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A Machine Learning Based Approach for Selection of SF_6 Alternatives

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SUMMARY

Finding a suitable replacement for sulphur hexafluoride (SF_6) in the gas insulated equipment is a major challenge facing the energy industry due to its incredibly high global warming potential (GWP), which is 25,200 times greater than that of CO_2 with an atmospheric lifetime of 3,200 years. There are around 114 million unique compounds in the PubChem database, and it is physically impossible to test all existing chemical compounds through laboratory scaled investigation. An iterative approach of mixture optimisation with a genetic algorithm can help narrow down the search space to a more feasible number of candidates. Besides a vast number of compounds and mixture combinations, there are also a range of parameters such as dielectric strength, boiling point, toxicity and GWP that must be collectively considered, which points to the application of advanced multi-objective optimisation techniques for balancing all the required properties. The benefit of the computational approach is clearly evident as the generated mixtures contain researched solutions reported in the literature such as C_3F_7CN and CF_3I. Furthermore, the developed approach is effective at identifying an optimal set of mixtures from a large space of possible mixture combinations and ratios.

KEYWORDS

Machine Learning, Sulphur Hexafluoride (SF_6), Environmentally Friendly Alternatives, Genetic Algorithms, Multi-objective Optimisation, Dielectric Strength and Gas Insulation.

1 Introduction

SF_6 is a potent greenhouse gas that is extensively used in electricity transmission and distribution equipment. It has been reported by National Grid to be their largest controllable element of direct emission at ~280,500 tonnes CO_2 equivalent in 2018/19 [1]. Replacing it with an environmentally friendlier insulating gas is an important step to decarbonise the electricity grid. Finding an alternative to SF_6 is a challenging task due to its outstanding insulating properties and the ability to remain gaseous at elevated pressure which allows the gas-insulated equipment to be compact and reliable. Besides that, any research cycle of new gas candidates will be costly and time consuming as it relies on extensive volume of experiments to evaluate the key properties of mixtures. In combination with a very large search space of chemical compounds, there is a high chance of missing the optimal solution. Therefore, there is a need for a computational approach to narrow down the number of potential candidates using pre-defined evaluation criteria.

Currently, there are several proposed alternatives that possess higher dielectric strength and reduced GWP than SF_6, the most prominent one being C_3F_7CN for high voltage (HV) applications. This gas possesses a dielectric strength double of SF_6 and a GWP of 2,240. Due to its boiling point limitation, it is typically used as part of a binary or tertiary mixture for new-build [2] and retro fill applications [3]. However, there is an increasing discussion on banning the use of Perfluoroalkyl and Polyfluoroalkyl Substances (PFAS) with 3M, the main manufacturer of C_3F_7CN, recently announcing that they will stop their production of PFAS materials by the end of 2025 [4]. It is clear there is no consensus on a single replacement candidate for SF_6 in the market and the upcoming F-gas regulation 2023 could drastically change the outlook on SF_6 alternatives. This reinforces the importance of developing a faster assessment approach to evaluate different candidates due to uncertainties in future regulation changes to production and use of PFAS. In this paper, a methodology combining clustering and genetic algorithms applied to optimisation of gas mixtures from compounds reported in the literature is presented.

2 Estimation of Gas Mixture Properties

As this work is concerned with selection of gas mixtures, calculation of key properties that define the suitability of any gas mixture for HV insulation is essential for evaluation of alternatives. Common calculation methods require a lot of input parameters or experimental data which are difficult to gather for a large set of compounds. Hence, simplified estimation algorithms were developed for properties where existing approaches are unfeasible. It is important to note that dew point and dielectric strength are dependent on temperature and pressure. Hence, this work calculates all values for standard ambient temperature of 25 °C at 1 bar absolute.

2.1 Global Warming Potential (GWP)

A method for GWP calculation of a mixture was found in [6]. It uses a weighted sum, shown in Equation (1). The overall mixture values are proportional to the GWP of the gases and the ratio of their molar mass in the mixture.

$$GWP_{mix} = \sum_{i=1}^{n} m_{\%i} * GWP_i \quad and \quad m_{\%i} = \frac{mole_{\%i} * m_i}{\sum_{j=1}^{n} mole_{\%j} * m_j} \tag{1}$$

where *mole%* - mole percentage, *m* – molar mass, *m%* - mass ratio, n – number of compounds.

2.2 Toxicity

Health effects of a gas are usually evaluated by measuring its acute toxicity estimate (ATE). Gas inhalation toxicity is typically expressed as LC_{50} – concentration of substance that kills 50% of test subjects in a given time period. The standard exposure time is 4 hours and, in this case, LC_{50} is equal to ATE in accordance with Globally Harmonized System of Classification and Labelling of Chemicals (GHS) [7]. The toxicity of a mixture is calculated using Equation (2).

$$\frac{100}{ATE_{mix}} = \sum_{i=1}^{n} \frac{C_i}{ATE_i} \tag{2}$$

where C_i is the concentration of *i-th* compound, n the number of compounds, and ATE_i is the acute toxicity estimate of the *i-th* compound [7].

Note that direct use of ATE in the algorithm is not suitable as there might be large differences between values even if gases are non-toxic, which will add unwanted bias to gases like oxygen (O_2). Hence, there needs to be a flooring function that keeps all toxicity values in a sensible range. Table 1 shows conversion of hazard categories and range values to a single ATE point suggested by GHS which is adopted in the algorithm before applying Equation (2). Note that gases with LC_{50} greater than 20,000 ppm/4h are assigned a value of 5,500.

Table 1. Conversion of hazard categories and ATE ranges to a point estimate [7]

Exposure route	Classification category or ATE range	Converted acute toxicity point estimate
Inhalation (ppmV)	0 < Category 1 ≤ 100	10
	100 < Category 2 ≤ 500	100
	500 < Category 3 ≤ 2500	700
	2500 < Category 4 ≤ 20000	4500

2.3 Dew Point

Boling point is a widely available property for many compounds but a more suitable quantity to describe the liquefaction temperature at a given pressure is the dew point. Dew point is the same as boiling point for single compounds and only diverges in mixtures [8], which means that both can be estimated if one of the properties is known.

The common way of calculating dew points of gas mixtures is to solve the Peng-Robinson Equation of State. However, this approach requires an input of a critical temperature, critical pressure, and an acentric factor for each compound which is unfeasible for the purpose of screening large volume of gas candidates. Therefore, a simpler estimation algorithm was developed that only requires boiling points of mixture components to be known and Excel tools with modified MATLAB programs by Carl Lira and Richard Elliot [9] were used to validate the method.

Initially, a simple weighted sum Equation (3) was tested for several binary mixtures, but the error of this linear estimation approach rapidly grows as the difference between boiling points of compounds increases. This led to implementation of another estimation method – a quadratic Bezier curve. Equation (4) shows how the Bezier curve is calculated from three input points.

$$T_d^{mix} = \sum_{i=1}^{n} mole_{\%i} * T_b^i \tag{3}$$

where T_d^{mix} is the dew point of the mixture, T_b^i the boiling point of a compound, *mole%* - mole percentage.

$$Q(t) = (1-t)^2 P_0 + 2(1-t)tP_1 + t^2 P_2, \qquad 0 \le t \le 1 \tag{4}$$

where P_0, P_1, P_2 are points on a two-dimensional plane.

As shown in Figure 1, the Bezier approach provides better estimation for mixtures with a wider difference in boiling point but overestimates smaller changes. Similar trend was observed for tertiary mixtures as it can be assumed as a binary mixture neglecting the compound with the boiling point that lies in the middle of the three compounds. Therefore, a composite method is used to determine the dew point of a mixture i.e., linear estimation when difference is less than 1.2 and Bezier curve above 1.2.

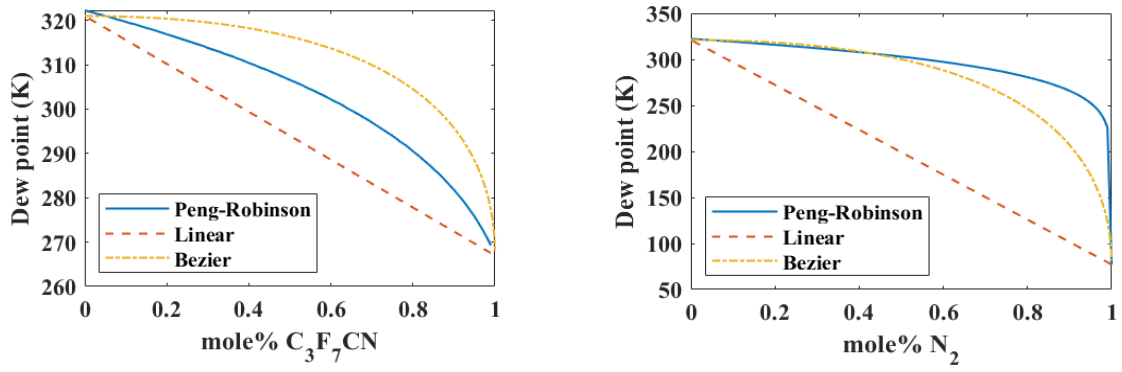

Figure 1. Comparison between dew point estimation methods (Bezier, linear) with calculated by Peng-Robinson Equation of State values [9], (a) – C_3F_7CN and $C_6F_{12}O$ mixture with boiling point difference of 1.2 °C (b) – $C_6F_{12}O$ and N_2 mixture with boiling point difference of 4.16 °C.

2.4 Dielectric Strength

Accurate estimation of the dielectric strength is the most challenging out of all the investigated parameters. A common method is the use of the Boltzmann equation which mathematically describes collision of particles of a given chemical compound. However, this requires accurate electron swarm parameters that can only be obtained experimentally.

Existing data of several mixtures shows that the change of $(E/N)_{crit}$ with respect to gas concentrations is highly dependent on the compounds. For example, mixture of CF_3I and N_2 has a linear trend but for SF_6 with N_2 the line is curved for 0 to 40% of SF_6 within the binary mixture [10]. Further complication is introduced for tertiary mixtures. For an initial screening process of different gas candidates, it was decided in this study to adopt a weighted sum for $(E/N)_{crit}$ estimation similar to Equation (3). Such simplified approach produces an error up to 30% for certain mixtures but the general trend is correct. Hence, it allows the genetic algorithm to select optimal mixtures correctly as shown in the results analysis despite the error margin.

3 Approach Development

The selection of gas mixtures as new SF_6 alternatives is based on several parameters that must be considered simultaneously. This makes the comparison of the mixtures challenging as candidates can be better or worse in different aspects which means there is no single one-for-one solution. To narrow down the search space, multi-objective optimisation techniques are used to identify a set of solutions as close to the desired properties range as possible. In this work the aim is to minimise dew point, GWP and to maximise dielectric strength and acute toxicity estimate.

There are several machine learning techniques that can perform multi-objective optimisation as summarised in [11], but the most suitable approach for gas mixture generation is an evolutionary algorithm. More specifically, a genetic algorithm is explored in this work for gas mixture optimisation as it allows a close representation of mixtures as virtual objects. It is a simple, yet powerful algorithm based on the natural evolution processes. In addition to that, clustering of data points is added at the initialisation stage to define the structure of mixtures generated in the main algorithm.

3.1 Genetic Algorithm

The mixture evolution program is based on a genetic algorithm and its structure is shown in Figure 2. Genetic algorithms start with a random population of candidate solutions called ‘individuals’ that are used to iteratively produce new generations by crossover and mutation of selected ‘parent’ candidates. The input to the program is a dataset of 33 compounds which are filtered to remove highly flammable gases, resulting in 26 data points with known LC_{50}, GWP, $(E/N)_{crit}$ and boiling point. To enhance mixture generation, clustering of the dataset (further explained in Section 3.2) is performed to define a structure for all individuals. A stopping condition can be based on a certain feature of the population but, in this case, there is no definitive measure of candidate performance. Therefore, the algorithm uses a fixed number of iterations that can be adjusted after manual analysis of the outputs.

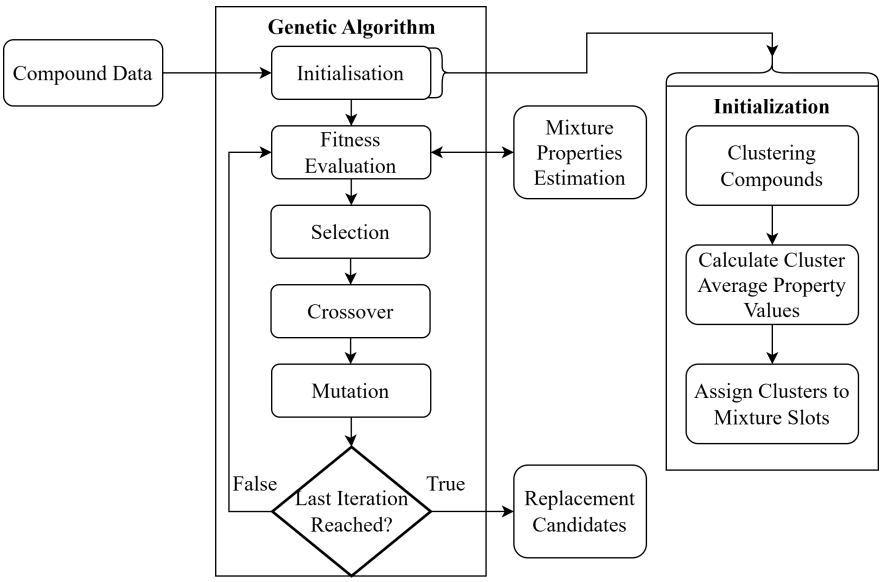

Figure 2. Mixture evolution program flowchart

Mixture generation requires both the compound combination to be identified and then the ratios to be assigned to each compound, making this a two-layer problem. This issue is tackled by exploring all possible ratios with a fixed step (typically 1 to 5%) for each mixture and selecting the best solutions which leaves only the compound combinations to be optimised.

All mixtures with their respective ratios have their properties evaluated and assigned as a multi-objective fitness. Then the optimal set of solutions are selected so that only the best candidates are used for further iterations. This is carried out by identifying a Pareto front which only contains non-dominated solutions with its principle illustrated in Figure 3.

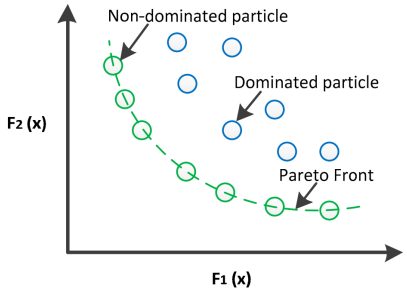

Figure 3. Pareto front and individual / particle domination principle [12]

A set of optimal mixtures is used to calculate the popularity of compounds which is simply their number of occurrences in the Pareto front. A sum of popularities of each compound in the mixture is treated as an overall fitness of the individual. This approach reduces the dimensionality of the problem to a single parameter and enables a simple parent selection with a roulette wheel where the probability of an individual being chosen is based on the total popularity of the compound in the mixture.

After two parent individuals have been chosen, there is a fixed probability that one of their compounds are randomly swapped to create 'offspring'. Mutation of the newly generated individuals is implemented as a small probability of a child-individual to have one of its compounds replaced by a random one from the dataset. The new generation has the same number of individuals so to keep the population size constant. Note that individuals can be both binary and tertiary mixtures.

3.2 Hierarchical Clustering

Random selection of mixture compounds is not a suitable way to generate and evolve candidate solutions because this can result in unusable combinations such as having three fluorinated gases. Therefore, having compounds clustered allows a more structured and informed approach to mixture

generation which narrows down the number of investigated candidates as well as gives control over the compound combinations searched by the algorithm.

Hierarchical clustering method was selected as any desired number of clusters can be easily identified by looking at the dendrogram which is not the case for other clustering methods. Besides that, the size of the input dataset is small, hence the pairwise distance calculations are quick. The features used to group the data points are toxicity, GWP, $(E/N)_{crit}$ and boiling point. Figure 4 shows the resulting five clusters of 26 compounds indicated by different shapes.

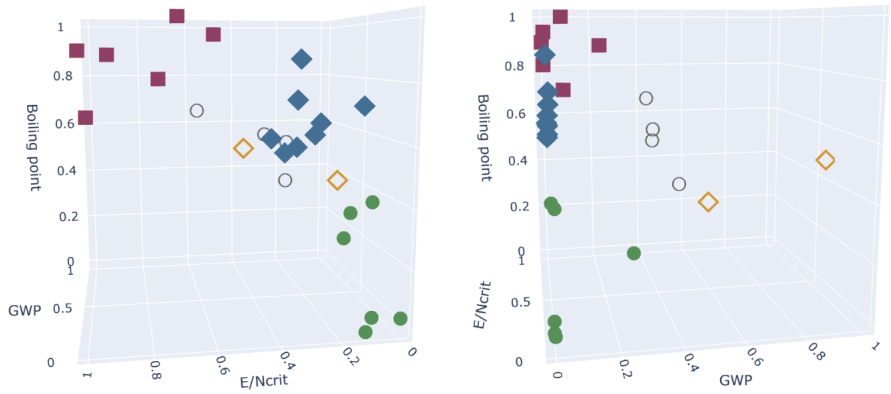

Figure 4. Clustered compound dataset of 26 compounds using scaled between 0 and 1 values of toxicity, GWP, $(E/N)_{crit}$ and boiling point. Filled circles - cluster mostly contains buffer gases such as N_2, CO_2, O_2, filled squares - cluster consists of fluorinated gases with high dielectric strength like C_3F_7CN and $C_5F_{10}O$. Filled/unfilled diamonds and unfilled circles - average parameters but a noticeable difference in the GWP range (includes SF_6, CF_3I, etc). Same plot shown from different angles. Toxicity is not shown on the axes.

Identified clusters are then used to define the structure of generated mixtures by assigning each group to the slots in the container object. This process is manually reviewed before running the genetic algorithm to ensure a reasonable position for clusters. Figure 5 visualises the structure of individuals. For the current dataset the first slot is filled with a compound from clusters with high dielectric strength, the second slot is open for any compounds and the third slot only allows compounds with low GWP and low boiling point.

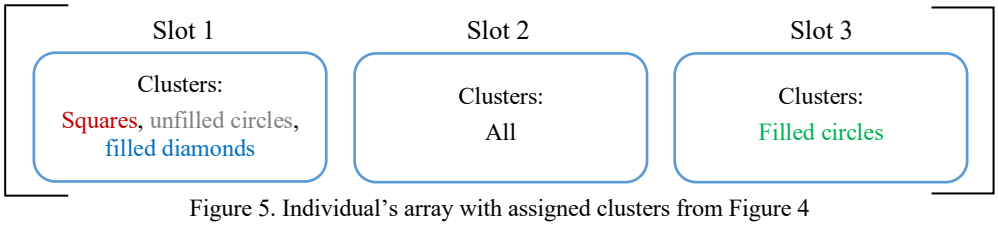

Figure 5. Individual's array with assigned clusters from Figure 4

4 Results and Analysis

The program ran for 100 iterations with 26 compounds to evaluate its performance. Population size was set to 200 individuals which is less than 7% of the total amount of compound combinations and fine-tuned probabilities of crossover and mutation were set as 0.7 and 0.3 respectively. Mixture structure used is shown in Figure 5 and ratio step used in mixtures is 5%.

It can be seen in Figure 6, that the generations have converged to a small set of gases with the highest occurrence in the Pareto front. CF_3I is the most popular compound with twice as many occurrences as the next leading compound in N_2. This is somewhat expected because it is one of the researched SF_6 alternatives [13] with a GWP of only 0.4. Another compound with high dielectric strength selected by the algorithm is C_3F_7CN but it has less occurrences than CF_3I due to a higher GWP value of 2,240.

O_2 and N_2 are also in the top four popular compounds which are the typical buffer gases used in existing applications. In practice, O_2 can only be used in lower concentrations as it is an oxidiser and can intensify combustion of flammable compounds. The use of CO_2 and N_2 is more common as the main buffer gas. The rest of the compounds are filtered out which is a great result considering the defined

evaluation parameters. Most of the dataset consists of unusable in practice gases which are either toxic, harmful to the environment or simply are outperformed by other gases in the dataset and the algorithm manages to select the better options to generate mixtures.

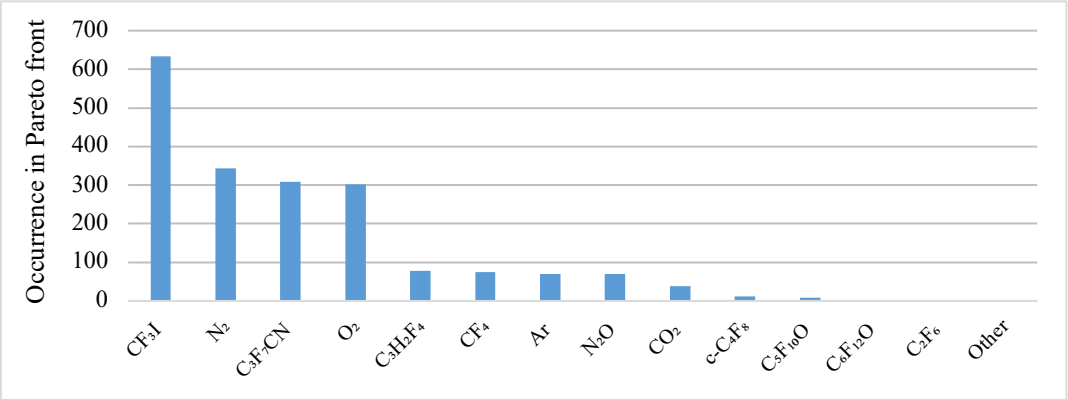

Figure 6. Compound occurrence in Pareto front after 100 iterations

The main output of the program is a set of optimal mixture combinations with concentrations, shown in Figure 7. Candidates in the resulting Pareto front can be split into three groups according to the property ranges. Green group is formed mostly by mixtures of CF_3I with various atmospheric gases as well as $C_3H_2F_4$. Its key property is low GWP but this comes at a cost of a limited dielectric strength of mixtures. Blue candidates are all combinations of CF_3I with C_3F_7CN and buffer gases. These solutions have the highest dielectric strength compared to other groups, but their liquefaction temperature is a concern.

Other solutions in the Pareto front are made up of compounds like CF_3I, $C_5F_{10}O$, C_3F_7CN combined with low boiling point gases which results in a middle parameter range. Many of the resulting mixtures, such as ones highlighted in Figure 7, are similar to the previously investigated SF_6 replacement candidates [13]. The rest of the output candidates contain promising compounds but must be experimentally validated before any judgment can be made regarding their suitability.

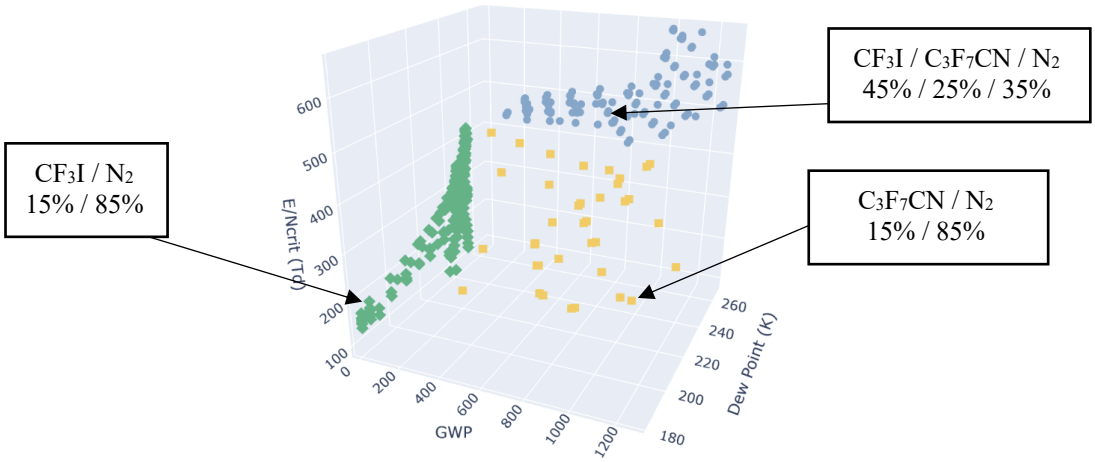

Figure 7. Pareto optimal solutions after 100 iterations: blue circle – $(E/N)_{crit}$ > 400 Td and boiling point > 255 K; green diamond – GWP < 100; and yellow square – other gases.

The developed program has successfully utilised a genetic algorithm to separate the gases with better features and use them in the mixtures. The ability of the program to identify the useful compounds based on many parameters is exceptional with 100% of the poor compounds being the least occurring in the output. However, a small size of the dataset limits the ability to test the algorithm. The convergence of the program on the desired compounds is fast in this setup but the same cannot be assumed for a larger variation of input gases. Therefore, there is no certainty in stability of the algorithm and larger scale runs must be done to evaluate the consistency of results.

5 Conclusions

A computational approach based on a genetic algorithm and hierarchical clustering has been developed to optimise gas mixtures from a set of compounds. The undertaken work has covered the issues related to the use of SF_6 as an insulation medium in power equipment and highlighted the benefit of using machine learning to select environmentally friendlier alternatives.

The main conclusions of this work are as follows:

1. Developed estimation methods for dew points, toxicity, GWP and $(E/N)_{crit}$ of mixtures minimise the amount of required input parameters and reproduce real trends close enough for the genetic algorithm to correctly evaluate the candidates.
2. Hierarchical clustering applied to a dataset allowed to manually define a structure of individuals in the genetic algorithm which focused generated mixtures on desired combinations.
3. Genetic algorithm for mixture optimisation produced a set of Pareto optimal solutions from a small starting population that is less than 7% of the total search space. Some of the resulting candidates are reinforced by previous research and all unusable in practice compounds have been filtered out with 4 out of 26 gases in CF_3I, C_3F_7CN, N_2 and O_2 being predominant in the resulting mixtures.

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