum static yield stress or lowest plastic viscosity.

Table 6. Coefficients of derived models

<table>
<thead>
<tr>
<th>Factors</th>
<th>Static yield stress (Pa)</th>
<th>Dynamic yield stress (Pa)</th>
<th>Plastic viscosity (Pa·s)</th>
<th>Thixotropy (Pa/s)</th>
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<tbody>
<tr>
<td>Interception</td>
<td>829.1</td>
<td>166.82</td>
<td>20.91</td>
<td>17369</td>
</tr>
<tr>
<td>A</td>
<td>−71.89</td>
<td>−18.56</td>
<td>−1.65</td>
<td>−2550</td>
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<tr>
<td>B</td>
<td>80.27</td>
<td>8.64</td>
<td>1.01</td>
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<tr>
<td>C</td>
<td>−125.5</td>
<td>−10.35</td>
<td>−0.3685</td>
<td>−725.9</td>
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<tr>
<td>AB</td>
<td>−87.19</td>
<td>−11.83</td>
<td>−0.0360</td>
<td>−262.1</td>
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<tr>
<td>AC</td>
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<td>1.70</td>
<td>1219</td>
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<tr>
<td>BC</td>
<td>17.24</td>
<td>3.56</td>
<td>−2.23</td>
<td>−244.5</td>
</tr>
<tr>
<td>R2</td>
<td>0.9765</td>
<td>0.9922</td>
<td>0.8354</td>
<td>0.9749</td>
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<tr>
<td>Adequate precision</td>
<td>23.26</td>
<td>41.08</td>
<td>11.20</td>
<td>17.90</td>
</tr>
</tbody>
</table>

Figure 8. The relationship between residuals and run orders: (A) Static yield stress; (B) dynamic yield stress; (C) plastic viscosity; and (D) thixotropy.
Figure 9. Predicted values vs actual values of different responses: (A) Static yield stress; (B) dynamic yield stress; (C) plastic viscosity; and (D) thixotropy.

Figure 10. 3D response surface of different responses: (A) Static yield stress; (B) dynamic yield stress; (C) plastic viscosity; and (D) thixotropy.
4.3. Printability test results

The mixture proportions of No.3 and No.6 in Table 4 were selected in printability test as they show the extremes of the rheological properties. The mixture proportion of No.3 possesses high yield stress (1,156 Pa) and high thixotropy (17,081 Pa/s). On the contrary, No.6 possesses a low yields stress (588.6 Pa) and low thixotropy (7,410 Pa/s). Final printed component with mixture proportion No.3 is illustrated in Figure 11. As shown from Figure 11, it indicates that the mixture proportion of No.3 can be printed well, and material can fully maintain its shape in the printing process.

However, mixture proportion of No.6 is not suitable for printing as shown in Figure 12. Large deformation of the structure occurred during the printing test since material static yield stress is insufficient to keep the printed layers to stand firmly; therefore, it is not suitable for 3D printing. Finally, the structure collapsed due to large deformation and misalignment. Therefore, materials with higher yield stress and higher thixotropy have better printability according to the printing test. The developed statistical models by using CCD can be an efficient method to optimize the dosage of chemical admixtures so that the material has the desired rheological properties, that is, high yield stress and high thixotropy, for 3DCP.

5. Conclusions

In this study, CCD was adopted to investigate the impact of chemical admixtures and their combined interactive effects on the rheological properties with respect of static/dynamic yield stress, plastic viscosity, and thixotropy. Two mixtures with the extremes of the rheological properties were selected for the printing test in this work. The printing test was successfully conducted when the mixture has high yield stress (1,156 Pa) and high thixotropy (17,081 Pa/s). However, the collapse happened during the printing test when materials have a low yields stress (588.6 Pa) and low thixotropy (7,410 Pa/s).

Four polynomial models are constructed to correlate the dosage of chemical admixtures with material rheological properties. The derived models are shown to be statistically significant based on ANOVA analysis. The results indicate that with an increase in the dosage of superplasticizer or retarder, the rheological properties decrease, while accelerator possesses an opposite effect on the rheological properties. The combined interactive effect of retarder and accelerator has a negative impact on the rheological properties. The interactive effect of retarder and superplasticizer possesses a positive impact on the dynamic yield stress, plastic viscosity, and thixotropy while it has a negative influence on the static yield stress. The interactive effect of accelerator and retarder has a positive influence on the yield stress whereas it possesses a negative impact on the plastic viscosity and thixotropy. The CCD derived model is not universally applicable since the results may possibly alter in conjunction with other factors such as the test range of parameters being studied. However, the CCD can minimize the efforts and time consumed in conducting experiments while obtaining sufficient information for data analysis. The method is particularly useful when one has to deal with a large number of variables in experiment. The second-order models can be further obtained through the CCD to make predictions.

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Conflict of interest

The authors report that they have no affiliations with or involvement in any organization or entity with any financial interest in the subject matter or materials discussed in this manuscript.

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