Simulation in rubber formulation development using AI^{Graf} Compounder

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The integration of artificial intelligence (AI) into rubber compound development marks a transformative step in materials science. This article explores the use of AI^{Graf} Compounder software to simulate and optimize rubber formulations. By leveraging feedforward neural networks, the system can predict material properties based on ingredient compositions, reducing the need for extensive physical testing and accelerating the development cycle (refs. 1-5). The methodology emphasizes the importance of high quality, structured datasets, especially those derived from Design of Experiments (DoE) in combination with other experimental strategies for accurate simulations. Several case studies demonstrate the software's effectiveness across various compound types, including EPDM and NR formulations, showcasing its predictive capabilities and alignment with experimental data. Challenges such as data inconsistency, measurement errors and the limitations of unstructured datasets are addressed through targeted validation and statistical analysis. Ultimately, the fusion of AI and structured experimental design enables more efficient, data driven decision making in rubber compound development, paving the way for innovation and performance optimization across the industry.

Use of artificial intelligence in rubber development

The rapid advancement of artificial intelligence (AI) has revolutionized numerous industries in recent years. AI has already led to profound changes in image and speech processing. However, the combination of high computational power and intelligent algorithms also opens up new possibilities in other areas, such as materials research and development (ref. 6).

The rubber industry faces challenges similar to those encountered in other material science disciplines: high development costs, lengthy testing cycles and the need to tailor materials precisely to specific requirements. Implementing AI to allow simulation processes offers a promising solution. Just as numerical simulations are used in the automotive industry to predict mechanical stresses, crack growth in lifetime experiments of metal, plastic parts or even in another field like chemistry to model reaction mechanisms, AI could significantly advance rubber development, as well.

Simulation as a basis for material development

A central aspect of rubber material development is the simulation of formulations and their properties. Every simulation requires precise data and relies on a well defined relationship between input parameters and the resulting material response. In an AI powered environment, rubber formulations with their properties can not only be predicted, but also analyzed in terms of their expected physical and chemical dependencies and correlations (refs. 7 and 8).

The core of such an AI driven approach is a neural network based on feedforward AI software (ref. 9). This structure enables the capture of complex relationships between formulation components and their properties. Depending on the application, recursive networks or deeper machine learning systems may also be utilized in the future. In this case, a feedforward network is employed, which is optimized through target criteria modification to provide increasingly precise predictions.

Once the criteria's fitness function identifies an optimal solution or the best balance between conflicting targets of the defined material properties, the system generates a concrete formulation recommendation. These recommendations are based on existing data and allow for further analysis and optimization.

A data driven model for the rubber industry

Data driven modeling is playing an increasingly important role in the rubber industry. Similar to the financial sector, where algorithms make credit decisions based on historical data of any customer, AI can create an objective decision making system in materials science. This reduces experimental effort and enables more efficient development cycles.

One major advantage of this method is the simultaneous consideration of multiple target variables. While humans can analyze only a limited number of factors at a time, a neural network allows for a comprehensive assessment of all relevant parameters. This leads to more precise and reliable predictions closer to what is in the mind of the expert. However, the quality of the results depends significantly on the underlying database. A well structured, sufficiently large dataset is essential for meaningful simulation outcomes.

Testing AI software

When developing new polymer formulations, AI software is often tested through validation experiments (ref. 10). Selected datasets are removed from the database, and the AI software must be able to calculate the missing values based on the remaining data sets. To perform this test, the physical properties are targeted which belong to the removed dataset. The success, pass or fail of this test depends on how accurately the AI reconstructs the formulation of the material with its ingredients and physical properties values.

An alternative testing strategy involves directly analyzing the ingredient values of the formulation. The results show that while the method is fundamentally effective, it has its limits, as already stated, by the homogeneity and, to some extent, even distribution of ingredient value data within the database. These tests are crucial for evaluating AI performance in simulation and identifying potential weaknesses early on. The problem may be solved with adding data to fill the data gaps.

Simulation and data analysis opportunities

A major challenge in the rubber industry is the accuracy of ma-

terial property measurements. Measurements are often error prone, but these errors are not always, if not seldom, normally distributed. As a result, simple statistical analyses of a historic database are not only insufficient for obtaining reliable insights, but rather impossible.

For example, the analysis of a small database containing 33 formulations can be visualized using different types of diagrams, such as bar charts or 3D scatter plots, to illustrate ingredient distribution and ingredient property influence. These methods provide an initial assessment of which calculations can be effectively performed. In fact, if there is an uneven distribution of factors, a full scale of solutions is restricted.

Additionally, correlation diagrams can be used to analyze relationships between specific material properties to have a first impression of the data. For instance, there is a physical correlation between hardness and modulus, as both parameters are linked to the stiffness of the material. Such correlations can be used to assess data quality and ensure that model assumptions are plausible.

Practical procedures for simulation

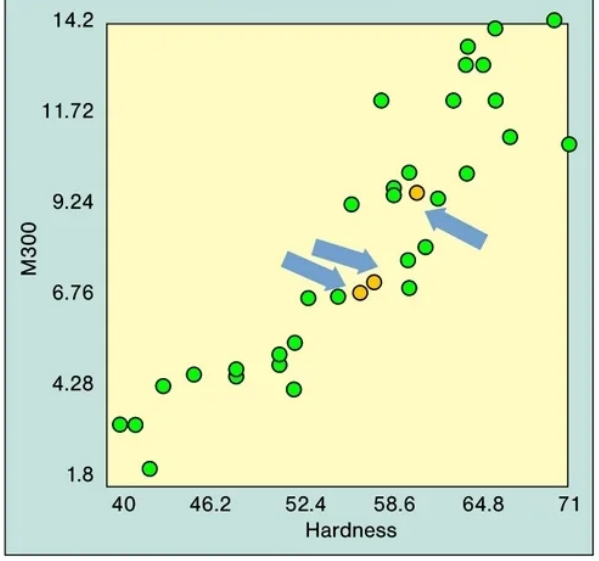
To conduct an AI driven simulation, data must first be imported into the system. This can be done through spreadsheet imports or direct database integration. Subsequently, all datasets can be analyzed collectively, or specific datasets can be excluded to simulate particular scenarios.

A useful strategy involves deliberately specifying contradictory criteria to observe how the model responds. The simulation results can then be cross-checked with expert knowledge and validated through further experiments.

Example 1: Correlation diagram

A literature database is used to analyze the relationships between formulation components and material properties (refs. 11-13). As expected, the correlation between hardness and modulus is relatively weak due to the influence of various ingredients in different formulations. The frequency distribution diagram reveals that not all solutions may be feasible, depending on the homogeneity of the database (figure 1).

Figure 1 - two compounds excluded; position in correlation diagram of predictions with remaining data indicated with arrows



For the first calculation, the target for carbon black CB N550 is set to 40 phr, with no plasticizer included. However, the formula with this composition was initially blocked. The calculation yields a close match to the blocked formula and its corresponding property data without requiring any modification of the criteria (weight/trade-off).

In the second calculation, a different formula is excluded, but its ingredients and corresponding values are used as criteria. The weighting for all criteria is set equally at 40. The results show that the amount of carbon black N330 does not exactly meet the target. However, by increasing the weighting, the second calculation successfully aligns with the target.

When integrating this compound into the hardness-modulus diagram, the data points are observed to be in close proximity (figure 1: first calculation, upper right; second calculation).

This example demonstrates that using ingredient based predictions instead of property based predictions can yield reliable results, provided that sufficient data are available to support the solution.

Example 2: DoE simulation and confirmation experiment

Expanding on the concept of using ingredients instead of properties, this example explores the prediction of a series of compounds following a DoE approach (ref. 14). A fractional factorial DoE plan was created utilizing data from example 1, which was subsequently validated through experiments. Further analysis allows for a comparison between the predicted and experimentally produced compounds, enabling statistical evaluation through the DoE program. This approach helps assess the accuracy of the data by comparing experimental results with database records.

Findings from this study contribute to the development of predicted compounds that align with the DoE scheme. This experiment, previously reported (ref. 15), utilized the same database as example 1. A fractional factorial design was chosen, with the following factors:

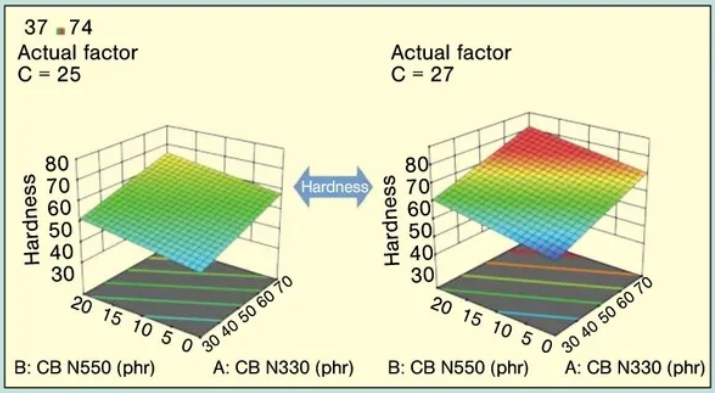
- CB N330: 30-70 phr
- CB N550: 0-20 phr
- Naphthenic oil: 5-45 phr

The compounds were then mixed, vulcanized and tested at Mahidol University, Bangkok, Thailand (ref. 15). The calculations for the compound compositions were performed using the AI software AI^{Graf} Compounder. Finally, the two DoE datasets were analyzed and compared using the DesignExpert software.

The results (figures 2 and 3) indicate differences largely influenced by variations in the molecular weights of the polymer, and to some extent by differing mixing procedures which remain unknown for past database entries.

The slopes of the curves in figures 4 and 5 are similar, and the midpoints of all the planes are in the same order. To quantify the alignment between results, the mean data of the experiment and simulation are provided in table 1.

Figure 2 - experiment (left) and simulated (right) Design of Experiments (DoE): hardness dependency from CB330 and CB550 shown in 3D plot



Only the value for Mooney scorch time deviates significantly beyond any expected measurement error. However, all other values fall within acceptable measurement error margins.

Overall, the results demonstrate a strong alignment between simulation and experimental outcomes except scorch time, with minor discrepancies attributable to differences in mixing procedures and polymer properties. Furthermore, they highlight the advantages of integrating DoE techniques with AI driven database analysis and laboratory practices.

NBR bushing/dynamic hardening dependent on fillers

This study examined the effect of silica beside the influence on carbon black on the dynamic hardening of an elastomer bushing (ref. 16). To evaluate the role of silica in a compound's dynamic behavior, three levels of carbon black N550 were selected, while the silica content was increased stepwise with the plasticizer remaining constant. Due to the symmetry of the data, it was possible to calculate formulations that fit within the expected scheme; however, minor variations in the plasticizer content had to be accepted.

The results show a significant influence of carbon black on M100 and static modulus (C_{stat}) (figure 4). The resulting curves looking almost identical, exhibited some non-uniform and erratic behavior at CB N550, 27 phr level, prompting further investigation of outliers and errors within the DoE.

By fitting regression curves, corrections were made, but only to the data sets used in the calculation. The fitted data replaced the original values, and the curves were recalculated. Notably, these corrections impacted the lower and middle levels of carbon black content, while the upper level remained unaffected due to a lack of data for high M100 and C_{stat} compounds (figure 5).

As described above, the dynamic hardening factor (C_{dyn}/C_{stat}) was calculated in the same manner. The first run used the untreated data, while the second run incorporated the corrected data. The resulting curves for the lower and upper levels of carbon black were nearly identical. However, at the middle carbon black level (CB550, 27 phr), a noticeable slope change was observed at lower silica concentrations (figure 6), indicated with an arrow. This suggests that further analysis, or even a complete repetition of the DoE, may be necessary to verify the findings.

Table 1 - data comparison of experimental and simulated DoE

Response name	*Units*	*Mean experiment*	*Mean simulation*
Gravity	g/ccm	1.11	1.11
Mooney	MU	32.71	38.42
Mooney t5	Minutes	8.97	26.30
Hardness	Durometer A	54.88	54.72
M300	MPa	8.27	7.75
Tensile	MPa	21.24	22.45
Elongation at break	%	547.13	585.28
Compression set, 70°C, 24 hours	%	50.79	32.37

Figure 3 - experiment (left)and simulated (right) DoE: §D plot shows result for compression set dependency on CB33 and oil

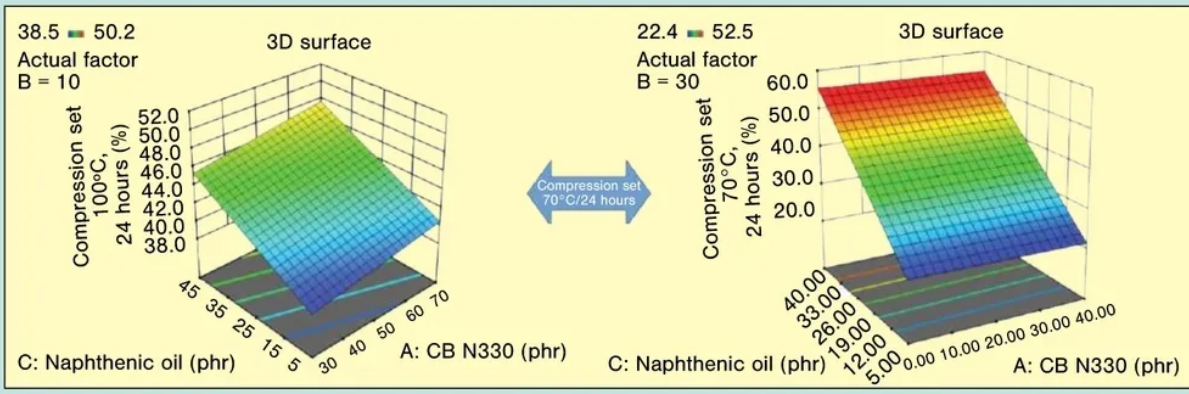


Figure 4 - modulus 100 over CB N550 and silica

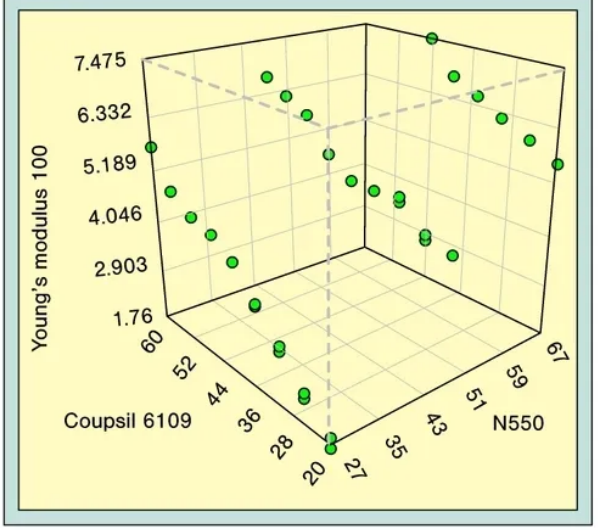
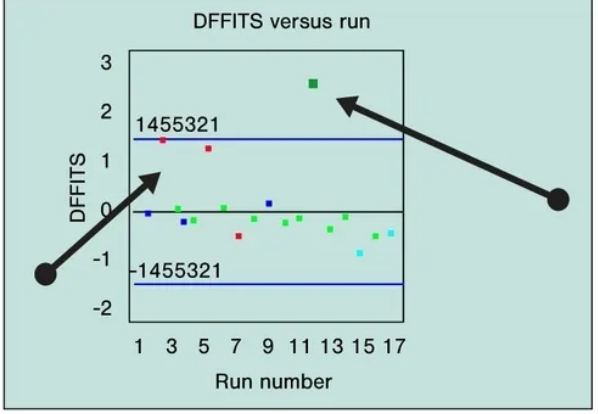


Figure 5 - difference in fits (DFFITS) versus run of experiment used for simulation; arrows indicate deviations



The simulation revealed a strong dependence of the AI software on data quality, which could be further optimized through statistical error analysis. This study demonstrated that a detailed statistical evaluation is required for each DoE factor, even when the predicted and actual data initially appear to align well.

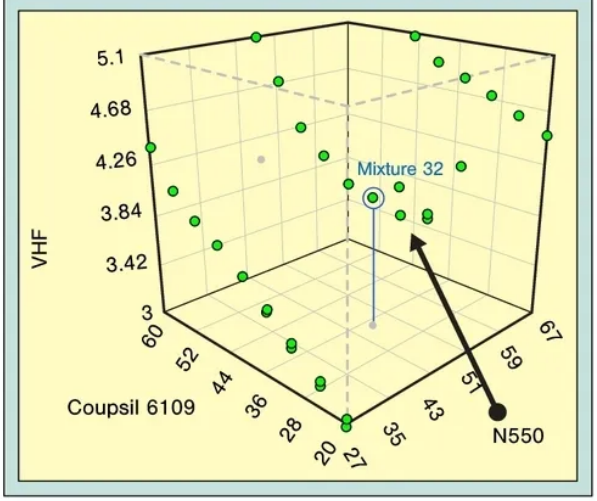
The graphs clearly highlight the dominant influence of fillers on the viscoelastic hardening factor (VHF), while the effects of almost all other ingredients appear minimal, or even negligible.

Simulation of accelerator combinations in EPDM compounds

This study investigates the impact of a co-accelerator under varying sulfur concentrations (ref. 17). While the simulation shows good agreement with experimental data, potential sources of error in the original dataset must be carefully considered.

The data used in this analysis come from a larger central composite DoE with seven factors. A major challenge in such extensive experimental designs is the significant time and effort required to mix and prepare all the compounds. Given the complexity, one would expect a larger deviation or an increase in errors. Surprisingly, however, no such trend is observed in the prediction versus actual plot.

Figure 6 - NBR compound dynamic hardening (VHF): 3 CB N550 levels and stepwise decrease of silica level



In the simulation, two sulfur levels were considered while the accelerator ZdiBC was increased stepwise in 0.5 phr increments (figure 7). Instead of producing smooth curves, the results showed a distribution of data points around a general trend line.

In this case, two possible approaches can be considered:

- Refining the dataset using DoE software to fit all data points onto a regression function
- Repeating the simulation procedure to validate the results

From a statistical perspective, the first approach introduces

Figure 7 - stepwise change of ZDiBC at two sulfur levels with questionable result due to faulty data

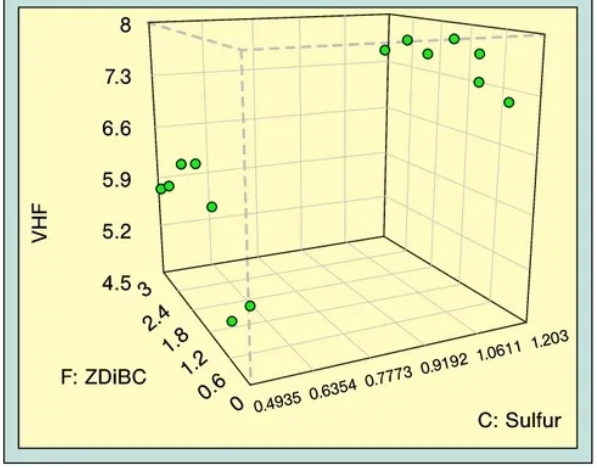


Figure 8 - modulus 100 over stepwise increase of MBTS at two sulfur levels, but constant SDT/S 2.2 phr

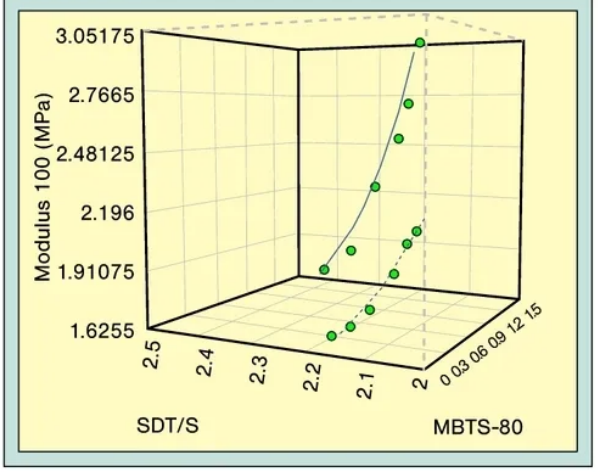
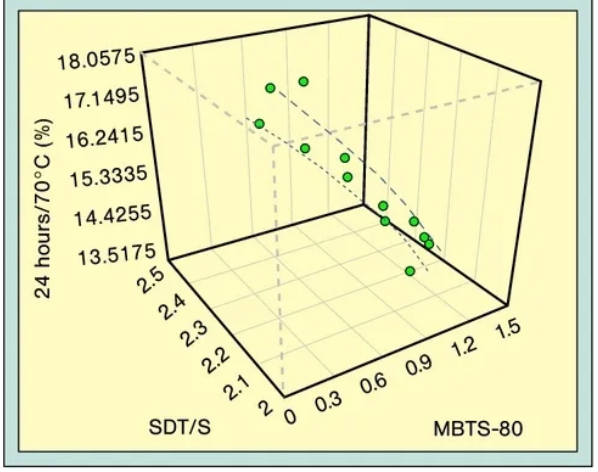


Figure 9 - compression set over MBTS stepwise increased at two sulfur levels, but constant SDT/S 2.2 phr



uncertainty, as it may distort the data's natural variability. Therefore, it is recommended to repeat the DoE with fewer factors per block, and use a folding technique to improve reliability.

In general, larger DoE studies require deeper statistical analysis to prevent misinterpretations and ensure accurate conclusions.

Natural rubber compounds: Simulation

In this example, a more unstructured database is used, consisting of trial-and-error experiments alongside small blocks of datasets generated using DoEs. All compounds are based on natural rubber (NR) (ref. 18). The distribution of ingredients and their values is sufficiently broad to allow multiple simulations.

Most common NR compounds utilize vulcanization systems based on sulfenamides and a so-called kicker. However, for the compounds in this study, the primary building blocks are dithiophosphates.

To demonstrate the simulation approach, the following conditions were examined:

- Sulfur at two levels: 1.5 phr and 2.2 phr
- SDT/S (poly-sulfur dithiophosphate) at 2.2 phr
- MBTS, with a stepwise increase from 0.0 to 1.5 phr

This setup was used to investigate the possible synergy between SDT/S and MBTS (figure 8).

The modulus 100 curves exhibit an almost parallel shift, but at higher MBTS concentrations, the slope becomes significantly steeper at the high sulfur level compared to the low sulfur level (figure 9). The elongation at break curves show a similar trend, with a flatter maximum at the low sulfur level and a steeper slope at higher MBTS levels. Interestingly, these curves appear to be an exact inverse of the modulus curves.

The compression set curves at both low and high sulfur levels demonstrate a strictly parallel shift with identical slopes across all MBTS levels. These curves are almost linear. Given that SDT/S and MBTS form a reversion resistant acceleration system, the compression set depends solely on crosslink density, and is not influenced by network breakdown due to amine based decomposition products. Since all other ingredients were held constant in this simulation, the observed dependency of the compression set on the accelerator combination was expected.

In conclusion, such simulations allow for fine tuning of a crosslinking system to optimize compound performance. However, the quality of the database must be sufficient to support accurate calculations. The simulation, the increase of MBTS concentration in the compound, was possible with this combinatin of SDT/S and sulfur only due to insufficient data for other simulations.

Conclusion

The implementation of artificial intelligence in rubber compound development represents a significant advancement, enabling faster, more precise and more efficient optimization of formulations. The examples above highlight key insights into the role of AI driven simulations and their dependencies:

- AI-driven simulations offer precise predictions but are highly dependent on data quality. The accuracy of any simulation is constrained by the reliability, consistency and completeness of the dataset used. Errors, missing values or inconsistencies in the data can lead to misleading results, emphasizing the need for thorough data validation before applying AI models.
- Design of Experiments (DoE) provides well structured and statistically distributed factor values, whereas random datasets often require significant cleaning. Well planned DoE studies ensure balanced data distribution, reducing the likelihood of errors and biases. In contrast, datasets derived from trial-and-error approaches may lack structure, requiring additional preprocessing to extract meaningful insights.
- The integration of AI and statistical experimental design enhances prediction accuracy and enables more targeted studies. Combining AI with DoE techniques creates a

powerful framework for data driven decision making. This approach not only improves model reliability, but also allows for deeper analysis of compound behaviors under varying conditions.

By merging AI driven simulations with systematic experimental design, the rubber industry can significantly accelerate its development processes, while maintaining a high level of accuracy and efficiency. This integration reduces the reliance on extensive physical testing, lowers costs and enables faster iterations in compound formulation. As AI models continue to evolve, their predictive capabilities will further refine material properties, unlocking new opportunities for innovation and performance optimization in rubber technology.

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