Chemical Reaction Space Elucidation Through Stochastically Guided Machine Learning

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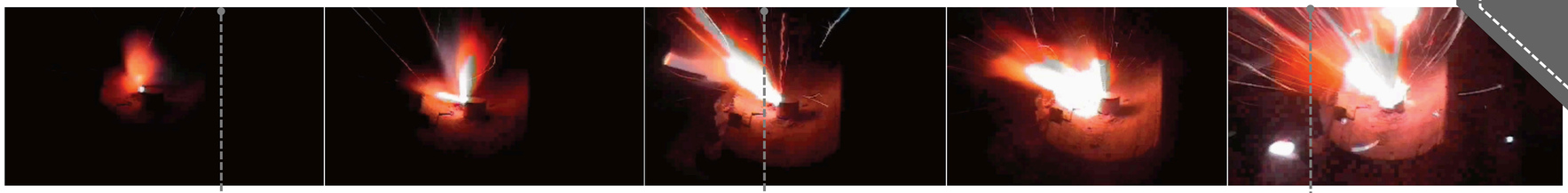
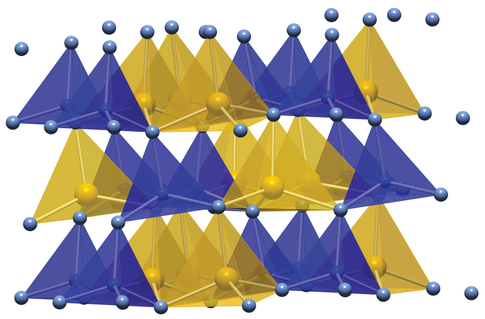


ABSTRACT

Chemical synthesis of compounds and materials are typically improved by understanding a reaction's mechanism and its kinetics. Many tools have been developed for performing these studies for solution and gas-phase chemistry. Solid-state chemistry presents a challenge, as reactions can be rapid and highly exothermic. Such chemistry is often used for the production of high performance ceramics. Optimization of these syntheses typically involves trial and error and this chemistry often is referred to as "shake and bake." One approach to dealing with this kind of challenge is to utilize schemes modeled after biological systems. By using processes observed in evolution, a robust algorithm can be developed for process optimization. This is called a genetic algorithm. In implementation of a genetic algorithm (GA) multiple "generations" of experiments are produced to optimize the reaction. This generational data is a representation of the reaction parameter space. By using this data as a training set, a neural network (NN) will be produced that can predict the reaction outcome from new reaction mixtures. Combining stochastic search routines with artificial intelligence offers an efficient method for improving and understanding solid-state syntheses.

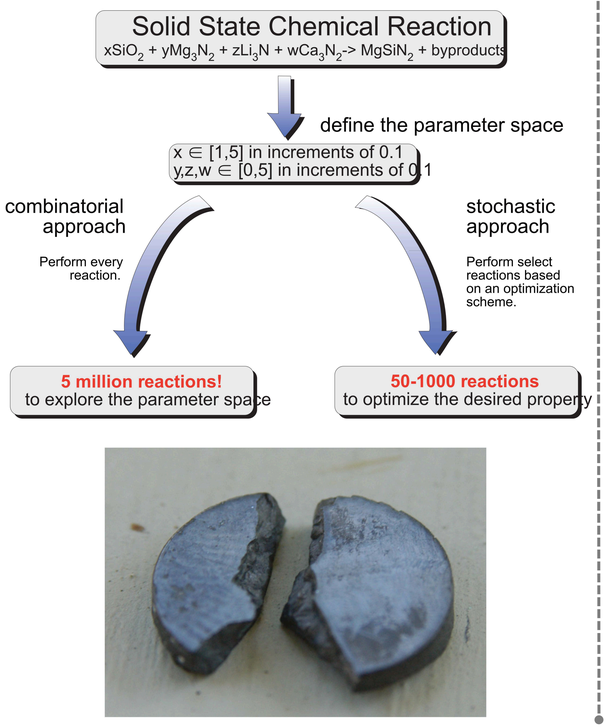
Introduction

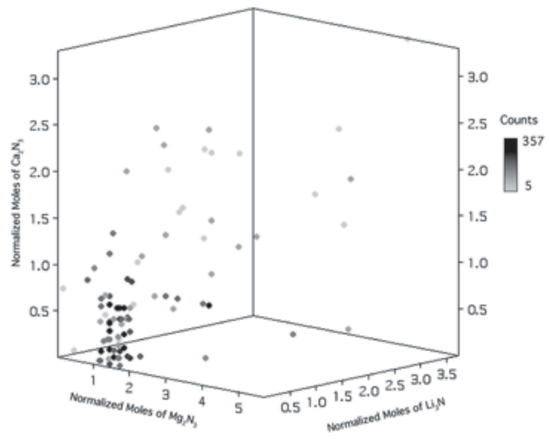


$MgSiN_2$ has many attractive properties such as high thermal conductivity, low dielectric constant, high hardness, high thermal stability, good oxidation resistance (up to 920˚C) and high electrical resistance at room temperature

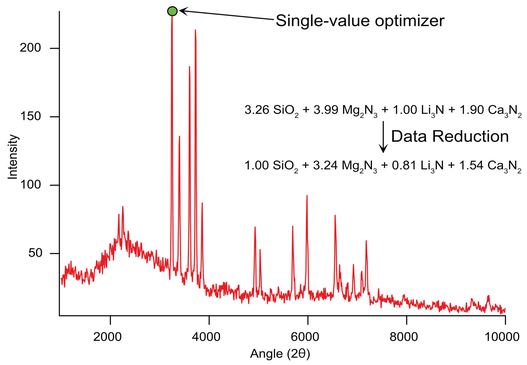
Mixed silicon nitrides as have attracted attention in applications required high thermal conductivity, low dielectric constants, and good thermal and mechanical stability. the Of these mixed nitrides, $MgSiN_2$ has been compacted to get a thermal conductivity up comparable to optimized Al_2O_3 with significant improvements in mechanical and chemical stability. A low cost route to $MgSiN_2$ from silicon dioxide would facilitate production of electrical insulators in power generation and transmission..

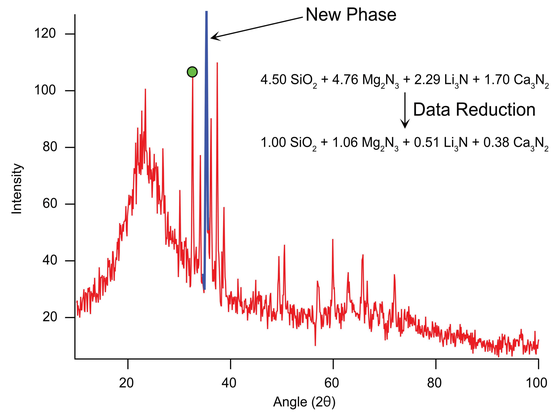
$MgSiN_2$ was first prepared from Mg_2Si or a mixture of Mg_3N_2 and Si_3N_4.[5] This was achieved by heating the reagent powders in nitrogen to 1200 ˚C for several hours. These approaches require significant inputs of energy to obtain the desired phase. A simple thermally initiated synthesis from silicon dioxide and metal nitrides would greatly reduce production costs. Previous work fond that $MgSiN_2$ could be produced by rapidly heating mixtures of SiO_2 and Mg_3N_2. However, significant reduction in yield was observed due to the formation os magnesium silicates. It was hypothesized that additions of alkali metal and alkaline earth metal nitrides could facilitate the formation of soluble silicates and $MgSiN_2$ with enhanced crystallinity. To that end a series of experiments were performed to stochastically optimize the crystallinity of the $MgSiN_2$ phase realized.





A map of reactions performed in a genetic algorithm search of the system: $xSiO_2 + yMg_2N_3 + zLi_3N + wCa_3N_2$ $MgSiN_2$ + byproducts. The intensity of the most intense $MgSiN_2$ peak, in counts, is indicated by the color of each point. The intensity of $MgSiN_2$ peak is indicative of product crystallinity. The $MgSiN_2$ crystallinity was increased from 170 to 357 counts.





If we take a look at these graphs of the powdered experiments, each graph represents different ratio compounds and taking account of the peaks, normally we get a single value optimizer for the benefit of finding the most optimized ratio of compounds. This makes it very inefficient in the long run because we don't get enough values to analyze the experiment as a whole, and this is where machine learning comes into play by taking and analyzing all the other values in order to gain a more refined optimized crystallized compound ratio. By finding this refined crystallized structure, it allows us to optimize the heat dissipation withing the final compound and ultimately apply it to final production. Because of the efficiency in almost all aspects of machine learning, leading energy corporations are able to apply this to everyday manufacturing and processes.

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