

Development of Premix Internal-Magnesium-Diffusion MgB₂ Wire Using a Data-driven Approach

Akiyoshi Matsumoto, Akimitsu Ishii, Rei Kawasaki, Takahiro Hosokawa, Akiyasu Yamamoto

Abstract— Optimizing various fabrication parameters is one of the major challenges in the development of superconducting wires, often resulting in prolonged transition periods from fundamental research to practical applications. Additionally, escalating costs for essential resources such as liquid helium have amplified the difficulty of experimental work, further underscoring the importance of data-driven research approaches. In this study, we focus on magnesium diboride (MgB₂) wires and demonstrate the effectiveness of Bayesian optimization in efficiently searching complex parameter spaces to identify optimal fabrication conditions. Specifically, we investigate the internal magnesium diffusion (IMD) process, employing Bayesian optimization and the BOXVIA visualization tool to explore key heat-treatment parameters—namely, heat-treatment time and temperature—with the aim of maximizing the engineering critical current density (J_c). Our results show that, under conventional conditions, the highest J_c was achieved at approximately 700 °C with a short holding time of less than one hour. Moreover, our process informatics approach enabled the discovery of optimal conditions even under unconventional parameter settings. This methodology substantially reduces the number of experimental iterations required and enhances the performance of superconducting wires. Overall, our data-driven optimization strategy offers a promising route for faster, more efficient wire fabrication and the accelerated commercialization of superconducting technologies.

Index Terms— Internal magnesium diffusion, Process informatics, Bayesian optimization, MgB₂, Premix-IMD

I. INTRODUCTION

IN recent years, the development of magnesium diboride (MgB₂) wires is actively advancing[1-2], driven by the emerging hydrogen economy and the vast potential of superconducting materials in cryogenic energy applications. High-temperature superconductors exhibiting superconducting transitions above 20 K are expected to play a crucial role in devices that harness the cryogenic energy of liquid hydrogen. Currently, high-temperature superconducting materials available in round wire form are primarily limited to MgB₂ and Bi₂Sr₂CaCu₂O₈. Notably, MgB₂ is distinguished by its relatively low cost and easy accessibility as a raw material[3]. These attributes make it a promising candidate for applications in magnetic resonance imaging magnets, rotating machinery (such as motors), and other related fields[4-6].

The manufacturing process of MgB₂ differs from advanced thin-film production, relying on a straightforward and

conventional fabrication approach. Among the various methods employed, the powder-in-tube (PIT) technique has been extensively utilized since the discovery of MgB₂ due to its simplicity and accessibility[7]. Concurrently, advancements have led to the development of the internal magnesium diffusion (IMD) method, which is particularly notable for achieving high critical current densities (J_c) [8]. Within the PIT approach, two primary methodologies exist: the ex-situ method, which uses pre-reacted MgB₂ as the raw material, and the in-situ method, which involves mixing magnesium (Mg) and boron (B) powders that react post-wire fabrication. The in situ PIT method presents challenges, such as Mg diffusion into the B side, leading to void formation and reduced critical current density(J_c). To address the crucial need for higher packing density and improved performance, the IMD method has emerged as a promising alternative for MgB₂ superconducting wires.

Recently, data-driven research has experienced rapid growth, particularly in materials informatics, where traditional datasets such as the MDR SuperCon[9] and Materials Project[10] have been utilized to explore novel superconducting materials[11,12]. Although no material has yet been identified that surpasses room-temperature superconductivity, these data-intensive methodologies have become powerful tools in aiding material discovery. The integration of machine-learning techniques is accelerating research by uncovering new principles from vast data collections, leading to insights that might otherwise elude human observation. Furthermore, innovative approaches are being developed, involving the robotic synthesis of new materials from extensive combinations of multiple elements. This not only facilitates the identification of compositions with enhanced properties but also significantly reduces the time required for experimental discovery. Additionally, process informatics (PI) is becoming increasingly prevalent, focusing on optimizing processes to achieve the desired properties with minimal data[13,14]. These advancements are transforming materials science research and providing promising pathways to accelerate the discovery and optimization of new substances.

In advancing MgB₂ wire technology, our research group has traditionally concentrated on the PIT method, exploring the impact of various additives and raw material modifications. Efforts to enhance high-field performance have involved an

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extensive examination of different additives, with particular emphasis on carbon-based compounds. Recognizing low packing density as a key factor limiting J_c , we developed an IMD technique. Although this approach achieved packing density of MgB_2 approaching 100%[8], the formation of cavities in the wire core limited improvements in engineering current density (J_c).

Historically, parameter optimization has been confined to narrow domains, guided by experiential insights and historical data. However, questions remain regarding whether past research efforts have fully explored the vast optimization landscape. Recently, Yamamoto *et al.* applied PI to bulk materials, achieving approximately double J_c value[13]. Nonetheless, such applications in superconducting wires remain limited.

In superconducting wire development, although discovering new methods to enhance material properties is valuable, it is equally important to determine optimal solutions with minimal experimental iterations. This consideration is especially relevant given the rising costs of helium gas, which limits experimental frequencies. Consequently, achieving optimal results with fewer experiments is becoming increasingly necessary.

This study aimed to apply the design of experiments—an approach that has gained recent attention—to the Premix-IMD method and evaluate its effectiveness. We begin by reviewing existing data and analyzing their interrelationships, followed by a demonstration of the optimal solution identification process using PI. Finally, we discuss the advantages of this methodology, highlighting its potential in revolutionizing the field of superconducting wire research.

II. Experimental

MgB_2 wires are produced using the internal diffusion method, which overcomes the packing density limitations inherent in the PIT approach. Unlike the conventional method, this technique utilizes an Mg rod of 2 mm in diameter surrounded by B powder (amorphous B powder from Pavezyum Co. Ltd.), where MgB_2 forms through heat treatment following cold processing. In our study, as depicted in Fig. 1, we incorporated pre-manufactured MgB_2 within the B powder during the internal diffusion process. The MgB_2 was synthesized using the method employed by S.Ueda *et al.*[15]. This study refers to the Premix-IMD approach, which integrates Premix-IMD techniques. We experimented with MgB_2 powder additions of 0, 10, and 30%, reduced the material from 6 mm to 0.8 mm in outer diameter of sheath material (Fe), and subjected it to heat treatments under diverse conditions.

After post-processing, the IMD wires were heat-treated across various environmental settings following vacuum sealing. Optimization was performed using Bayesian optimization, a strategy widely applied across multiple scientific domains. This methodology constructs a regression model between the explanatory and target variables through Gaussian process regression. It then employs the estimated values and their variances to identify sample candidates that are

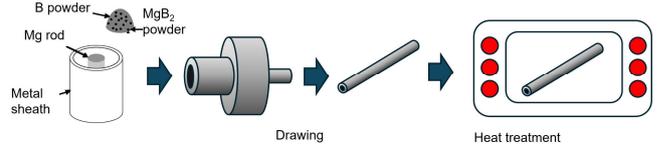


Fig. 1. Premix-IMD manufacturing method. An Mg rod is placed in the center, surrounded by a mixture of B and MgB_2 powder. The wire diameter is then reduced by wire drawing, and MgB_2 is generated by heat treatment.

expected to maximize or minimize the target variable. Essentially, this approach connects data points to approximate the shape of the function, enabling exploration of uncharted territories, while leveraging the best available data.

To visualize this process, we used BOXVIA, a tool for representing Bayesian optimization[16]. To define the parameter optimization search space, we set maximum temperature (T_{\max}) and holding time (t_{hold}) as explanatory variables, ranging from 550 to 900 °C and from 0 to 3000 min, respectively. The target for optimization was J_c at 4.2 K and 7 T. BOXVIA allows multiple points to be proposed simultaneously, a valuable feature given the escalating costs of He, which constrains experimental frequency. To address He consumption, we obtained five proposals from a one-time optimization calculation. In addition, two measurements were conducted for each proposal to account for sample-dependent variations in the measurement results.

III. RESULTS AND DISCUSSION

Defining the search range is a crucial step in Bayesian optimization, as ensuring a realistic range helps avoid challenging scenarios. We leveraged previous findings to guide our choices, extending the range into less frequently explored regions. The initial experiments were conducted under specific boundary conditions, with the results shown in Fig.2. MgB_2 samples demonstrated minimal variance, indicating no sample

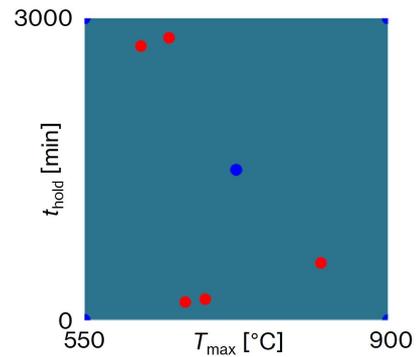


Fig.2. 2D distribution of the mean function. The blue and red points indicate the initial data and the next candidates proposed using Bayesian optimization. Due to the scarcity of data, a uniform distribution indicated by steel blue was obtained, resulting in nearly random proposals. T_{\max} and t_{hold} are maximum temperature and holding time of heat treatment, respectively.

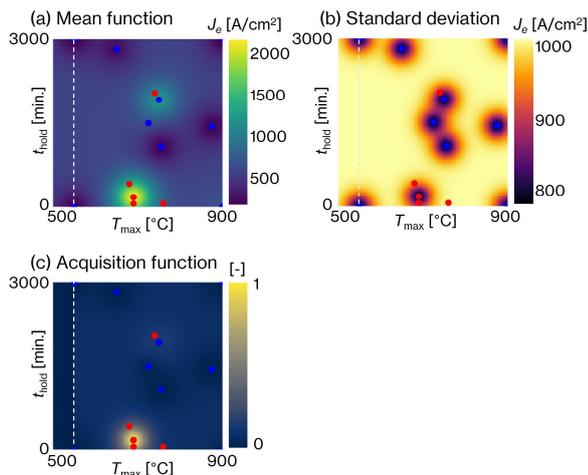


Fig.3. 2D map of (a) the mean function of J_e at 0% composition, (b) its standard deviation, and (c) the acquisition function. The blue and red points indicate the given data and proposed candidates. The white dotted line indicates 550 °C, representing the lower limit of the proposed temperature.

dependency at the representative central points. Contrary to the initial assumption that the J_e would be negligible across all boundary conditions, some configurations yielded small J_e values. These conditions were then used as the baseline for applying BOXVIA, which led to the extraction of five subsequent experimental proposals.

The initial experimental locations, marked in blue, included four boundary points and one central point as shown in 1st try of Fig.2. The subsequent experimental suggestions by BOXVIA, marked in red, highlight the exploratory nature of Bayesian optimization, particularly in data-scarce conditions, resulting in proposals that may appear nearly random. This methodology enables the systematic extension of experimentation into novel parameter spaces, while refining the search for optimal conditions.

Figure 3 presents a two-dimensional (2D) distribution analysis based on experiments conducted using the initial set of proposed points, focusing on the mean function, standard deviation distributions, and the subsequently computed acquisition function. As shown in Fig. 3, the blue markers represent the first set of proposals, while the red markers indicate subsequent experimental proposals. Notably, as shown in part (a), a pronounced peak appears near 700 °C within the short-time heat treatment region, along with a more subtle peak near the central point. These observations suggest that potential candidates for optimal conditions were identified within approximately two exploratory rounds.

Further analysis revealed that the next set of proposed points concentrated on four locations in the region exhibiting the highest values, while also expanding exploration into a secondary peak region. Nevertheless, relying solely on Bayesian optimization could potentially leave portions of the extensive exploration space unexamined. Therefore, additional search spaces beyond those proposed by Bayesian strategies were designated for exploration as the next experimental frontier. The use of BOXVIA not only supports Bayesian

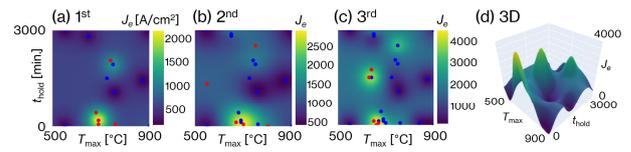


Fig.4. Changes in the mean distribution function obtained after adding the (a) first, (b) second, and (c) third set of experimental data. (d) shows a 3D representation of the 2D distribution shown in (c).

optimization but also enhances experimental visualization, facilitating the identification of unexplored areas. Incorporating experiential insights with Bayesian recommendations is crucial for reducing the number of experimental iterations required.

The results from the three experimental iterations are illustrated in the 2D distribution of the average functions in Fig. 4, with section (d) providing a three-dimensional (3D) visualization for improved clarity. The visual representation confirms that the region around 700 °C for short-time heat treatment consistently yields the highest J_e . Figure 4 plots the mean distribution function of J_e obtained from each experimental iteration, linking the peak J_e achieved per cycle. Initially, the highest J_e reached was approximately 1000 A/cm², which then rapidly tripled. Subsequent increases stabilized by the second and third cycles.

These findings underscore that, despite the expansive search space explored for the MgB₂ IMD method, optimal values can be effectively reached within a few experimental cycles. Although this current study targeted optimal values at 4.2 K and 7 T, modifying the target parameters allows for the exploration of heat treatment conditions tailored to various properties with relatively few experiments. Thus, Bayesian optimization not only helps new researchers navigate general exploration areas but also facilitates progression towards more detailed assessments, thereby optimizing research efficiency.

Figure 5 illustrates the unique results obtained from the Premix-IMD method incorporating various amounts of MgB₂ powder. This includes scenarios with 10 and 30% MgB₂ powder, along with a 0% powder sample for comparison. In Bayesian optimization, candidate data points for subsequent experiments were identified at peaks in the acquisition function, which was calculated using both the mean function and standard deviation. As this iterative process progressed into the second and third rounds, there is a clear trend towards the convergence of the J_e values.

Analysis of the proposed points revealed three discernible peaks within the operational space. The presence of a peak at approximately 700 °C within a shorter time frame was consistent with traditional methodologies. However, the emergence of peaks at higher temperatures and extended heat-treatment durations represents a novel and unexpected finding.

Bayesian optimization is expected to minimize the number of experimental iterations, thereby enhancing the experimental efficiency. Furthermore, recent findings have underscored its

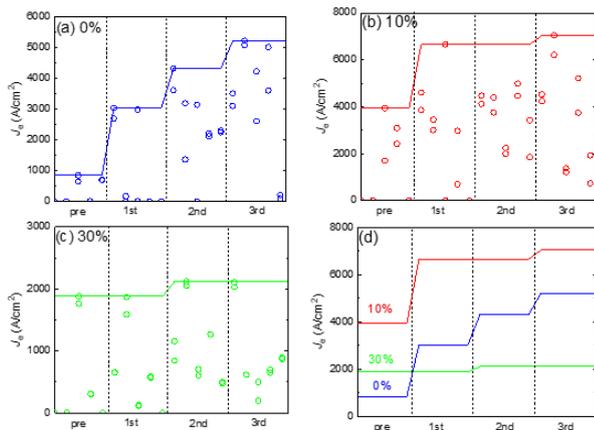


Fig. 5. Comparison of J_c transitions and the maximum value obtained at each proposal number: (a) 0% composition, (b) 10% composition, (c) 30% composition, and (d) maximum value transitions.

efficacy in identifying distinct material characteristics by defining optimal exploration ranges[16,17]. In this study, a comprehensive exploration of the entire parameter space under the specified boundary conditions — ranging from 550°C to 900°C in 10°C increments, and from 0 to 3000 minutes in 10-minute intervals — traditionally demands 35 x 300 experimental trials. Remarkably, our approach unveiled the general trend with only 20 sampling points (4 x 5), highlighting its efficacy in substantially reducing the number of experiments required. Since these experimental findings were systematically integrated into a comprehensive database, they have the potential to enrich the correlation diagrams presented in Fig. 4 with new and insightful data. Although the current phase necessitates extensive data acquisition, this study demonstrates how PI can play a pivotal role in effectively optimizing data collection strategies. We have performed Bayesian optimization on MgB₂ wires fabricated via premix IMD. Our findings suggest that Bayesian methods can be effectively employed to optimize the heat treatment process for superconducting joints in MgB₂. Furthermore, these methods hold potential in identifying optimal heat treatment conditions for enhancing the efficacy of pinning centers in REBCO wires[18].

IV. CONCLUSION

The study on MgB₂ superconducting wires, initially perceived as data-limited, revealed substantial data potential through the systematic organization of fabrication parameters. This study employed the internal diffusion method for wire production to enhance traditional techniques. Furthermore, different MgB₂ powder percentages were tested, with Bayesian optimization streamlining the experimental process. This approach utilized Gaussian process regression models to identify optimal experimental parameters, with the BOXVIA tool effectively defining the search space for parameters such as temperature and heat treatment. The results highlighted unexpected J_c values under certain conditions and revealed distinct peaks in the experimental space, thereby identifying novel optimal conditions at higher temperatures and longer durations.

Consequently, Bayesian optimization proved effective in reducing experimental iterations and expanding exploration ranges, thus enabling the efficient identification of unique material properties.

V. ACKNOWLEDGEMENT

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