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NEURAL NETWORKS AS TOOLS FOR PREDICTING MATERIALS PROPERTIES

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Introduction

Materials science is of fundamental significance to science and technology because our industrial base and society depend upon our ability to develop advanced materials. Materials and materials processing cuts across almost every sector of industry. From microelectronics to polymers, from pharmaceuticals to petrofuels, from aerospace alloys to smart materials, our industrial base is built on our ability to invent and craft new materials. The key in all of these areas is the ability to rapidly screen possible designs which will have significant impact. However up to now materials design and processing have been to a large extent empirical sciences. As an example of the general problem, it has been found that empirical methods can sort out on the order of tens of thousands of drugs a year, however estimates show that only one cancer drug in 40,000 has clinical significance and that perhaps one in a million would be curative (at this rate it would take 100 years to obtain our "first cancer-curing drug"). In addition we are still unable to design new alloys and polymers to meet application specific requirements. Being able to do so quickly and at minimum cost would provide an incredible advantage, subsequently leading to significant advances in quantity and quality of materials products.

In order for a given material to be used in technology it must first satisfy a number of performance criteria and these criteria are becoming more and more stringent each year. At the same time the range of applications of some of the more common materials (i.e., polyethylene) have been nearly stretched to their limits. Materials suitable for advanced applications can have considerably more complex chemical structures (or can be blends of homomaterials, such as polymer blends) [1]. In short, the development of new high-performance materials to be used in a particular technology requires significant R&D. Obviously, the ability to predict physical, chemical, or mechanical properties of compounds prior to their synthesis is of great technological value in optimizing their design, processing, or recycling [1-3]. Such capabilities represent a great challenge to present computational methods since there are a large number of possible structures for a given compound (isomers) in addition to numerous possible compositions, atomic connectivities, and different processing techniques [4-6]. In this light, the development of new predictive computational schemes to evaluate potential candidates for specific applications has taken on a new level of importance. The computational methods that address this challenge should allow simple, rapid, and accurate determination of key properties for a large range of compounds. In addition, in order to realize the ultimate goal of *materials by computational design*, the reverse problem, prediction of chemical structure based on desired properties, has to be resolved.

Research at Oak Ridge National Laboratory has lead to the development of a novel computational paradigm (coupling computational neural networks with graph theory, genetic algorithms, wavelet theory, fuzzy logic, molecular dynamics, and quantum chemistry) capable of performing accurate *computational synthesis* (both predictions of properties or the design of compounds that have specified performance criteria) [7]. The computational paradigm represents a hybrid of a number of emerging technologies and has proven to work very well for test compounds ranging from small organic molecules [8] to polymeric materials [7]. Fundamental to the method is the neural network-based formulation of the correlations between structure and properties. The advantages of this method is in its ease of use, speed, accuracy, and that it can be used to predict both properties from structure, and also structure from properties.

Computational Scheme

Computational neural networks provide powerful tools for modeling of materials. In the chemical sciences, the use of computational neural networks has rapidly increased over the past 5 years (approximately

850 publications). There is now sufficient utilization of neural networks in chemistry that several review articles and a book is available on the subject[9-13]. It is the goal of this paper to present results on the use of computational neural networks in materials science as a method for making accurate predictions of polymer properties based on their molecular structure and for designing molecular-based compounds that have specified properties.

Figure 1 schematically shows the overall approach that was used in the research presented in this paper.

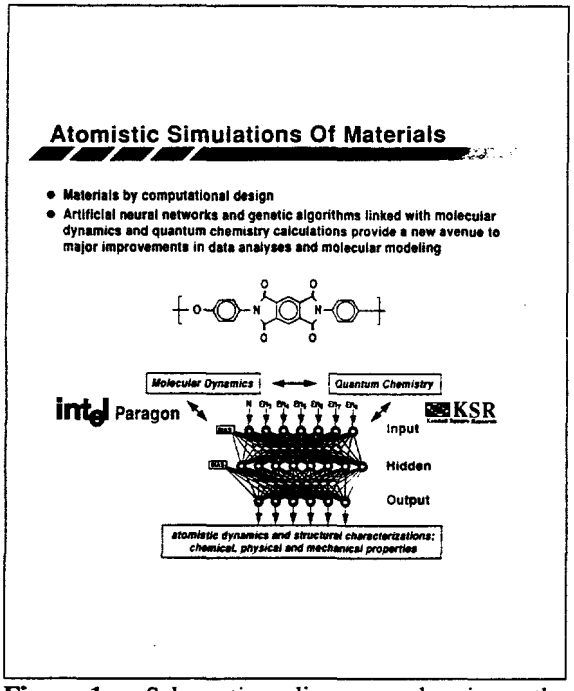

Figure 1. Schematic diagram showing the computational paradigm used to carry out materials modeling.

A given molecular structure for a repeat unit of a particular polymer, Kapton as shown, is transformed into a set of numerical descriptors (using graph theory for example). These descriptors are used as inputs to a computational neural network and a set of desired properties are used as outputs. The neural network will then adjust its connection weights in order to optimize the output prediction error. Once this phase (called training) is complete, the neural network can be given descriptors for polymers that were not originally used in the training phase and will output a set "predicted" properties, using the internal correlations that were learned from the training phase. This overall scheme can also serve to screen out descriptors that are not necessary to formulate the correct correlations. This task can be accomplished by performing sensitivity analyses on the trained network (changing one input and monitoring the changes made in the outputs) [14].

The underlying engine of the method resides in a general purpose neural network simulator (ORNLNET) [7] that utilizes a modified form of the learning algorithm of back-propagation (a hybrid stochastic-conjugate gradient method) to adjust the connection weights in a feedforward neural network consisting of several layers of nodes including an input layer, some hidden layers (the number of nodes is automatically optimized), and an output layer. The initialization of the connection weights is generally performed using adaptive simulated annealing or genetic algorithms which tends to start the network in a state that is much more optimal than that accessible for a random distribution. The overall goal is to teach the network to associate specific output states (desired answers) to each of several input states [14-17]. Having learned the fundamental relationship(s) between inputs and outputs, the neural network should then be able to produce the correct output for unknown input.

Forward Process: Prediction of Properties from Chemical Structure

In order to perform neural network predictions on polymer properties from a set of structural desriptors, a suitable training set must be obtained. In this study a number of different neural network architectures and topologies were used since a number of different experiments were performed. The more general network was one with an architecture of a single hidden layer and topology of 18 inputs (structural descriptors), 3 hidden, and 9 outputs (9 different properties). The network also had direct connections between the input and output layer and bias nodes. This particular network was determined to be optimal for the problem of making accurate predictions of 9 different properties (simultaneously) for a polymer data set.

As a starting point a number of descriptors that are useful for characterizing molecular structure were examined [5]. For the purpose of accurately predicting polymer properties, the use of the descriptors defined by Bicerano [1] appear to be an excellent choice. These descriptors are based topological indices obtained by using graph theory, and reflect both topological connectivity and information on the electronic structure. A new set of indices are incorporated into this basic set which take into account properties that depend on the amount of material present, called extensive, and those which do not, called intensive.

The indices developed by Bicerano were used as input to a neural network which then could sort out

which ones were needed to predict a set of polymer properties. The properties that were predicted were: the molar volume (V), heat capacity at 298 K (C_p), change in heat capacity at the glass transition temperature (ΔC_p), cohesive energy (E_{coh}), solubility parameter (δ), glass transition temperature (T_g), refractive index (η), thermal conductivity (λ), and the dielectric constant (ϵ). A total of 357 polymers were examined, although sufficient information on all 9 of the properties was not available for all of the polymers. An adequate set of complete data could be constructed for the purpose of neural network training and predictions. Table I summarizes the errors obtained from the prediction of the properties listed above for twenty unknown polymers (test set).

Table I. Physical property estimates by ORNLNET.

Property	Range	std. dev.	c. coef.
V (cc/mol)	24-214.3	0.5	0.9999
C_p(J/mol k)	38.3-235.9	1.8	0.9992
ΔC_p at T_g	9.4-77.8	1.1	0.9982
E_{coh} (J/mol)	8089-71087	335	0.9996
δ ($J^{1/4}/cm^{3/2}$)	12.7-20.9	1.3	0.9994
T_g (K)	171-482	6	0.9978
η	1.3500-1.5750	0.0045	0.9971
ϵ	2.10-3.25	0.03	0.9971
λ (J/K m s)	0.110-.230	0.019	0.9970

The correlation coefficient (c. coef.) and standard deviation (std. dev.) for each property was determined for the neural network predictions. In each case, the neural network was able to account for more than 99.4 % of the variation of the data for the given properties. The standard deviation was at most 11.8 % of the average value of the property (the worst was for the thermal conductivity). The average prediction error was < 1% with a maximum error of only 8.2 %. Overall, the neural network clearly demonstrated the capability of making accurate predictions of at least 9 polymer properties from a set of 18 descriptors.

Extensions to more properties such as oxygen permeabilities at room temperature, or temperature dependencies of the various properties should be relatively straight forward. Predictions of mechanical properties such as shear and bulk moduli, tensile, and impact strength have also proven to be accurately predicted by the neural network technique.

The generality of this approach was also tested on other types of materials, such as hydrocarbons (including their isomeric forms gave an average prediction error for 6 physical properties of < 3%), fluorohydrocarbon compounds (average prediction error for 5 physical properties of < 6%), and energetic materials (predictions on the detonation velocity and sensitivity of energetic materials gave an average prediction error of < 3%). In all cases, the accuracy of the predicted properties was similar to that discussed above.

Reverse Process: Designing Molecular Structures that have Desired Properties

The prediction of properties based on chemical structure has numerous possible applications, however, the real potential is in the reverse problem: predicting the structure of compounds that give a set of specified properties or meet certain performance criteria. Simply inverting the neural network predictions is not possible since the relationships are ambiguous. The approach that we have employed is to use a combinatorial optimization technique, genetic algorithms [18-20], to search through the numerous possible structures that could give a set of specified performance criteria (the neural network is used as the function to optimize). It has been found that a genetic algorithm (using real strings and special operations) is an efficient technique for generation and selection of chemical structures. The overall procedure is to use a trained neural network-genetic algorithm hybrid to determine possible structures that satisfy desired performance criteria. As an example of the capabilities of this computational scheme, we have used this method to design a number of fluorohydrocarbon compounds that had the desired properties of Freons but without the environmentally unsatisfying ozone depletion potential and atmospheric lifetime. In addition we have used the method to design energetic materials that had optimal detonation velocities.

For the flurohydrocarbon problem, several compounds were designed that had no experimental data available: ($CH_3CF_2CH_2F$, $CF_2HCHFCHF_2$, $CH_2FCF_2CFH_2$). All of these compounds can be synthesized and measurements of at least one of the predicted compounds confirm the prediction accuracy (the maximum error was 6%) of the overall method.

Similar results have been obtained for designing energetic materials. In this case, the method designed compounds, $C_6H_6N_6O_6$ triaminotrinitrobenzene and $C_4H_8N_8O_8$ cyclotetramethylene tetranitramine, that already was known to have optimal properties. In any case, the fact that the method was able to determine these candidate compounds is very exciting and demonstrates the potential applicability to a number of problems in materials design.

Conclusions

In this paper, *computational synthesis*, a method for predicting the properties or for designing materials that have desired properties, was discussed. It entails a computational paradigm for making rapid and accurate estimations of physical properties for molecular systems. The method uses a set of descriptors as a numerical representation of the chemical structure for a given compound and relates these to a set of properties by utilizing a computational neural network. The neural network is capable of efficiently formulating all of the correlations necessary to make accurate predictions. Results have been obtained for up to 10 properties of 357 different polymers (average prediction error of $< 1\%$), for making accurate predictions on the detonation velocity and sensitivity of energetic materials (average prediction error of $< 3\%$), and for predicting properties of a series of isomeric forms of saturated hydrocarbons (average prediction error of $< 3\%$) and fluorohydrocarbon compounds (average prediction error of $< 6\%$). In comparison with other techniques for obtaining quantitative structure-property relationships (QSPR), the neural network method offers advantages in both ease of use and accuracy, features which are essential for use in technological applications. The method can also be operated in the reverse direction (with the aid of genetic algorithms), that is, designing molecular-based compounds that have desired properties.

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