Autonomous robotic experimentation system for powder X-ray diffraction

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Abstract

Accelerating materials research using AI technologies requires high-quality, reliable data across a wide range of measurement conditions. Powder X-ray diffraction (PXRD) is crucial for analyzing crystal structures and quantifying phase compositions in materials science. However, PXRD reliability heavily depends on accurate sample preparation and data analysis, including precise measurements over a broad angular range, especially at low angles. Recent advancements in AI-driven PXRD data analysis have improved accuracy and efficiency, shifting the bottleneck and reproducibility issues to manual sample preparation and measurement processes. To address these challenges, we developed an autonomous robotic experimentation (ARE) system for PXRD that integrates sample preparation, measurement, and data analysis into a single automated workflow. Our system achieves high precision and reproducibility in sample preparation, enabling quantitative phase analysis with only one-hundredth of the conventional sample quantity (reducing from $300\,\mathrm{mg}$ to $3\,\mathrm{mg}$) while maintaining a standard deviation below $1\,\%$. By combining robotic precision with machine learning-based data analysis, our approach enhances reproducibility and enables more efficient materials discovery compared to traditional manual methods.

1 Introduction

As artificial intelligence (AI) technologies are increasingly utilized in materials science, the importance of reliable data and comprehensive measurements has grown significantly. These data serve as reference databases for human researchers as well as essential training data for AI algorithms. Among various experimental techniques, powder X-ray diffraction (PXRD) provides crucial data in materials science, offering diverse information such as crystal structures [1–3], phase identification and quantification [4], and crystal polymorph characterization [2,5–9]. The versatility and importance of PXRD data make it a cornerstone in materials research and development. To ensure the reliability of PXRD data, accurate sample preparation and precise measurement data analysis are crucial. Recent developments in AI-driven automated analysis methods have greatly improved the accuracy and efficiency of data interpretation [10–12]. However, this progress has highlighted a new challenge: sample preparation and measurement processes have become the bottleneck in the workflow.

Laboratory automation using robots to perform repetitive tasks reproducibly has become increasingly important in materials research and development [13–24]. This trend towards automation has naturally extended to PXRD experiments, with several research groups making significant contributions to the development of automated systems for PXRD sample preparation and analysis [22,24,25]. While these studies have made significant strides in PXRD automation, several critical challenges remain to be fully addressed. These include ensuring sample homogeneity, achieving optimal surface smoothness, and effectively reducing background noise, particularly in the low-angle region.

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The minimization of background noise is a critical challenge in obtaining high-quality PXRD measurement data. Background noise affects analysis results, potentially hindering the accurate estimation of important parameters such as weight fractions. Conventional methods of fixing powders on sample holders, such as the use of grease or Kapton tape, often increase background noise, which is particularly problematic for low-angle measurements. The accuracy of low-angle measurements is crucial for characterizing a wide range of materials, including organic compounds [26,27] and lead halide perovskites [28–31], where it is essential for identifying reactants, products, and verifying reaction occurrences. Therefore, minimizing background noise and enhancing low-angle measurement precision are critical for expanding the applicability of automated PXRD systems to a broader range of materials, including complex structures like metal-organic frameworks.

To address these challenges, we have developed an autonomous robotic experimentation (ARE) system for PXRD that aims to achieve high-precision quantitative analysis. This ARE system integrates and automates the entire PXRD workflow, from sample preparation to measurement and data analysis. By combining robotic precision in sample preparation with data analysis through black-box optimization, our system demonstrates significant progress in PXRD automation, potentially offering improvements in efficiency and data quality compared to conventional methods.

Our approach minimizes human error, enabling efficient acquisition of reproducible, high-quality data, particularly in the low-angle region.

In summary, our contributions are as follows:

1. We developed an autonomous experimentation system for PXRD that integrates the entire workflow from sample preparation to data analysis; compared to manual methods, our system achieved high precision and reliability in sample preparation.
2. We demonstrated the ability of precise robotic sample preparation to obtain low-background patterns, especially at low angles.
3. We investigated the effect of the sample quantity on the accuracy and consistency of quantitative analysis using our autonomous system; thus, reliable results could be obtained with reduced sample amounts than those used in manual preparation methods.
4. We validated the autonomous system with different mixture ratios and showed its accuracy in quantifying phase compositions.

2 Development of the ARE system for PXRD

We have developed the ARE system for PXRD that integrates sample preparation, measurement, and data analysis into a single automated workflow. Figure 1 shows the ARE system's workflow and components.

The key components of our ARE system are as follows:

1. The 6-axis robotic arm (DENSO, COBOTTA) with a multifunctional end effector is capable of preparing powder samples and transferring the sample holders. (Figure 2-a)
2. A detachable protective cover made of paper is used to protect the soft gel attachment on the robotic arm's end effector from environmental contamination. The robotic arm's automated movements enable the cover to be attached before sample preparation and detached after use. This ensures a clean environment for each sample without human intervention.
3. An XRD instrument (Rigaku, MiniFlex 600-C) is equipped with a single-axis actuator to control the door. The single-axis actuator enables the automatic opening and closing of the doors of the XRD instrument.
4. A custom-designed sample holder with a frosted glass surface and embedded magnets serves as a sample support during XRD measurements. (Figure 2-b)
5. A drawer-based sample hotel serves as a storage unit for multiple sample holders and has 20 tiers to accommodate up to 40 samples in total.
6. A sample preparation station is used for processing powder samples into an optimized form for XRD analysis; this features an integrated pull-out funnel for precise centering of the powder within the holder.

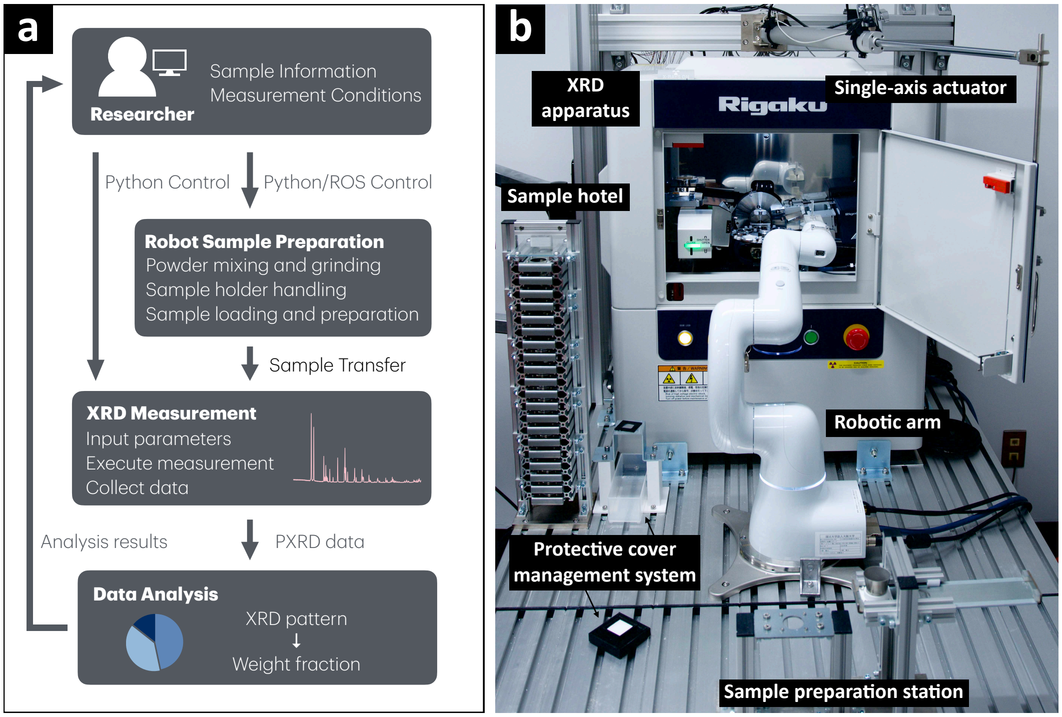

Figure 1: Autonomous PXRD system overview. (a) Schematic of the autonomous PXRD experimental workflow. (b) Key components of the system setup.

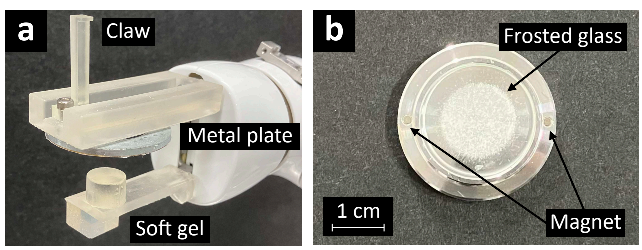

Figure 2: Close-up view of the key components of the autonomous PXRD system. (a) Multifunctional end effector. (b) Custom-designed sample holder.

The end effector of the robotic arm (Figure 2-a) is a key component of our ARE system. It integrates three attachments: a claw for drawer manipulation, a metal plate for magnetic coupling with the sample holder, and a soft gel for surface flattening. This integrated design improves system efficiency and autonomy, allowing for sample preparation, loading, and unloading without changing attachments. The soft gel attachment is crucial for creating low-background patterns by gently and uniformly pressing the powder sample, resulting in a smooth surface that minimizes background noise.

Our custom-designed sample holder (Figure 2-b) is a crucial component of the ARE system. It features a frosted glass central area that supports the powder sample while reducing background noise, particularly beneficial for low-angle measurements. The frosted surface also prevents the powder from slipping off during manipulation. Embedded magnets in the outer frame of the sample holder ensure secure attachment to the metal plate of the end effector, enabling precise and stable sample handling throughout the automated process, from transfer to measurement.

The integration of these components into the ARE system allows for a fully automated XRD sample preparation and measurement process, reducing human errors and improving reproducibility. The system's modular design enables adaptation to other analytical methods, with components such

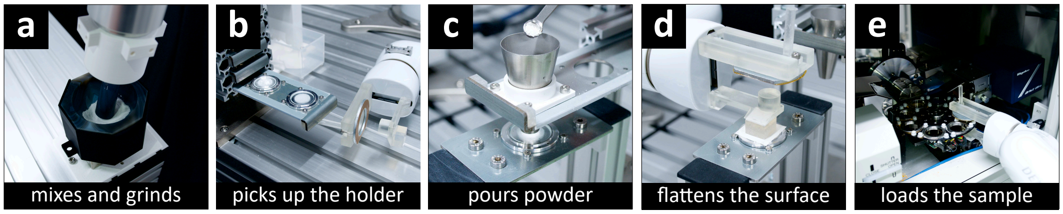

Figure 3: Key processes performed by the robotic arm in the autonomous PXRD system. (a) The robotic arm mixes and grinds the powder sample to ensure homogeneity and appropriate particle size. (b) The robotic arm picks up the sample holder using a multifunctional end effector. (c) In this study, the prepared powder sample is manually poured into the sample holder due to the need to measure the amount of powder used. To note, the robotic arm is technically able to perform this step; however, in the current experiment, this was manually performed. (d) Using a soft gel attachment on the end effector, the robotic arm gently flattens the surface of the powders to ensure a smooth and even surface for the XRD measurement. (e) The robotic arm loads the prepared sample into the XRD instrument for measurement.

as the robotic arm, sample hotel, and control software being adaptable to different measurement configurations.

3 Detailed workflow of the autonomous PXRD system

The PXRD system operates according to a workflow that reduces human intervention through automation. Figure 3 illustrates the key processes in our PXRD system workflow. The operation of our system can be viewed in a demonstration video[1]. The detailed steps in the workflow are as follows:

Step 1: The researcher sends a command to the control PC specifying the sample information and measurement parameters.

Step 2: The control PC processes the command and sends instructions to the robotic arm and the XRD instrument.

Step 3: The robotic arm (Universal Robots, UR5e) mixes and grinds the powder sample to ensure homogeneity, as described in our previous work[32,33]. (Figure 3-a)

Step 4: The robotic arm retrieves the specified sample holder from the sample hotel and transfers it to the sample preparation station. (Figure 3-b)

Step 5: The ground powder is filled into the sample holder placed at the sample preparation station. While the robotic arm is technically capable of performing this step autonomously, in this study, it was done manually to measure the precise amount of powder used. (Figure 3-c)

Step 6: The robotic arm uses soft gel attached to the end effector to gently flatten the surface of the powder sample. (Figure 3-d)

Step 7: The XRD instrument automatically opens its door using a single-axis actuator, and the robotic arm loads the prepared sample into the instrument. (Figure 3-e)

Step 8: The XRD instrument closes its door, and the measurement software is automatically controlled using the PyAutoGUI library to start the measurement according to the specified parameters.

Step 9: After the measurement is complete, the XRD instrument sends the raw data (XRD pattern) to a workstation, and the robotic arm retrieves the sample from the XRD instrument and returns it to the sample hotel.

Step 10: The workstation automatically analyzes the XRD pattern using an automated Rietveld analysis method; the pattern is converted into weight fractions of its constituent phases.

[1] `https://youtu.be/nXfL7gmZPMw`

Step 11: The analyzed results are sent back to the researcher, and the automated PXRD experiment is complete.

Our developed autonomous PXRD system provides two main advantages over traditional manual methods. First, automation of the workflow from sample preparation to data acquisition aims to reduce human intervention and increase reproducibility of the experimental results. The automated system helps maintain consistent measurement conditions across experiments. Second, the system improves sample throughput by enabling continuous operation. This allows for analysis of multiple samples sequentially, while researchers can focus on tasks such as data interpretation and experiment planning.

4 Automation of quantitative XRD data analysis

Automating the analysis of material characterization data is a key component of a fully closed-loop autonomous measurement workflow. As discussed in the previous section, our developed autonomous PXRD system integrates the entire process from sample preparation to data acquisition. To complete the automation process and achieve a fully self-operating PXRD experiment, automating the analysis of the acquired data is needed.

PXRD experiments often use a data analysis method called Rietveld analysis, which enables the quantitative analysis of the phase composition, lattice constants, and other structural parameters. However, Rietveld analysis is a time-consuming and effort-intensive task. This analysis involves the optimization of a large number of parameters, which can be several dozen. These parameters include not only those related to the crystal structure but also those related to the equipment and other aspects, such as line shape or background. Additionally, the quality of Rietveld analysis results may vary depending on the researcher's skill and experience, potentially impacting the reliability of the analytical outcomes. Such variability could introduce uncertainties in the interpretation of PXRD data.

To address this challenge, Lee et al. developed an automated Rietveld analysis approach that utilizes machine learning techniques [10]. Their method employs a convolutional neural network (CNN) for phase identification, followed by support vector regression (SVR) for phase fraction prediction. This approach demonstrated high accuracy in both synthetic and real-world datasets. However, it requires a large amount of training data and may struggle with previously unseen phases or complex mixtures.

Szymanski et al. proposed a different approach that does not rely on Rietveld analysis [11]. Instead, they introduced a machine learning method that uses a dual representation of XRD patterns and pair distribution functions (PDFs). This method showed improved accuracy in phase identification, especially for low-intensity features and in the presence of experimental artifacts. However, it may face challenges in providing detailed structural information compared to Rietveld-based methods.

Ozaki et al. introduced a novel approach called BBO-Rietveld, which combines Bayesian Black-box Optimization (BBO) with traditional Rietveld refinement [12]. This method treats the Rietveld refinement process as a black-box optimization problem, using the tree-structured Parzen estimator (TPE) algorithm to efficiently search the high-dimensional parameter space. BBO-Rietveld demonstrated superior performance compared to both human experts and rule-based automation systems.

After careful consideration of these approaches, we chose to integrate the BBO-Rietveld method into our autonomous PXRD system. The Bayesian optimization approach employed in BBO-Rietveld allows for efficient exploration of the parameter space, potentially surpassing human expert performance in terms of speed and accuracy. This efficiency is crucial for high-throughput materials characterization, enabling rapid analysis of a large number of samples. Furthermore, BBO-Rietveld can be easily integrated with existing crystallographic software packages, facilitating its implementation in our automated workflow. This integration capability is particularly important as it allows us to leverage well-established crystallographic analysis tools while benefiting from advanced optimization techniques. By incorporating BBO-Rietveld into our autonomous PXRD system, we aim to achieve high-quality, efficient, and adaptable quantitative phase analysis. This integration brings us closer to realizing a fully self-operating materials characterization platform capable of handling a wide range of samples and experimental conditions.

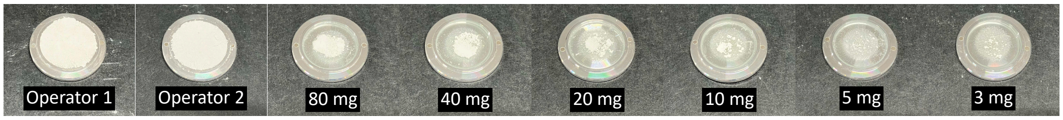

Figure 4: Photographs of the TiO_2 samples prepared by manual operation and the robotic arm in different quantities.

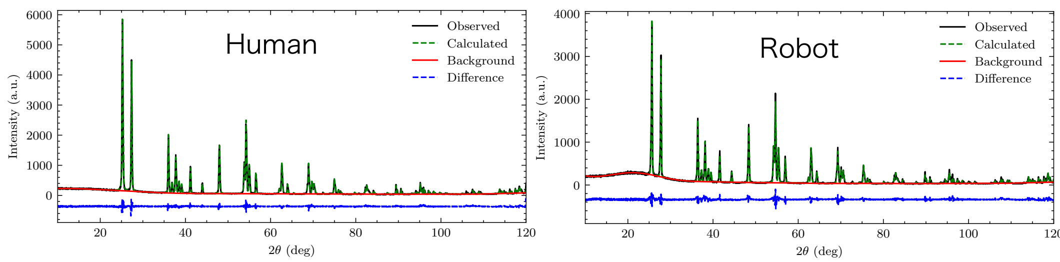

Figure 5: Comparison of the XRD patterns and Rietveld refinement results for samples prepared by manual operation and the robotic arm ($80\,\mathrm{mg}$).

5 Results and discussion

In this study, we conducted three experiments to evaluate the performance and capabilities of the autonomous PXRD system, using titanium dioxide (TiO_2) in its anatase and rutile phases as a model system.

The first experiment, described in Section 5.1, focused on assessing the precision of the robotic arm in sample preparation and its impact on the accuracy of quantitative XRD analysis. The goal was to determine whether the automated system could achieve results comparable to or better than those of the manual sample preparation methods in terms of reproducibility, consistency, and reliability.

The second experiment, detailed in Section 5.2, investigated the effect of the sample quantity on the accuracy and precision of the quantitative XRD analysis. By preparing samples with varying amounts of material using the robotic arm, we aimed to identify the minimum sample quantity needed to obtain reliable anatase content results with a target standard deviation of less than $1\,\%$ for the replicate measurements.

Finally, the third experiment, presented in Section 5.3, validated the autonomous system's performance using samples with different mixture ratios of anatase and rutile TiO_2. The purpose of this experiment was to demonstrate the system's accuracy in quantifying the phase compositions across a range of sample compositions; this aspect is essential for high-throughput material discovery and optimization.

Through these three experiments, we aimed to comprehensively evaluate the capabilities, limitations, and potential of the autonomous PXRD system for advancing material research and accelerating the discovery of new materials with desirable properties. All XRD measurements were performed under standardized conditions to ensure consistent and high-quality data acquisition across various sample types. The detailed measurement parameters, including radiation characteristics, voltage, current, scan specifications, and beam conditioning, are provided in Appendix A.1.

5.1 Powder sample preparation by the robotic arm

We evaluated an autonomous PXRD system to investigate the precision of robotic arms in sample preparation and its impact on the accuracy of the quantitative XRD analysis. Our objective was to assess whether an autonomous PXRD system using a robotic arm could enhance the reproducibility, consistency, efficiency, and accuracy of the PXRD sample preparation. We hypothesized that the robotic arm could reduce human error, increase throughput, standardize procedures, and ultimately match or even surpass the accuracy and consistency of the manual sample preparation methods.

Table 1: Comparison of the sample preparation methods: Manual operation and robotic arm.

Method	Average Anatase Content (%)	Standard Deviation (%)
Operator 1	52.4	0.4
Operator 2	52.2	0.1
Robotic Arm	51.6	0.7

We used a robotic arm to prepare samples containing precise quantities of TiO_2 in its anatase and rutile phases. Figure 4 shows the samples prepared by manual operation and the robotic arm. The anatase reagent (Kojundo Chemical Laboratory, $99\,\%$ purity) did not contain any detectable rutile impurities; however, the rutile reagent (Kojundo Chemical Laboratory, $99.99\,\%$ purity, $2\,\mu\mathrm{m}$ particle size, $>90\,\%$ rutilization rate) contained approximately $2.8\,\%$ anatase as an impurity. To determine the impurity levels, samples containing only the individual reagents were prepared and analyzed using the BBO-Rietveld method prior to the main experiment. Equal amounts of the two reagents were mixed; this resulted in samples with anatase and rutile weight fractions of approximately $51.4\,\%$ and $48.6\,\%$, respectively. Each sample prepared by the robotic arm weighed $80.0\,\mathrm{mg}$, while samples prepared by human operators weighed approximately $300\,\mathrm{mg}$. To ensure consistency, the robotic arm's repeatability was evaluated across multiple experiments. The resulting data were analyzed via the BBO-Rietveld method.

Figure 5 shows a comparison of the XRD patterns and Rietveld refinement results for the samples prepared by the robotic arm and manual operation. The good agreement between the measured and calculated patterns, along with the small residuals, demonstrates the high quality of the samples prepared by both methods. Importantly, compared to previous studies using automated sample preparation methods, the robotic arm-prepared samples exhibit lower background intensity at low angles [22,25]. This achievement can be attributed to the robotic arm's precise control in Step 6; this involves gently pressing the powder to create a smooth surface, effectively minimizing the unwanted background signals that often hinder accurate analysis in the low-angle region. The comparable background levels between the robotic arm-prepared samples and those prepared by skilled human operators confirm the effectiveness of the automated sample preparation technique in producing high-quality samples suitable for a wide range of materials, including those with important structural features at low angles.

Table 1 lists the average anatase content and standard deviation for samples prepared by two human operators (Operator 1 and Operator 2) and the robotic arm. The results demonstrate the variability in the sample preparation between different human operators, as indicated by the differences in the average anatase content and standard deviation. Although the standard deviation of the robotic arm is slightly larger than that of the human operators, it is still sufficiently low; these results indicate that the automated sample preparation can achieve a level of consistency comparable to that of human operators. Thus, automated sample preparation has the potential to reduce human-induced variability.

The reliability and standardization potential of the robotic arm show the importance of integrating automated sample preparation with automated XRD measurements and data analysis for the future of material science research.

5.2 Optimizing the sample quantity for quantitative analysis

We investigated the effect of the sample quantity on the accuracy and precision of the quantitative XRD analysis by conducting an experiment to determine the minimum sample quantity needed to obtain the anatase content results with a standard deviation of less than $1\,\%$ for the replicate measurements. A robotic arm was used to prepare samples with six different amounts (3.0, 5.0, 10.0, 20.0, 40.0, and $80.0\,\mathrm{mg}$) of the TiO_2 mixture; this was the same mixture used in Section 5.1. For each sample quantity, the robotic arm was used to prepare five separate samples; this resulted in a total of 30 samples (6 quantities $\times$ 5 replicates). Figure 4 shows the samples prepared by the robotic arm in different quantities.

Figure 6-a shows the XRD patterns of the different quantities of the TiO_2 samples prepared by the robotic arm. Based on these patterns, although the peak intensities decrease as the sample quantity decreases, the overall shapes of the patterns are maintained, and the background intensities remain sufficiently low; these results show the consistency of the automated sample preparation method.

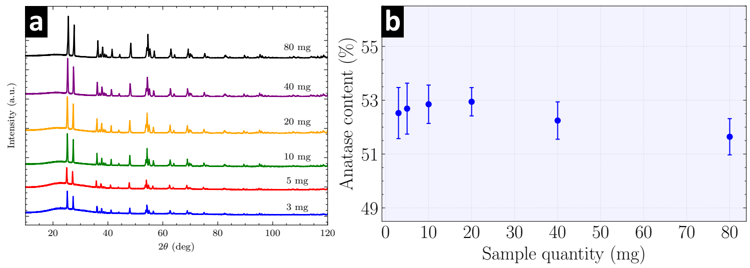

Figure 6: XRD analysis of TiO_2 samples with varying quantities. (a) XRD patterns for different sample quantities. (b) Effect of sample quantity on anatase content precision, with error bars representing standard deviation.

Table 2: Quantitative analysis results for the anatase content in TiO_2 samples with different mixture ratios prepared by the autonomous system.

Mixture Ratio (Anatase:Rutile)	Average Anatase Content (%)	Standard Deviation (%)
9:1	89.8	1.1
7:3	70.9	0.7

Figure 6-b shows the quantitative analysis results for the anatase content of the TiO_2 samples. Each data point is the average anatase content from the five samples at each sample quantity, and the error bars represent the standard deviation. The results indicate that as the sample quantity decreases, increased variability is observed in the estimated anatase content between the samples and is shown by the larger error bars. However, even at the minimum sample quantity of $3.0\,\mathrm{mg}$, which is the lowest amount our system can prepare, the standard deviation of the anatase content was found to be $0.9\,\%$; thus, quantitative results with the desired precision can be obtained.

When proper sample preparation and analysis techniques are employed, reliable quantitative data can be obtained even with small sample quantities. This finding is significant considering that manual sample preparation typically involves using sample quantities of approximately $300\,\mathrm{mg}$. Our system was able to automatically prepare the samples with sufficient accuracy and precision for the quantitative analysis using only $1\,\%$ of the sample quantity traditionally used in manual preparation. By minimizing the sample quantity, the amount of material needed for synthesis can be reduced, potentially leading to shorter synthesis times and more efficient use of resources. As a result, high-throughput material characterization workflows can become more efficient, enabling the rapid analysis of a larger number of samples. The optimization of the sample quantity, along with other key parameters, is essential for the development of efficient and reliable autonomous XRD systems for material discovery and optimization.

5.3 Validation of the autonomous system with different mixture ratios

To further validate the effectiveness of the autonomous PXRD system, we prepared samples with different mixture ratios of anatase and rutile TiO_2 using the optimized sample quantity of $3\,\mathrm{mg}$ determined in Section 5.2. Two mixture ratios were tested: anatase:rutile = 9:1 and 7:3 by weight. For each mixture ratio, the robotic arm was used to prepare five samples $3\,\mathrm{mg}$ for each mixture; this resulted in a total of ten samples (2 ratios $\times$ 5 replicates).

Table 2 provides a summary of the quantitative analysis results for the anatase content in the TiO_2 samples with different mixture ratios. The average anatase contents for the 9:1 and 7:3 mixtures were found to be $89.8\,\%$ and $70.9\,\%$, respectively. These values were in good agreement with the expected anatase content based on the prepared mixture ratios and demonstrated the accuracy of the autonomous system in quantifying the phase compositions. Moreover, the low standard deviations ($1.1\,\%$ for 9:1 and $0.7\,\%$ for 7:3) of the anatase content for each mixture ratio highlight the high precision and reproducibility of the sample preparation and measurement processes performed by the autonomous system.

These results further validate the effectiveness of the autonomous PXRD system in accurately and precisely characterizing materials with different phase compositions, even when using small sample quantities. This capability is beneficial for high-throughput material discovery and optimization, where the ability to reliably analyze a large number of samples with varied compositions is essential.

5.4 Limitations

While our autonomous PXRD system demonstrates significant improvements in sample preparation and analysis, it is important to acknowledge certain limitations:

1. Although we investigated TiO_2 as a typical sample, the optimal sample quantity may vary when using different reagents. Further studies with a wider range of materials are necessary to establish the system's versatility across various compounds.
2. Our sample preparation method may not be universally applicable, particularly for materials with significantly different physical properties such as viscosity. Some materials may require alternative preparation techniques, which could limit the current system's applicability in certain cases.

These limitations highlight the need for continued research and development to expand the system's capabilities and ensure its broad applicability in materials science.

6 Conclusion

In this study, we developed an autonomous robotic experimentation system for PXRD that integrates sample preparation, measurement, and data analysis into a single automated workflow. Our system demonstrates significant improvements in efficiency, reproducibility, and reliability compared to traditional manual methods. Key contributions include:

1. Development of an integrated system achieving high precision and reliability in sample preparation.
2. Demonstration of low-background patterns, particularly at low angles, crucial for characterizing complex materials.
3. Optimization of sample quantity, enabling reliable results with significantly reduced material usage.
4. Validation across different mixture ratios, showcasing the system's accuracy in quantitative phase analysis.

Future work will focus on extending the system's capabilities to include automated reagent handling, aiming to achieve a fully closed-loop material research process. Recent advancements in robotic manipulation, such as dual-arm solid dispensing systems[34], demonstrate the feasibility of further automating complex tasks. By integrating similar capabilities into our autonomous PXRD system, we aim to accelerate material discovery and optimization processes.

Acknowledgments

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A Appendix

A.1 XRD Measurement Conditions

All XRD measurements were performed under the following conditions:

- Radiation: Cu Kα (λ = 1.5419 Å)
- Voltage: $40\,\mathrm{kV}$
- Current: $15\,\mathrm{mA}$
- Scan range: $10°$–$120°$ 2θ
- Step size: $0.01°$
- Scan speed: $4\,°/\mathrm{min}$
- Sample spin rate: $80\,\mathrm{rpm}$

The incident beam was conditioned using a $5\,\mathrm{mm}$ incident slit and a $1.25°$ divergence slit to control and reduce the irradiated area on the sample surface.

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