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Integrated classical/quantum–classical neural networks, DFT, and experiments for predicting bandgap and color of $\text{Cr}_x\text{Sb}_x\text{Ti}_{1-2x}\text{O}_2$ yellow-orange pigments

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Abstract

The accurate prediction of the bandgap (E_g) and color of ceramic pigments is of considerable importance in materials science research. In this work, we investigate the E_g and color of $\text{Cr}_x\text{Sb}_x\text{Ti}_{1-2x}\text{O}_2$ rutile pigments by combining experimental, density functional theory (DFT and DFT+ U) calculations, and neural network approaches. The main novelty of this study is the application of classical/hybrid quantum–classical neural networks to the prediction of pigment color and E_g . Cr/Sb co-doped rutile pigment was synthesized and characterized, and the electronic structures of undoped and doped TiO_2 were analyzed using first-principles calculations. In parallel, classical and hybrid quantum–classical neural network models were developed and trained on datasets compiled from the Materials Project database and literature-reported rutile-based pigments. The models successfully captured the trends in E_g and chromaticity, with the hybrid architectures providing slightly improved predictive performance and more consistent agreement with experiment. Overall, this work demonstrates an effective framework that links experiment, first-principles calculations, and classical/quantum–classical neural networks for predicting key properties of ceramic pigments, and it points toward the future use of fully quantum machine learning models for materials design.

Keywords: Neural network, DFT, Rutile, Color, Bandgap

1 Introduction

Recent advances in computational power, algorithm development, and the growing availability of large experimental and computational datasets have made classical machine learning an important tool for analyzing high-dimensional data and predicting material properties. In materials science, these approaches are increasingly used together with first-principles methods such as DFT to accelerate the discovery and design of new materials by revealing relationships between composition, structure, and physical properties. Nevertheless, accurately describing electronic structure and related quantities such as the E_g remains computationally demanding for many complex systems. At the same time, progress in quantum computing and the emergence of noisy intermediate-scale quantum (NISQ) devices have stimulated interest in parameterized quantum circuits and variational quantum algorithms as new computational models. These developments have led to quantum machine learning, where quantum circuits are combined with classical optimization routines for data-driven tasks. Because current quantum hardware is still limited by noise, qubit number, and circuit depth, hybrid quantum-classical approaches provide a practical framework that leverages classical processing while exploiting the ability of quantum circuits to represent complex correlations and entangled states [1–5].

Several studies have begun to investigate how quantum-enhanced learning models can address problems in materials science. For example, a hybrid classical-quantum approach based on a variational quantum classifier has been used to distinguish ABO_3 perovskites from non-perovskite structures [6]. Similarly, a quantum support vector classifier combined with data-balancing techniques such as SMOTE-ENN has been applied to predict perovskite structures [7]. Beyond structure prediction, quantum machine learning has also been explored in electronic-structure calculations, where hybrid algorithms incorporating restricted Boltzmann machines have achieved accurate estimates of ground state energies and potential energy surfaces for small molecules [2]. In the context of materials informatics, quantum neural networks have further been investigated for multivariate regression problems, showing favorable generalization behavior in tasks such as predicting the melting points of metal oxides [8]. With respect to electronic properties, recent work has shown that quantum circuit learning can successfully classify whether the E_g in perovskite oxides is direct or indirect [9]. Despite these advances, existing studies have largely remained limited to classification tasks or qualitative property prediction. However, the direct prediction of the numerical value of the E_g in crystalline materials using hybrid quantum-classical neural network architectures remains largely unexplored, pointing to an important gap in the current literature.

Titanium dioxide (TiO_2) is an abundant and inexpensive polymorphic compound that occurs mainly in three crystalline forms: rutile, anatase, and brookite. Among these, rutile is the stable phase, while anatase and brookite are metastable. Rutile (space group $P4_2/mnm$) and anatase (space group $I4_1/amd$) both possess tetragonal crystal structures composed of TiO_6 octahedra, which share two edges in rutile and four edges in anatase. In the rutile phase, the structure consists of chains of TiO_6 octahedra connected through vertex sharing along the c -axis, enabling the incorporation of many transition metals and even heavier elements. Owing to its robust structure and

excellent optical properties, rutile is widely used as a ceramic pigment [10–13]. The $\text{Cr}_x\text{Sb}_x\text{Ti}_{1-2x}\text{O}_2$ is a widely used rutile-based ceramic pigment with a yellow-orange color. It has been synthesized by different experimental methods in recent years [12–15], but it has not yet been investigated from a computational perspective. A previous first-principles study examined the electronic structure of Cr/Sb co-doped TiO_2 for the anatase phase [16]. To the best of our knowledge, a similar theoretical analysis for the rutile phase is still lacking, despite its practical importance as a ceramic pigment.

Over the past few years, we have conducted several studies on the properties of ceramic pigments and on predicting their color using DFT [10, 17–22]. In our previous work [23], we investigated the prediction of the E_g and color of a cobalt-doped gahnite ceramic pigment by combining classical machine learning, DFT calculations, and experimental approaches. Building on this research, in the present study, we implement for the first time, the prediction of pigment color using classical/quantum–classical neural networks. In addition, motivated by the existing gap in the literature, we also address the prediction of the E_g using these approaches. As a model system, $\text{Cr}_x\text{Sb}_x\text{Ti}_{1-2x}\text{O}_2$ ceramic pigments were selected. This work aims to predict the E_g and color of $\text{Cr}_x\text{Sb}_x\text{Ti}_{1-2x}\text{O}_2$ by integrating classical and hybrid quantum–classical neural networks with DFT calculations and experimental methods. To this end, the $\text{Cr}_{0.0625}\text{Sb}_{0.0625}\text{Ti}_{0.875}\text{O}_2$ pigment was synthesized and characterized. DFT and DFT+ U calculations were performed for undoped rutile TiO_2 , as well as for a rutile structure doped with one Cr atom and one Sb atom, and the corresponding electronic properties were investigated. In parallel, four predictive models, including classical and hybrid quantum–classical architectures, were developed. For E_g prediction, a dataset containing 1527 materials with tetragonal crystal systems and energy gaps in the range of 1–2 eV was constructed from the Materials Project database, and two prediction scenarios were examined. The E_g of rutile TiO_2 was then predicted and compared with the DFT and DFT+ U results. For color prediction, we constructed a dataset comprising 195 rutile-based ceramic pigments collected from the literature. The colors of $\text{Cr}_x\text{Sb}_x\text{Ti}_{1-2x}\text{O}_2$ pigments ($x = 0, 0.015, 0.05$, and 0.0625) were predicted and compared with the corresponding experimental results.

2 Results

2.1 Experimental results

The results of scanning electron microscopy (SEM), X-ray diffraction (XRD) pattern, absorption spectrum, and Tauc plot of the $\text{Cr}_{0.0625}\text{Sb}_{0.0625}\text{Ti}_{0.875}\text{O}_2$ ceramic pigment are shown in Fig. 1. SEM image (Fig. 1a) shows micron-sized agglomerates with irregular, non-spherical morphology. The aggregates show uneven, tortuous external boundaries without discernible faceted crystal surfaces. The surfaces display a granular, cauliflower-like texture, hinting at additional fine structural details. The inset shows the macroscopic appearance of the pigment powder, which exhibits a yellow-orange color with CIE $L^*a^*b^*$ coordinates of $L^* = 58.14$, $a^* = 13.35$, and $b^* = 46.77$.

We analyzed the XRD pattern (Fig. 1b) using the QualX program [24]. The diffraction peaks are predominantly indexed to the rutile phase of TiO₂ (COD ID: 00-900-4142), with a few low-intensity peaks corresponding to the anatase phase of TiO₂ (COD ID: 00-720-6075). We estimated the crystallite size to be approximately 83 nm from the most intense diffraction peak using the Scherrer equation:

$$D = \frac{0.89\lambda}{\beta \cos \theta}, \quad (1)$$

where D , λ , β , and θ denote the average crystallite size, the X-ray wavelength, the full width at half maximum (FWHM) of the peak, and the Bragg diffraction angle, respectively [17].

The absorption spectrum of Cr_{0.0625}Sb_{0.0625}Ti_{0.875}O₂ (Fig. 1c) shows a broad band in the blue-green region. The absorption band can be attributed to spin-allowed transitions of ${}^4A_2({}^4F) \rightarrow {}^4T_2({}^4F)$ and ${}^4A_2({}^4F) \rightarrow {}^4T_1({}^4F)$ in an octahedral coordination of Cr³⁺ cation [12, 15]. Fig. 1d shows the corresponding Tauc plot, from which the direct E_g of the pigment is estimated to be approximately 2.2 eV.

2.2 DFT and DFT+ U results

The electronic band structure, density of states (DOS), and projected density of states (PDOS) of rutile TiO₂ and (Cr, Sb)-doped rutile TiO₂ for both DFT and DFT+ U methods are shown in Fig. 2. The k -path was selected using the SeeK-path tool [25]. From the results, it can be seen that for rutile TiO₂, the DFT method yields a E_g of 1.4 eV, which increases to 1.9 eV after applying the Hubbard U correction to the Ti-3d orbitals. The PDOS indicates that the valence band maximum (VBM) and conduction band minimum (CBM) are mainly composed of O-2p and Ti-3d states. After applying U , the Ti-3d states in the conduction region shift toward higher energies and become more localized, whereas the O-2p manifold in the valence region remains nearly unchanged. Therefore, the E_g enlargement mainly arises from an upward shift in the Ti-3d-derived conduction band edge, reflecting improved treatment of electron correlation within the d shell. For the Cr/Sb co-doped rutile TiO₂ structure, the DFT approach yields a E_g of zero, which can indicate quasi-metallic character. The PDOS shows impurity peaks near the Fermi level, mainly originating from Cr-3d states, whereas Sb has a minor contribution near the band edge. In this case, the Cr-derived peaks are relatively broad and strongly hybridized with the host bands, leading to an overlap between impurity states and the band edges. When the Hubbard U correction is included, the E_g opens to 1.3 eV, and the system recovers semiconducting character.

The charge density and electron localization function of undoped and doped rutile TiO₂ structures that were calculated with the DFT+ U method are shown in Supplementary Fig. 1. The rutile TiO₂ displays uniform charge distribution and shows no evident asymmetry. Here, the electron density is localized around the atomic centers and extends along the Ti–O bond directions, indicating Ti-3d and O-2p hybridization and thus a mixed ionic-covalent bond character. Upon Cr/Sb co-substitution, the Cr site exhibits charge density extending along Cr–O directions and indicates partial electron sharing between Cr and O atoms, pointing to moderate covalent contributions

through Cr-3d and O-2p hybridization. Around the Sb site, it can be seen that the charge density remains strongly localized near the atomic core, while the interatomic regions along Sb–O directions exhibit reduced density compared with Ti–O bonds. This behavior points to reduced orbital overlap and a more ionic bonding tendency for Sb–O.

2.3 Scenario 1 of the E_g prediction with neural networks

As described in the Methods section, we considered two scenarios to predict E_g using classical and hybrid quantum–classical neural network models. The heat map of the Pearson correlation coefficient matrix is shown in Supplementary Fig. 2. For Scenarios 1 and 2, the features of CBM, Fermi level asymmetry relative to E_g center (asym), and lattice parameters of a and b are most strongly correlated with the target, whereas the VBM shows the weakest correlation.

2.3.1 Effect of variational circuit depth

Schematics of the C-NN diagram and quantum circuits are shown in Supplementary Figs. 3-6. To ensure a fair comparison, the same depth was used across all models; specifically, we employed quantum circuits with a depth of four layers. Supplementary Table 1 summarizes the evaluation metrics of hybrid quantum–classical models for different variational circuit depths. In general, the results show that a circuit depth of four layers provides a reasonable balance between model expressibility and training complexity. Increasing the circuit depth generally leads to a larger number of trainable parameters and entangling operations, which not only raises the computational cost but can also make the optimization process less stable and may also increase the risk of barren plateau effects [26]. For the HQCNN-QI, the best performance is obtained at a circuit depth of two layers. The performance drops at depth three and recovers somewhat at depth four, although it still does not reach the accuracy achieved with two layers. Nevertheless, a depth of four layers was used for all models to enable a fair comparison across architectures.

2.3.2 Performance and interpretability of optimized models

The comparison between the true and predicted E_g for all models, together with their evaluation metrics, is presented in Fig. 3 and Supplementary Table 2. In all cases, the predicted values follow the diagonal trend closely, showing an agreement between the predicted and true E_g values across the dataset. The evaluation metrics and cross-validation results confirm the performance of the models. HQCNN-QI has the lowest value of R^2 (0.9450) and the highest values of MAE (0.0651 eV) and MSE (0.0045 eV). For the other models, $R^2 \geq 0.98$ and the MAE and MSE values are lower. The cross-validation results remain consistent with the corresponding metrics obtained from the primary evaluation.

The SHAP summary and corresponding bar plots of mean absolute SHAP values for all models are presented in Fig. 4. Detailed SHAP dependence analyses are provided in Supplementary Note 2. For the C-NN, the SHAP summary plot indicates that the prediction is largely governed by electronic features. Among them, CBM shows the

largest contribution to the model output. The Fermi energy (efermi) also appears with a noticeable influence, followed by VBM. The ranking based on the mean absolute SHAP values confirms this pattern. CBM contributes the most to the predictions, while efermi and VBM provide secondary effects. Most remaining features have SHAP values near zero, suggesting only minor contributions in the model's predictions. For the HQCNN-SE, HQCNN-HEA, and HQCNN-QI models, CBM and VBM dominate the prediction, while the efermi appears only as a weak feature. Other features remain of near-zero importance. The ranking based on the mean absolute SHAP values confirms this situation. This behavior is consistent with the physical origin of the E_g , which is defined by $E_g = \text{CBM} - \text{VBM}$. In general, the SHAP analysis indicates that while all models capture the general electronic trends associated with E_g formation, the hybrid quantum-classical models produce a more compact and physically consistent representation of the prediction mechanism.

2.4 Scenario 2 of the E_g prediction with neural networks

In Scenario 2, we removed the VBM, CBM, and efermi features to examine the robustness of the models when explicit band-edge information is not available. Instead, we introduced the asym as an alternative feature. This quantity is defined as

$$\text{asym} = \frac{\text{VBM} + \text{CBM}}{2} - E_F, \quad (2)$$

which represents the position of the Fermi level relative to the center of the E_g . When the Fermi level lies close to the middle of the gap, the asym value approaches zero, whereas positive and negative values indicate a shift of the Fermi level toward the valence band (p-type tendency) or toward the conduction band (n-type tendency), respectively. By using this combined quantity rather than the individual CBM and VBM values, the model does not receive the band-edge positions as separate features, while still retaining information about the electronic asymmetry within the E_g .

To allow a direct comparison with Scenario 1, the model configurations were kept unchanged. In particular, the hybrid quantum-classical models employed variational circuits with a depth of four layers, identical to those used in the previous scenario. Illustrations of the C-NN architecture and the quantum circuits are provided in Supplementary Figs. 7-10.

2.4.1 Models performance and interpretability

As shown in Fig. 5, the predicted E_g values follow the general diagonal trend, although the points show more scatter than in Scenario 1. Despite the larger scatter, the main upward trend remains visible across all models, and the relationship between the true and predicted E_g values is still clear. The results in Supplementary Table 3 confirm this behavior. Although the MAE, MSE, and R^2 metrics show lower accuracy than in Scenario 1, the models continue to provide stable and physically meaningful estimates. The models show similar behavior across both scenarios in terms of their strongest performer, with HQCNN-HEA again achieving the best results. However, the weakest model shifts from HQCNN-QI in Scenario 1 to the C-NN in Scenario 2. Taken together,

these results show that even with fewer features in Scenario 2, the models still follow the main E_g trend.

The SHAP analysis for Scenario 2, presented in Fig. 6 shows a clear and consistent pattern across all models. The *asym* feature clearly has the strongest impact on prediction compared to all other features. This suggests that after removing VBM, CBM, and *efermi*, the models depend chiefly on the electronic asymmetry represented by *asym*. The structural parameters such as *a* and *b* still contribute, but their influence is fairly limited. Their SHAP values are relatively small, suggesting that these features act as secondary adjustments rather than primary drivers of the output. All models display the same overall ranking of feature importance. The *asym* consistently carries the largest contribution, while only a limited set of structural features shows measurable but minor effects. Although the exact importance values vary among architectures, the overall pattern remains the same. Further SHAP dependence results are provided in Supplementary Note 2. In summary, the SHAP analysis shows that *asym* effectively replaces the missing electronic features (VBM, CBM, and *efermi*) and becomes the central feature for predicting E_g in Scenario 2, while structural parameters offer only minor refinements.

2.5 Prediction of the E_g of rutile TiO_2

In this section, we are interested to comparing the E_g values obtained from DFT, DFT+*U*, and neural network models for the rutile TiO_2 structure. These results are presented in Table 1. The DFT calculation significantly underestimates this value, yielding a E_g of 1.4 eV. Applying the *U* correction raises the E_g to 1.9 eV. This value is close to the E_g reported for the entry mp-2657 in the Materials Project database, which is 1.781 eV. The predictions obtained from the neural network models show that Scenario 1 generally performs better than Scenario 2. Within Scenario 1, the HQCNN-HEA model gives the most accurate estimate, with an error of 0.001 eV. For Scenario 2, the HQCNN-SE model provides the closest prediction, showing an error of 0.0497 eV. The largest error is related to the C-NN model in Scenario 2, with a deviation of 0.1485 eV. Taken together, these results show that both scenarios are capable of producing acceptable predictions for the E_g of rutile TiO_2 , although Scenario 1 generally leads to more accurate outcomes.

2.6 Color prediction results

Supplementary Fig. 11 shows the heat map of the Pearson correlation coefficient (*r*) matrix for the features and the color coordinates *x* and *y*. The results indicate that *x* has the strongest correlation with *y* ($r = 0.63$), while *y* similarly shows its highest correlation with *x* ($r = 0.63$). Among the remaining features, the synthesis temperature (*T*) exhibits the next highest correlation with the targets ($r = 0.25$ for *x* and $r = 0.21$ for *y*). In addition, the Cr shows a noticeable positive correlation with *x* ($r = 0.23$). In contrast, Sb shows the weakest correlation with *x* ($r = -0.14$), while Nb exhibits the lowest correlation with *y* ($r = -0.022$), suggesting that these features have a minimal linear relationship with the color coordinates.

2.6.1 Impact of variational circuit depth

Supplementary Table 4 reports the performance of the hybrid quantum–classical models for predicting the x -component of the color coordinate across different variational circuit depths. The results show that increasing the circuit depth generally enhances model accuracy, although the extent of improvement varies between architectures. For the HQCNN-SE model, performance improves consistently as the depth increases, with the lowest error values observed at four layers. The HQCNN-HEA model shows a less uniform trend, with its accuracy dropping at a depth of two layers and then recovering at deeper circuits, eventually reaching its best performance at a depth of four. In contrast, the HQCNN-QI model performs best with a two-layer circuit, while deeper circuits do not lead to further improvements. Despite these variations, a depth of four layers was ultimately selected for all architectures to maintain consistency in model comparison. Although this depth is not individually optimal for every model, it provides a common basis for evaluating their relative capabilities. For the same reason, the y -coordinate prediction task was also conducted using a four-layer circuit depth to ensure comparability between the x and y components of the color space. Schematics of the C-NN architecture and the quantum circuits are presented in Supplementary Figs. 12-15.

2.6.2 Insights into the performance and interpretability of the models

The comparison between the true and predicted colorimetric coordinates (x and y) for all models, along with their evaluation metrics, is presented in Fig. 7 and Supplementary Table 5. The results show that the models predict the y coordinate more accurately than the target x coordinate. In all models, the predicted y points fall closer to the diagonal trend line, while the predicted x points show a larger spread around this line. The evaluation metrics and cross-validation results support this observation. MAE and MSE are lower for the y predictions, and the R^2 values are higher compared to those of x . The C-NN model yields the highest R^2 value for predicting the x target ($R^2 = 0.7799$), whereas the HQCNN-SE model achieves the best performance for the y target ($R^2 = 0.8731$). In contrast, HQCNN-QI exhibits the weakest performance for both coordinates with $R^2 = 0.7009$ and $R^2 = 0.7788$ for x and y , respectively.

The SHAP summary distributions together with the mean absolute SHAP values (Fig. 8 and Supplementary Fig. 16) consistently indicate a strong statistical coupling between the two chromaticity coordinates. In every model, the complementary coordinate (y for predicting x and x for predicting y) exhibits the largest contribution to the model output, underscoring the tight interdependence inherent to the chromaticity space. For the C-NN model, the three most influential features for predicting x are y , Cr, and Nb, while Sb, T , Ti, and Ni appear as secondary factors. In the prediction of y , the three largest contributions come from x , Ti, and Nb, followed by Cr, T , Sb, and Ni with decreasing influence. In the HQCNN-SE model, the most important features for x are y , Cr, and T , whereas Ti, Sb, Nb, and Ni show lower contributions. For y , the top three are x , Ti, and Cr, and the remaining features play lesser roles. Within the HQCNN-HEA model, the leading features for x are y , Cr, and T . Notably,

in this model, Ni exhibits a stronger influence than Ti, distinguishing it from the other cases. Ti, Sb, and Nb contribute more modestly. When predicting y , the most relevant variables are x , Cr, and Ti, followed by T , Nb, Sb, and Ni. Furthermore, in the HQCNN-QI model, the three dominant factors for x include y , Cr, and T , with Ti, Nb, Sb, and Ni contributing to lesser degrees. For y , the top-ranked variables are x , Cr, and Ti, while T , Sb, Ni, and Nb appear as lower-priority variables.

2.6.3 Color prediction of $\text{Cr}_x\text{Sb}_x\text{Ti}_{1-2x}\text{O}_2$ pigments

Here, the predictive performance of all neural network models in estimating the chromaticity coordinates of the pigment series $\text{Cr}_x\text{Sb}_x\text{Ti}_{1-2x}\text{O}_2$ is evaluated. Four representative compositions, $x = 0$ [27], $x = 0.015$ [28], $x = 0.05$ [28], and $x = 0.0625$, were selected. Fig. 9 compares the experimental chromaticity coordinates of these pigments with the predictions obtained from the neural network models. For the undoped sample ($x = 0$), the HQCNN-SE and HQCNN-HEA models show the closest agreement with the experimental point, while the C-NN model gives the largest deviation among the models. For $x = 0.015$, HQCNN-SE provides the most accurate prediction, closely matching the experimental coordinate. In this case, HQCNN-HEA shows the weakest performance and produces the point farthest from the measurement. Also, a similar trend can be observed for the $x = 0.05$ sample. For the highest dopant level investigated ($x = 0.0625$), HQCNN-HEA yields the best agreement with experiment. In contrast, C-NN and HQCNN-QI exhibit the lowest predictive accuracy for this composition. Overall, the results show that, for each pigment composition examined, the most accurate predictions are obtained from the hybrid quantum-classical models. HQCNN-SE provides the closest match to the experimental chromaticity coordinates at $x = 0$, $x = 0.015$, and $x = 0.05$, whereas HQCNN-HEA gives the best agreement with the measurements at $x = 0$ and $x = 0.0625$. Although the model with the lowest accuracy differs among compositions, and certain hybrid architectures such as HQCNN-HEA do not surpass the C-NN in some cases, the hybrid framework consistently includes the most reliable predictors across the full set of samples.

3 Discussion

The aim of this study was to examine whether classical and hybrid quantum-classical neural network models could be used to predict two important properties of ceramic pigments, namely the E_g and color. For this purpose, $\text{Cr}_x\text{Sb}_x\text{Ti}_{1-2x}\text{O}_2$ rutile pigments were selected as a model system and investigated through a combination of experimental, DFT/DFT+ U , and classical/quantum-classical neural networks approaches. The $\text{Cr}_{0.0625}\text{Sb}_{0.0625}\text{Ti}_{0.875}\text{O}_2$ pigment was synthesized and characterized, while the electronic structures of undoped and doped rutile TiO_2 were analyzed using DFT and DFT+ U . In parallel, classical and hybrid quantum-classical neural network models were trained to predict the E_g of TiO_2 and chromaticity coordinates of rutile-based pigments using datasets derived from the Materials Project database and experimental pigment data reported in the literature.

The experimental results showed that the synthesized $\text{Cr}_{0.0625}\text{Sb}_{0.0625}\text{Ti}_{0.875}\text{O}_2$ pigment exhibited micron-sized agglomerates (irregular and non-spherical morphology) and a yellow–orange color with CIE $L^*a^*b^*$ coordinates of $L^* = 58.14$, $a^* = 13.35$, and $b^* = 46.77$. XRD analysis confirmed that the main crystalline phase corresponded to rutile TiO_2 , with only a minor contribution from anatase. The absorption spectrum revealed a broad band in the blue–green region, which was consistent with electronic transitions associated with Cr^{3+} ions in octahedral coordination. The Tauc analysis indicated a direct optical E_g of approximately 2.2 eV.

The DFT calculations provided additional insight into the electronic structure of the system. For undoped rutile TiO_2 , the calculated E_g was 1.4 eV using standard DFT and increased to about 1.9 eV when the Hubbard U correction was applied to the Ti-3d orbitals. The PDOS indicated that the valence band and conduction band edges were primarily composed of O-2p and Ti-3d states. In the Cr/Sb co-doped structure, standard DFT predicted a vanishing E_g . When the U correction was included, these states shifted, and a E_g of approximately 1.3 eV was recovered, indicating semi-conducting behavior. Charge density and ELF results showed that the Ti–O bonds exhibited mixed ionic–covalent character, whereas Cr–O bonds show enhanced covalent contributions via Cr-3d/O-2p hybridization, in contrast to the more ionic Sb–O bonding with weaker orbital overlap.

The classical and hybrid quantum–classical neural network models were trained to predict the E_g using two different scenarios. In Scenario 1, all models produced highly accurate predictions, and the predicted values closely matched the reference E_g values across the dataset. The SHAP analysis showed that CBM and VBM dominated the predictions in all models, which was consistent with the physical definition of the E_g . In Scenario 2, the prediction accuracy decreased, but the models still reproduced the general trend of the E_g values. In this case, the asym emerged as the most influential feature, suggesting that it contained relevant information about the electronic structure even when the band-edge energies were not explicitly included. The results of the E_g prediction for rutile TiO_2 showed that the conventional DFT approach underestimated the E_g . The inclusion of the Hubbard U correction increased the calculated E_g and improved the electronic structure description. The predictions obtained from the neural network models indicated that Scenario 1 generally performed better than Scenario 2. Within Scenario 1, the HQCNN-HEA model showed the most accurate estimate. In Scenario 2, the HQCNN-SE model showed the closest prediction.

The color prediction results showed that all models predicted the y chromaticity coordinate more accurately than x . The C-NN performed best for predicting x , while HQCNN-SE achieved the highest accuracy for y . SHAP analysis revealed a strong intrinsic coupling between the two chromaticity coordinates. When applied to the $\text{Cr}_x\text{Sb}_x\text{Ti}_{1-2x}\text{O}_2$ pigments, the hybrid quantum–classical models consistently produced predictions closer to experiment than the C-NN. HQCNN-SE and HQCNN-HEA provided the most accurate results across different compositions, highlighting the improved reliability offered by the hybrid architectures in modeling colorimetric behavior.

Some limitations, nevertheless, needed to be considered. For example, one limitation arose from the availability of structural data for doped materials. Databases

such as the Materials Project mainly contained undoped crystal structures, and doped configurations were rarely reported. Another limitation was the absence of a comprehensive experimental database for ceramic pigments that simultaneously reported composition, synthesis conditions, and quantitative color coordinates. Because of this limitation, the dataset used for color prediction contained a relatively limited number of samples.

Overall, the results of this work showed that classical and hybrid quantum-classical neural networks could be used to predict the E_g and color of rutile-based ceramic pigments with reasonable accuracy. The hybrid models provided slightly improved predictive performance and clearer feature attribution compared with the classical architecture. The framework adopted in this study, which combined experiment, first-principles calculations, and classical/quantum-classical neural networks, therefore offered a useful strategy for studying and predicting the E_g and color of crystalline materials and ceramic pigments. In future work, this approach could be extended to fully quantum machine learning models implemented on quantum hardware, allowing the predictive framework to move beyond hybrid architectures and further explore the potential of quantum computation for materials property prediction.

4 Methods

4.1 Classical and hybrid quantum-classical neural network models

4.1.1 Datasets and learning protocol

In this work, we have E_g and colorimetric coordinates as targets. For E_g , we generated a dataset of 1527 materials (see Supplementary Data 1) with a tetragonal crystal system and energy gap between 1 and 2 eV using the Python Materials Genomics (pymatgen) library [29] from the Materials Project [30]. We used two scenarios to predict the E_g . The list of features and their symbol descriptions is presented in Supplementary Table 6. For the color prediction, we constructed a dataset comprising 195 rutile-based ceramic pigments collected from the literature [14, 15, 27, 28, 31–45] and the present study (see Supplementary Data 2). We converted the color space from CIE $L^*a^*b^*$ to CIE xyY , where x and y were used as the target variables. The input features consisted of the atomic stoichiometries (i.e., the subscripts in the chemical formula) and the synthesis temperature.

We developed a set of learning models to predict the E_g and color, including hybrid quantum-classical neural networks and a fully classical quantum-equivalent baseline (C-NN). We used the same input features, training protocols, and evaluation procedures for all models to ensure a controlled comparison of their architectures. We normalized the features and targets to $[0, \pi]$ and $[-1, 1]$ using min-max scaling, respectively. The data was split into 80% training and 20% testing sets. All classical layers and training routines were implemented in PyTorch, while hybrid quantum layers were constructed using PennyLane [46] with TorchLayer. All random seeds were fixed to guarantee reproducibility. Hyperparameters were optimized using the Tree-Structured Parzen Estimator (TPE) in Optuna [47]. We trained the models using the

AdamW optimizer [48]. Given an input feature vector $\mathbf{x} \in \mathbb{R}^d$, the models learn a nonlinear mapping

$$\hat{y} = f_{\Theta}(\mathbf{x}). \quad (3)$$

Herein, $\hat{y}$ is the predicted (scaled) target and Θ represents all trainable parameters (classical weights and quantum circuit parameters) [49, 50]. All models were trained to minimize the mean squared error (MSE) as a standard loss function in regression problems.

4.1.2 Input embedding layer

Each model begins with a classical linear transformation

$$\mathbf{h}_1 = W^{(1)}\mathbf{x} + \mathbf{b}^{(1)}, \quad (4)$$

followed by a LeakyReLU activation [51]

$$\mathbf{h}_1 = \max(\alpha \mathbf{h}_1, \mathbf{h}_1), \quad (5)$$

with slope parameter $\alpha = 0.1$. This layer maps the raw features into a latent space that matches the dimension required for quantum embedding.

4.1.3 Quantum-equivalent classical baseline

To isolate the structural contribution of the quantum circuit, we implemented a fully classical surrogate designed to match the depth and feature dimensionality of the quantum layer. Let

$$\mathbf{h}_1 = \text{LeakyReLU}\left(W^{(1)}\mathbf{x} + \mathbf{b}^{(1)}\right) \quad (6)$$

denote the embedded representation produced by the input layer, where $\mathbf{h}_1 \in \mathbb{R}^d$ and d equals the number of qubits in the quantum model. The classical quantum-equivalent block then applies L successive fully connected nonlinear transformations while preserving this dimensionality. Defining $\mathbf{z}_0 = \mathbf{h}_1$, the block is given by

$$\mathbf{z}_{k+1} = \tanh\left(W^{(k)}\mathbf{z}_k + \mathbf{b}^{(k)}\right), \quad k = 0, \dots, L-1, \quad (7)$$

where each $W^{(k)} \in \mathbb{R}^{d \times d}$ and $\mathbf{b}^{(k)} \in \mathbb{R}^d$. The final output of the block is $\mathbf{z}_L$. This construction preserves network depth, parameterized nonlinear transformations, and dense feature mixing comparable to the variational quantum circuit, while deliberately omitting quantum entangling operations.

4.1.4 Variational quantum circuits (VQCs)

We designed three hybrid quantum–classical neural networks, each using a different variational quantum circuit ansatz: a strongly entangling architecture (HQCNN-SE), an enhanced hardware-efficient ansatz (HQCNN-HEA), and a QAOA-inspired ansatz (HQCNN-QI). Quantum layers were implemented as parameterized unitary operators

$U(\mathbf{h}_1, \boldsymbol{\theta})$ acting on $n = d$ qubits, where the number of qubits was chosen to match the number of input features, with each feature encoded into a distinct qubit. Expectation values of Pauli-Z operators provide classical outputs:

$$z_i = \langle 0|U^\dagger(\mathbf{h}_1, \boldsymbol{\theta}) Z_i U(\mathbf{h}_1, \boldsymbol{\theta})|0\rangle, \quad (8)$$

following standard VQC measurement protocols. The measurement vector $\mathbf{z} \in [-1, 1]^n$ was passed to classical dense layers.

Hybrid quantum-classical models were simulated with a noiseless statevector backend. For the HQCNN-SE, classical data were embedded into the quantum circuit via single-qubit rotations along the X -axis, implementing an angle-embedding scheme as discussed in Schuld *et al.* [52]. We implemented this encoding as

$$U_{\text{enc}}(\mathbf{x}) = \bigotimes_{i=1}^n R_X(x_i), \quad (9)$$

where each classical feature x_i is mapped to a rotation on the i -th qubit. This procedure encodes each data point into a rotation-based quantum state, offering a direct implementation of angle embedding. For the HQCNN-QI and HQCNN-HEA, a trainable affine encoding was used,

$$U_{\text{enc}}(\mathbf{x}, \boldsymbol{\alpha}, \boldsymbol{\beta}) = \bigotimes_{i=1}^n R_Y(\alpha_i x_i + \beta_i), \quad (10)$$

which also supports data re-uploading [53].

The HQCNN-SE circuit consists of L layers of parametrized single-qubit rotations interleaved with entangling operations arranged in a ring topology. The HQCNN-HEA circuit consists of repeated layers of single-qubit R_X , R_Y , and R_Z rotations on each qubit, followed by full all-to-all CNOT entanglement. This design has become a standard architecture, widely adopted in near-term variational quantum algorithms [54]. The HQCNN-QI circuit employs the same all-to-all entangling block followed by axis-resolved single-qubit rotations, forming a hardware-efficient variant of the QAOA mixing dynamics [55]. The circuits are described in detail in Supplementary Note 1.

4.1.5 Classical post-processing network

The quantum outputs were processed through classical dense layers as

$$\mathbf{h}_{L+1} = \text{LeakyReLU}\left(W^{(L+1)}\mathbf{h}_L + \mathbf{b}^{(L+1)}\right), \quad (11)$$

$$\hat{y} = W^{(L+2)}\mathbf{h}_{L+1} + \mathbf{b}^{(L+2)}. \quad (12)$$

4.1.6 Evaluation metrics

We assessed the models performance using mean absolute error (MAE), MSE, and coefficient of determination (R^2). A nested 5×3 cross-validation scheme, comprising a

five-fold outer loop for unbiased performance estimation and a three-fold inner loop for hyperparameter optimization, was employed only for two representative models. Due to the high computational cost of training hybrid quantum-classical neural networks, this validation was restricted to C-NN and HQCNN-SE.

4.2 DFT and DFT+ U calculations

We modeled the undoped rutile TiO_2 along with the doping of one Cr atom and one Sb atom into rutile. We constructed the $2 \times 2 \times 2$ supercells to match the doping level with the experimental sample. The crystal structure of the samples, visualized using the VESTA program [56], is shown in Supplementary Fig. 17. We also used this program to plot the charge density and electron localization function. We carried out the DFT and DFT+ U calculations using the Quantum ESPRESSO package [57, 58]. Structural optimizations were done using the variable-cell relaxation scheme with BFGS dynamics, allowing both the lattice parameters and atomic coordinates to fully relax. We set the total energy and force convergence thresholds for structural optimization to 10^{-5} Ry and 10^{-4} Ry/Bohr, respectively. Non-spin-polarized calculations were adopted, as the study does not focus on magnetic properties, and the low concentration of dopants in the supercell makes magnetic interactions negligible. The effective Hubbard U parameters of 4.8 and 6.7 eV, obtained using density functional perturbation theory (DFPT) [59], were applied to the Ti 3d and Cr 3d orbitals, respectively. The GGA-PBE exchange-correlation functional with PAW pseudopotentials was used. The calculations were done using a $2 \times 2 \times 4$ k -point mesh. The kinetic energy cutoff for the wave functions and the charge density were set to 90 and 900 Ry, respectively.

4.3 Pigment synthesis and characterization

The $\text{Cr}_{0.0625}\text{Sb}_{0.0625}\text{Ti}_{0.875}\text{O}_2$ pigment was synthesized using the conventional ceramic route. All synthetic precursors were provided from industry with purities of 98% or higher. First, TiO_2 (0.875 mol), Cr_2O_3 (0.0625 mol), Sb_2O_3 (0.0625 mol), and distilled water (as a solvent) were mixed by magnetic stirrer for 30 min. The mixture was then transferred to an oven and dried at 120 °C. Finally, the sample was gradually heated in a furnace to 1000 °C over a period of 2 h, and subsequently held at this temperature for an additional 2 h to ensure complete processing.

The microstructure of the pigment was examined using a VEGA3 TESCAN microscope. XRD analysis was done with an X’Pert Pro instrument (40 kV, 30 mA) using Cu $K\alpha$ radiation ($\lambda = 1.54$ Å) under ambient conditions. UV-Vis spectroscopy and colorimetric measurements were performed using an X-Rite i1Publish Pro 3 device.

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Contributions

S.Y.V.: Conceptualization, Methodology, Neural network model development, DFT calculations, Writing – Original Draft, Writing – Review & Editing; M.K.A.: Conceptualization, Supervision, Writing – Review & Editing; R.D.B.: Materials synthesis and characterization, Writing – Review & Editing.

Competing interests

All authors declare no financial or non-financial competing interests.

Data availability

All data generated or analyzed during this study are included in this published article and its supplementary information file.

Code availability

The Quantum ESPRESSO package (<https://www.quantum-espresso.org>) is an open-source software suite. The codes used to develop and train the neural network models underlying the results of this study are not publicly available; however, they may be provided by the corresponding author upon reasonable request.

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Figures

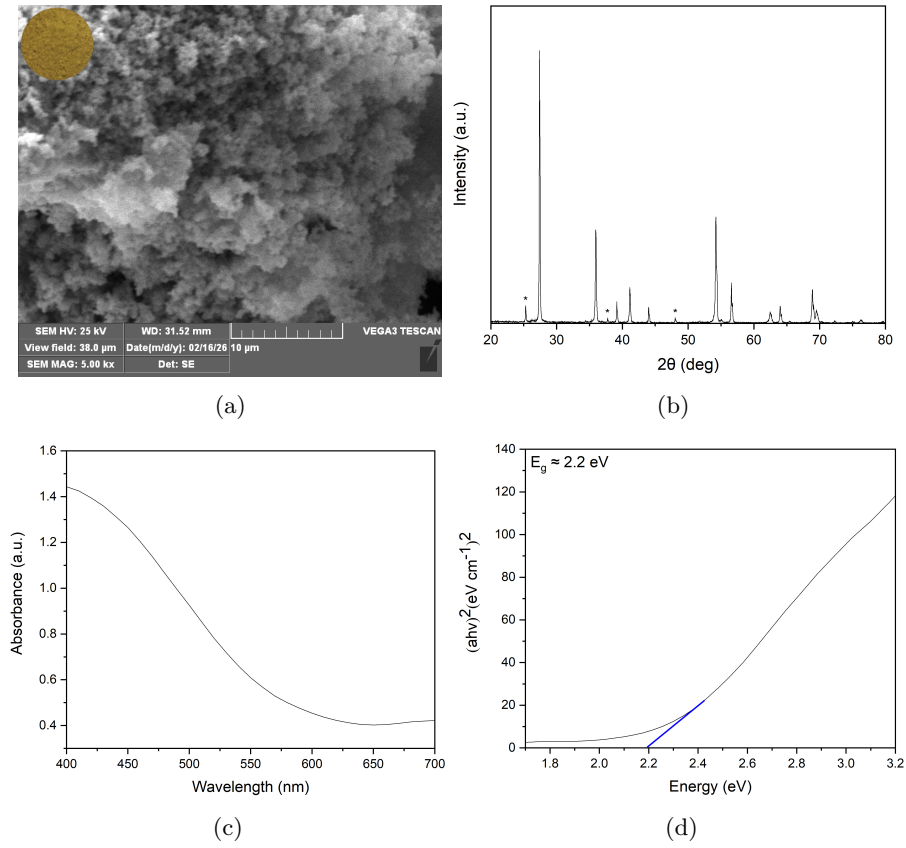


Fig. 1: SEM image (a), XRD pattern (b), absorption spectrum (c), and Tauc plot (d) of the $\text{Cr}_{0.0625}\text{Sb}_{0.0625}\text{Ti}_{0.875}\text{O}_2$ ceramic pigment. Diffraction peaks corresponding to the rutile phase are not labeled in the XRD pattern, whereas peaks marked with an asterisk (*) correspond to the anatase phase.

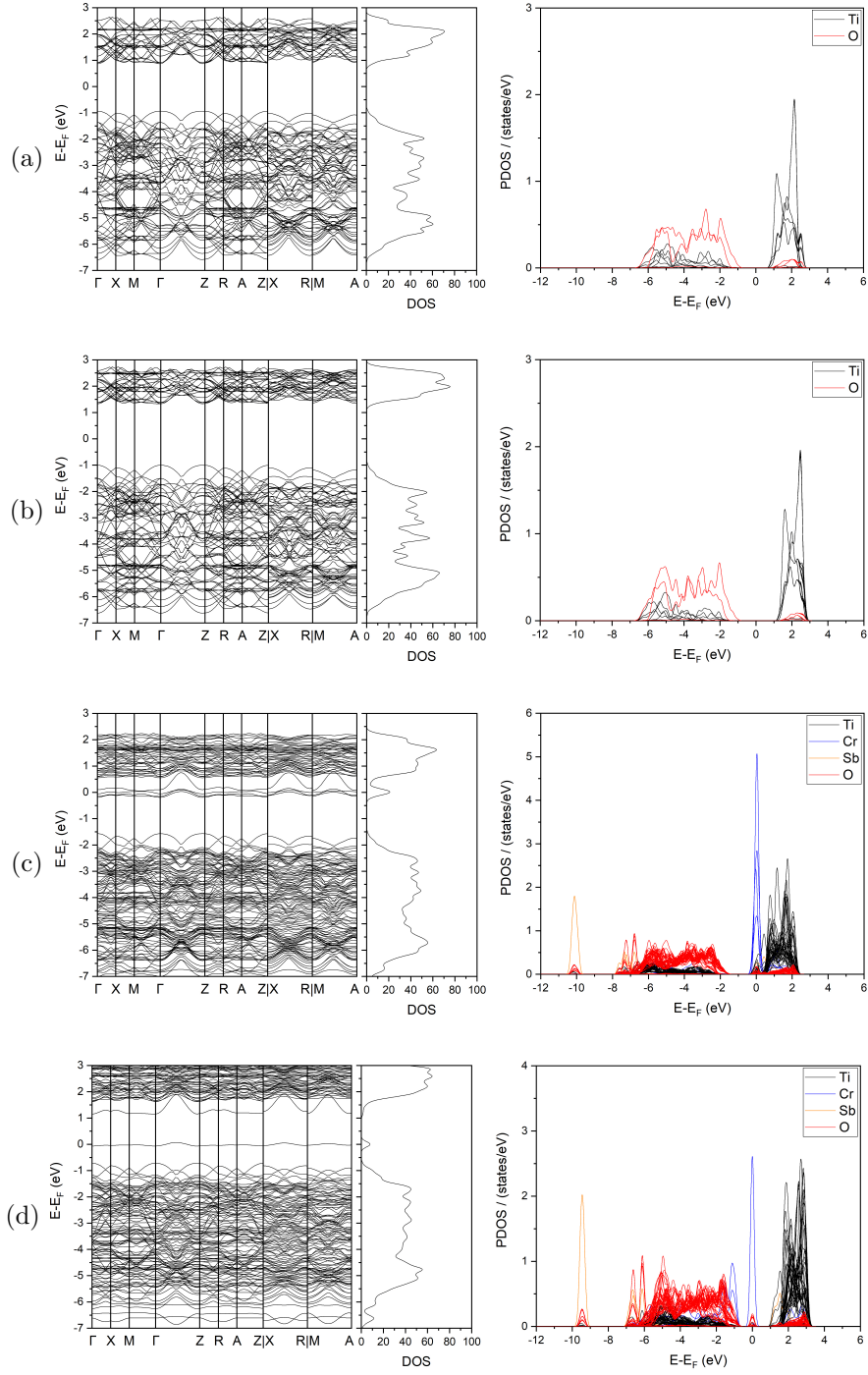


Fig. 2: Band structure, DOS, and PDOS of rutile TiO_2 : (a) DFT method, (b) DFT+ U method, and rutile TiO_2 co-doped with one Cr atom and one Sb atom: (c) DFT method, (d) DFT+ U method.

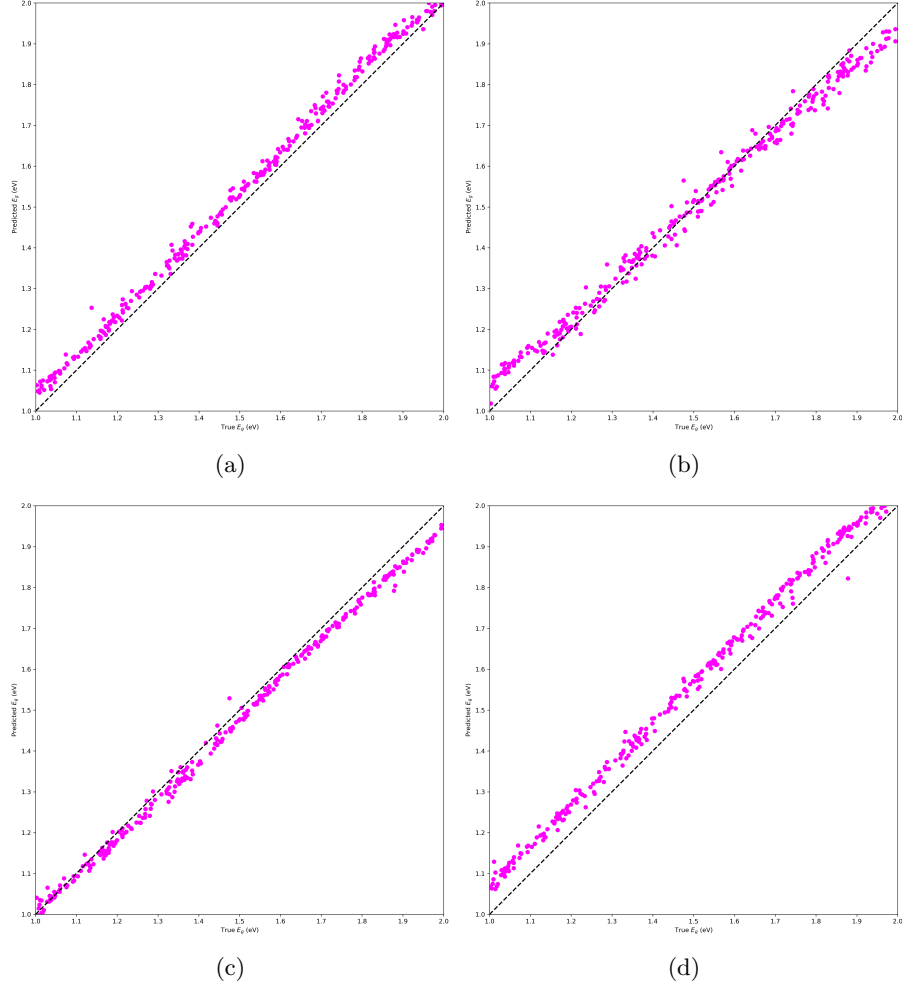


Fig. 3: Comparison between the true and predicted E_g (Scenario 1) for C-NN (a), HQCNN-SE (b), HQCNN-HEA (c), and HQCNN-QI (d) models.

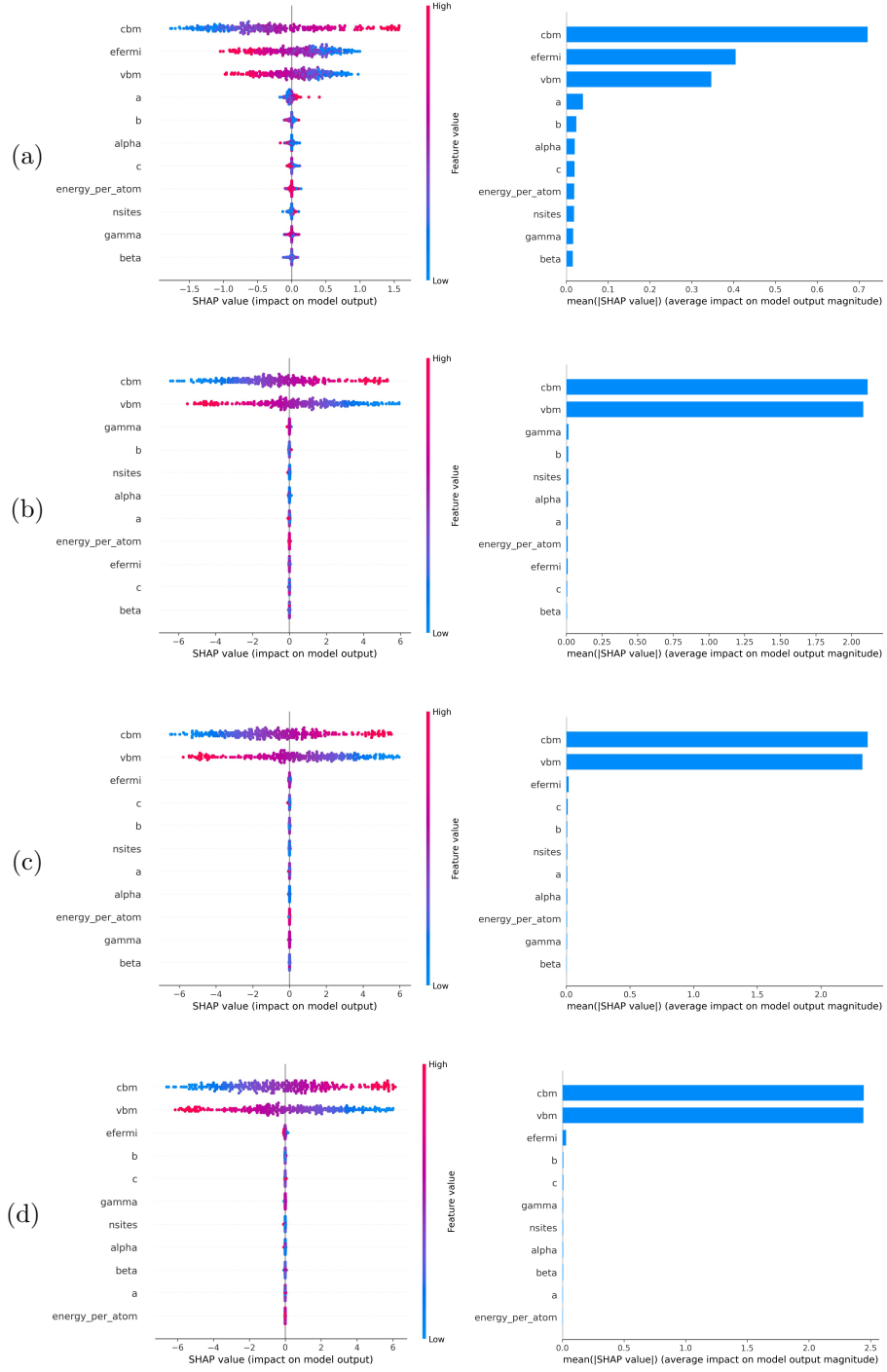


Fig. 4: SHAP summary plots (left) and mean absolute SHAP value bar plots (right) for the C-NN (a), HQCNN-SE (b), HQCNN-HEA (c), and HQCNN-QI (d) models for E_g prediction in Scenario 1.

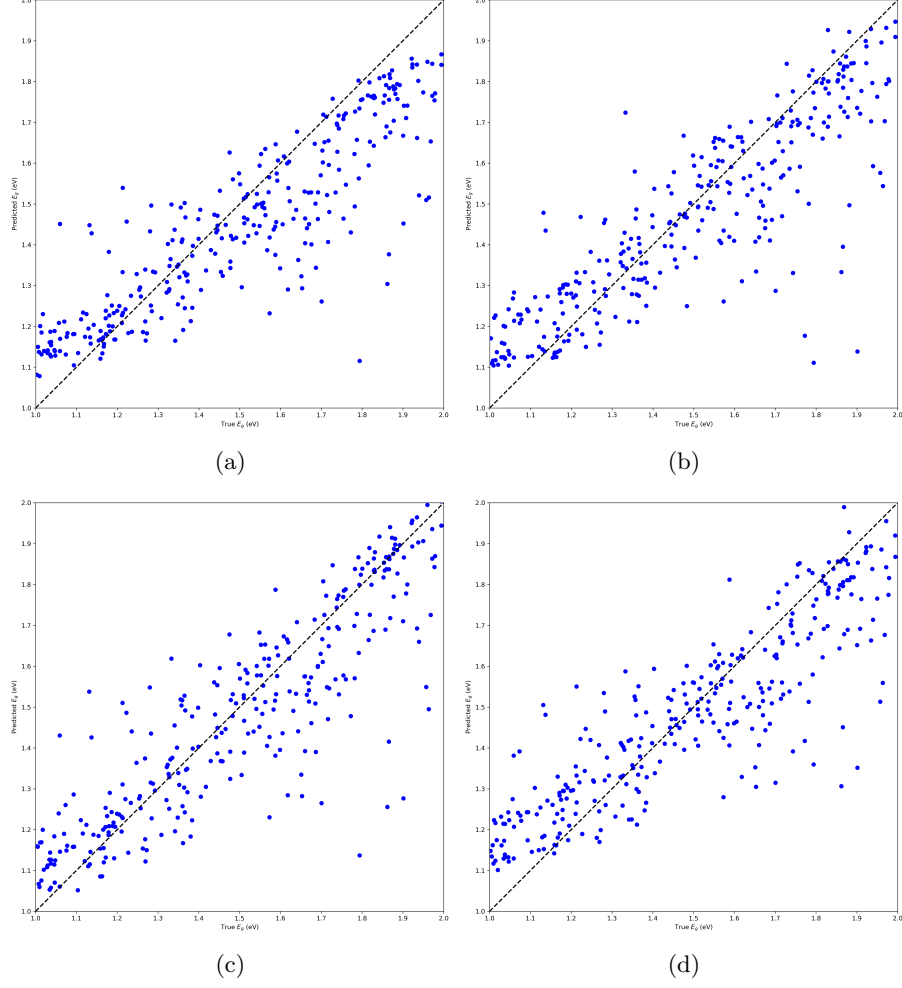


Fig. 5: Comparison between the true and predicted E_g (Scenario 2) for C-NN (a), HQCNN-SE (b), HQCNN-HEA (c), and HQCNN-QI (d) models.

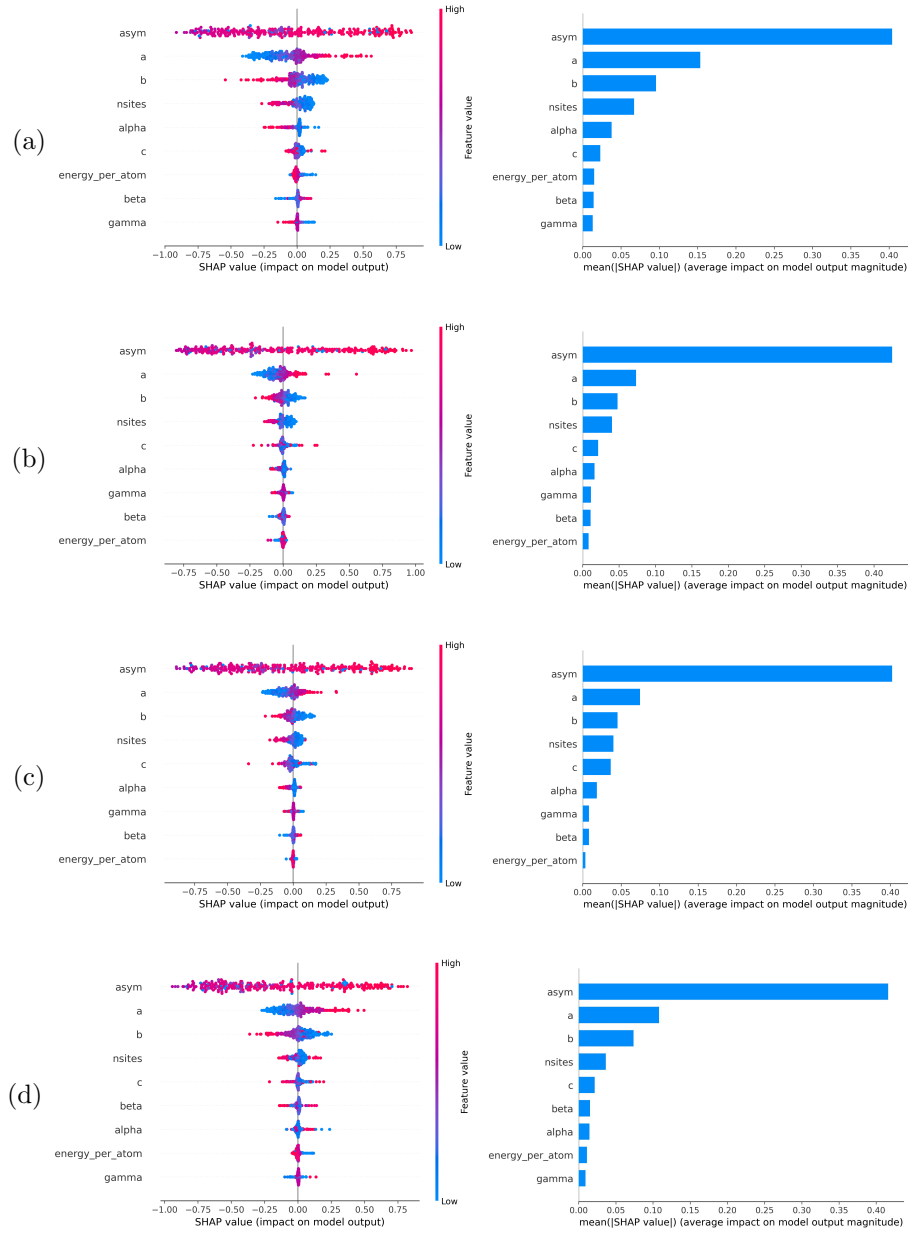


Fig. 6: SHAP summary plots (left) and mean absolute SHAP value bar plots (right) for the C-NN (a), HQCNN-SE (b), HQCNN-HEA (c), and HQCNN-QI (d) models for E_g prediction in Scenario 2.

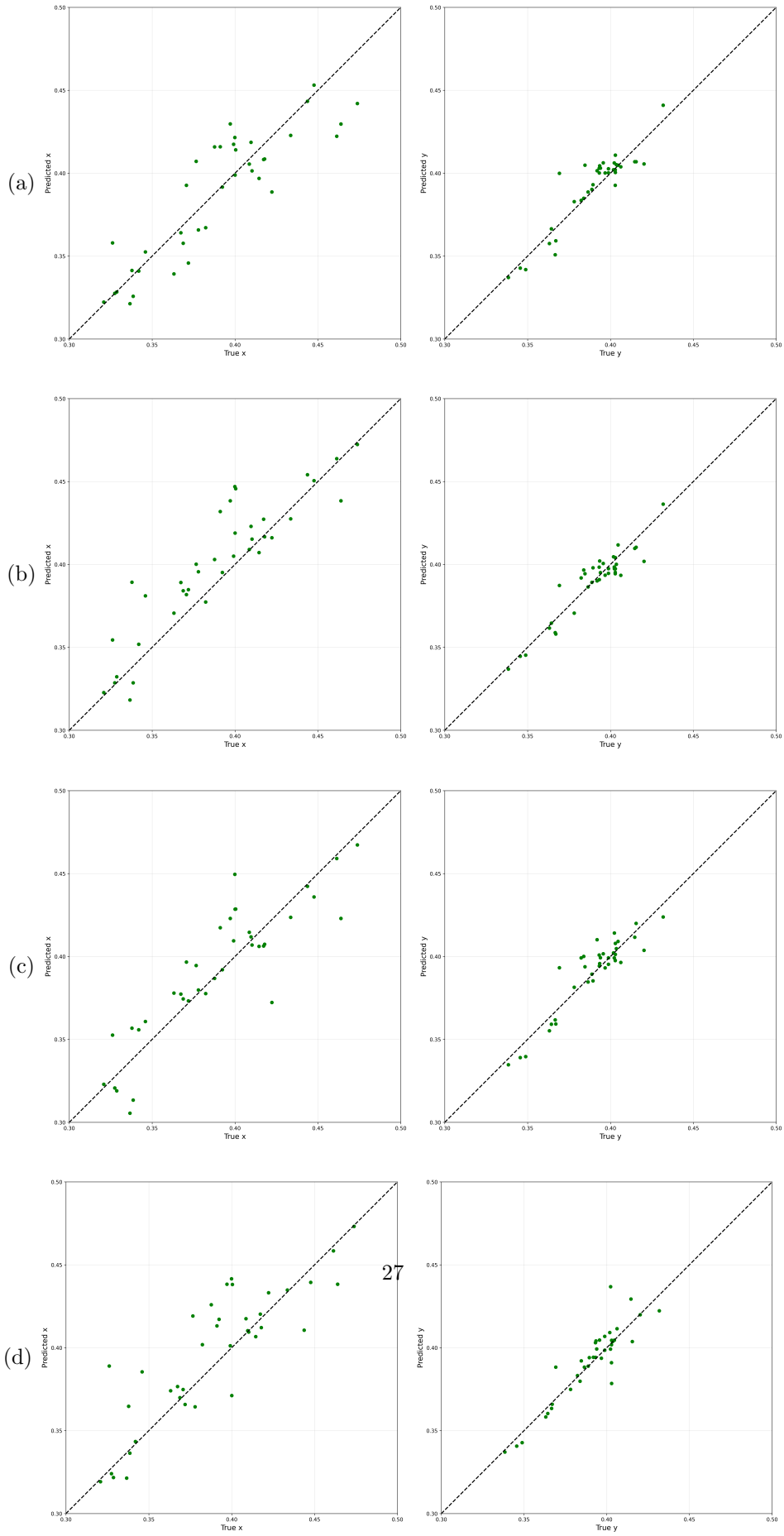


Fig. 7: Comparison between the true and predicted colorimetric coordinates (x and y) for C-NN (a), HQCNN-SE (b), HQCNN-HEA (c), and HQCNN-QI (d) models.

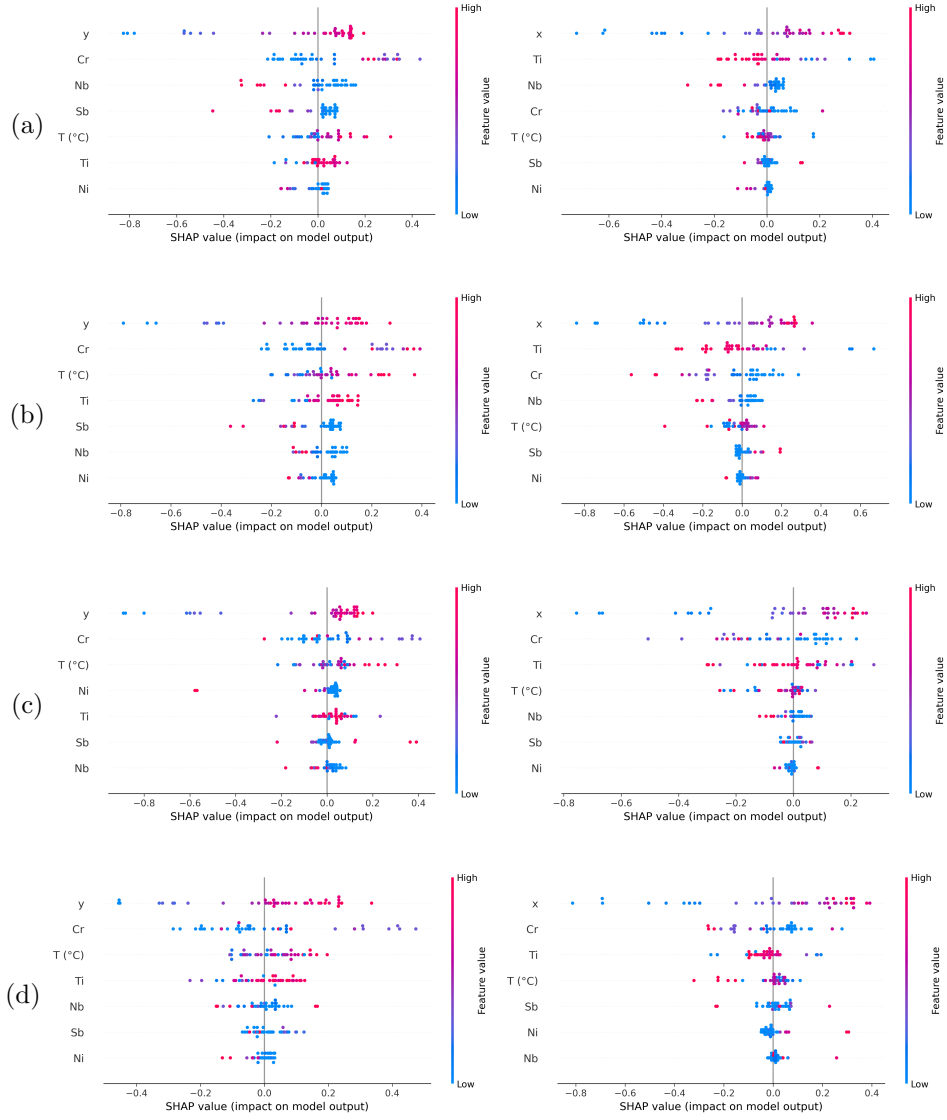


Fig. 8: SHAP summary plots of the C-NN (a), HQCNN-SE (b), HQCNN-HEA (c), and HQCNN-QI (d) models for the prediction of the x (left panels) and y (right panels) colorimetric coordinates.

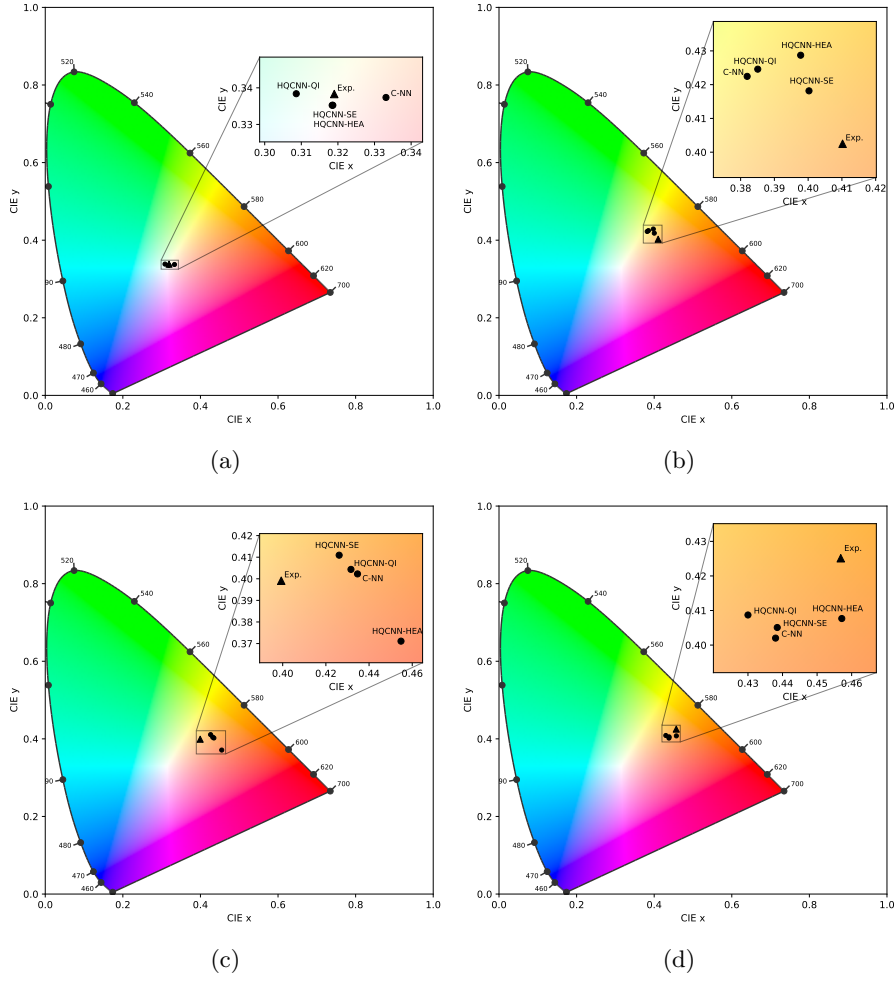


Fig. 9: CIE 1931 chromaticity diagrams of $\text{Cr}_x\text{Sb}_x\text{Ti}_{1-2x}\text{O}_2$ pigments, showing a comparison between experimental chromaticity coordinates and those predicted by neural network models for $x = 0$ [27] (a), $x = 0.015$ [28] (b), $x = 0.05$ [28] (c), and $x = 0.0625$ (d).

Tables

Table 1: Comparison of E_g values for rutile TiO_2 obtained from DFT, DFT+ U , and neural network models. Errors denote the absolute deviation from the mp-2657 reference value.

Method / Model	E_g (eV)	$E_g^{\text{Reference}}$ (eV)	Error (eV)
DFT	1.4	–	–
DFT+ U	1.9		
C-NN (Scenario 1)	1.7676	1.781	0.0134
C-NN (Scenario 2)	1.6325		0.1485
HQCNN-SE (Scenario 1)	1.7785		0.0025
HQCNN-SE (Scenario 2)	1.7313		0.0497
HQCNN-HEA (Scenario 1)	1.7820		0.0010
HQCNN-HEA (Scenario 2)	1.6996		0.0814
HQCNN-QI (Scenario 1)	1.7749		0.0061
HQCNN-QI (Scenario 2)	1.6673		0.1137

Supplementary Files

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