NEURAL-NETWORK BASED EXPLORATION IN THE DISCOVERY AND DESIGN OF MATERIALS

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Abstract: In previous work we reported on materials design and property estimation using empirical or ordinal representation approaches (Pao et al., 1999, Pao et al., 2000). In this paper, we report in detail on a Basic Concepts approach, which combines some of the merits of both the *ab initio* quantum theoretic first principles approach and the commonly used empirical approach. We show that it is possible to establish mappings between sets of basic atomic characteristics and properties of materials systems formed from those constituent atomic species. Such mappings are implemented in neural networks and are trained using large bodies of high-quality well-filtered data. We also report on the adaptive extrapolation and exploration of such neural network mappings. Such exploration can be used to suggest likely compositions of new compounds that would have certain required properties. This is accomplished by a combined technique of neural networks and evolutionary search. The feasibility and the effectiveness of this method are illustrated with examples. *Copyright 2000 IFAC.*

Keywords: Materials Design, Materials Discovery, Basic Concepts, Neural-Networks, Evolutionary Search.

1.INTRODUCTION

When given the need for a material with certain specified properties, it should be possible to identify some candidate materials compositions and fabrication procedure computationally with a high degree of accuracy. That scenario is not far fetched. It would be the natural extension of present day practice in the use of handbooks, or extrapolation based on existing knowledge of associations in data and experiences.

This paper is concerned with that matter. The expectation is that even early advances in computational materials science in this area would be of significant help in the systematic discovery and design of new materials required to fill some need.

Non-relativistic quantum mechanics is a mature theory and in principle it is possible to start with a description of a materials system at the level of nuclei and electrons, and computationally estimate certain chemical-physical properties of that system. In practice some of that can indeed be done (Bernholc , 1999, Ordejón et al., 1995, Rohlfing and Louie, 1998), but each such computation is a long and costly endeavor.

Presently, empirical or semi-empirical means are used in the search of likely candidate materials systems. It is bemusing to note that in empirical or semi-empirical search for new materials there is a basic question of what is the space to be searched. In present empirical practice, that space is rather small and specific. That space is usually defined in terms of a few variables representing available data for a set of known materials systems. The task then is to build an associative bridge or mapping between known values of property A and inferred values of property B for the same type of materials systems. That is indeed a practicable procedure and can be successful depending on the circumstances. The methodology has some failings in that a considerable amount of experimental data is required for the establishment of the mapping or association and it is not always evident whether the data are relevant or whether the mapping would be valid for new materials; or indeed whether the new desired material is located in that input space circumscribed in such arbitrary manner.

This present paper proposes and describes instead a procedure where the input space is specified in

terms of 'Basic Concepts', features which are so fundamental that one could carry out an ab initio first principles computation of the properties of interest, starting with those 'Basic Concepts' values. Candidate material systems are points in that space and the task is that of establishing mappings between that space and the property values of interest.

Specifically, in materials discovery and design the activities of importance are those of exploration and prediction. One identifies candidate materials systems in 'input space' and predicts what the property values will be in 'output space'. To be able to do this one needs to synthesize an accurate mapping from input space to output space. In the Basic Concepts approach experimental data are needed for populating the output space but the input space is essentially invariant with regards to changes in output space.

The Basic Concepts approach is described in skeletal form in Section 2 of this paper.
Several demonstrations of applications of this procedure are described in Section 3. In one case we use this procedure to estimate whether binary, ternary or quaternary sets of elements would form compounds or not. The prediction results are 95.4%, 98.1% and 99.7% correct respectively, of accuracy high enough to be interesting and possibly of practical value.

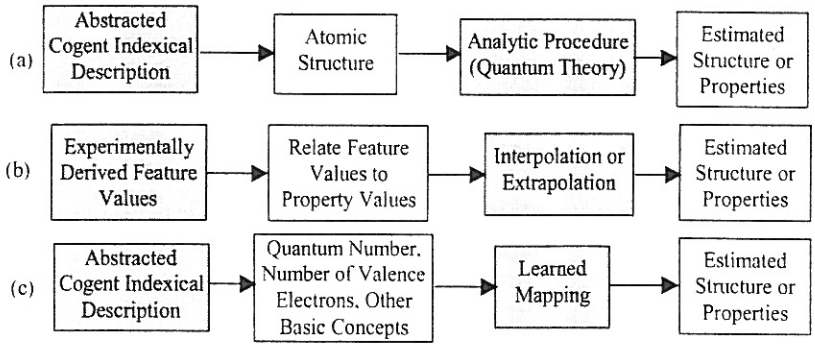
In our work we have also used this approach to predictively estimate the values of certain properties of binary compounds. We present the results for the estimate of melting points in Section 3. Similar results were obtained for other properties.

In Section 4, we discuss in further detail issues of representation, adequacy of training data and accuracy of estimated results. The discussions are illustrated with an account of how a mapping can be improved adaptively until it becomes adequate for identifying candidate materials compositions with property values initially out of the range of previously experienced values. That adaptation can be "boot-strapped" successively.

2.THE BASIC CONCEPT APPROACH TO MATERIALS DISCOVERY AND DESIGN

Three different approaches to information processing in materials research are illustrated schematically in Figure 1. These approaches are the *ab initio* first principles approach, a generic empirical approach and the presently discussed Basic Concepts approach. The context is primarily that of the predictive estimation of property values of materials, for the purposes of discovery and design of new materials with interesting properties or with specific properties needed for some application.

In the interest of brevity, we illustrate the similarities and differences with the sketch exhibited in Figure 1. Emphasis is on the fact that in both the first principles and the Basic Concepts approaches, the 'input' delineation of the materials system is so basic that the same representation can be used for many different types of tasks and no new sets of experimental data are required on changing from one property to another.



(a) The *Ab Initio* First Principles Approach
(b) The Empirical Approach
(c) The Basic Concepts Approach

Fig. 1. Different Approaches for Materials Research

NVE	1	2	3	4	5	6	7	8	9	10	11	12	3	4	5	6	7	8
GN	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18
QN																		
1	H																	He
2	Li	Be											B	C	N	O	F	Ne
3	Na	Mg											Al	Si	P	S	Cl	Ar
4	K	Ca	Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	Ga	Ge	As	Se	Br	Kr
5	Rb	Sr	Y	Zr	Nb	Mo	Tc	Ru	Rh	Pd	Ag	Cd	In	Sn	Sb	Te	I	Xe
6	Cs	Ba	A*	Hf	Ta	W	Re	Os	Ir	Pt	Au	Hg	Tl	Pb	Bi	Po	At	Rn
7	Fr	Ra	B*	Rf	Db	Sg												
			A*	La	Ce	Pr	Nd	Pm	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu
			B*	Ac	Th	Pa	U	Np	Pu	Am	Cm	Bk	Cf	Es	Fm	Md	No	Lr

Fig. 2. Periodic Table of Elements

In the present case we characterize an element in terms of two numbers, one being the principal quantum number of the valence shell (QN) and the other the number of valence electrons (NVE). The QN and NVE of the elements are shown in Figure 2. However, there exist some overlaps in the values of NVE, which make some elements indistinguishable. In order to give each element a unique representation, except for the Lanthanides and the Actinides which have similar properties, the group number (GN) is used instead of NVE. GN is closely related to NVE in the following way:

Block	NVE
s, *d*	GN
p	GN-10

Such replacement is for computational purposes only. The purpose of using this feature is still to specify the valence electrons in an ordinal manner. The group number distinguishes the valence electrons filling the *d* orbits from those filling the *p* orbits.

We note that the 'Basic Concepts' are not necessarily limited to the quantum number and the group number. Other concepts that are equally basic can be incorporated when needed. Some examples are discussed in Section 4 of the paper and in reference (Zhao, 2000). But all such 'Basic Concepts' remain basic in the original sense. They do not change from task to task.

Prediction is made possible through use of neural networks. The input to the neural network is the Basic Concepts description (QN and GN in this case). The output is the value of the property of interest. The mapping between the inputs and the outputs is 'learned' using large bodies of high-quality well-filtered data. Detailed implementation of the approach is available in reference(Zhao, 2000), including comments on measures for differentiating between elements in the Actinide and Lanthanide series.

3.APPLICATIONS TO PREDICTION OF CATEGORY MEMBERSHIP AND OF PROPERTIES

In our work, as a precautionary check, we applied this methodology to the prediction of some properties of the elemental species such as the first ionization potential, the atomic radii and so on. Neural networks were used to learn a model of the mapping relationships between the Basic Concepts representation of elemental species and the exemplar values. The model was then used to provide estimates of other elemental species. The prediction results of the first ionization potential of some elemental species are shown Figure 3. The estimates agree well with the experimental data.

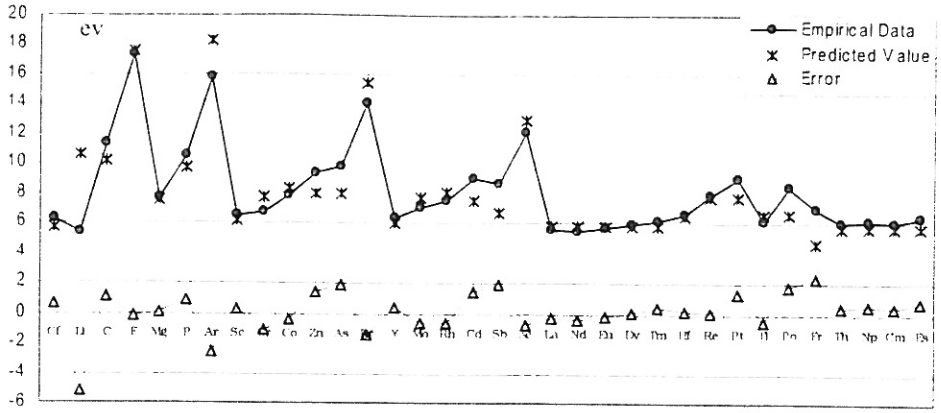


Fig. 3. Predictive Estimates of First Ionization Potential for Elemental Species

	# of Training Data	# of Generalization Data	# of Test Data	Prediction Accuracy
Binary Systems	627	706	692	95.4%
Ternary Systems	804	1352	4278	98.1%
Quaternary Systems	833	1699	4959	99.7%

Fig. 4. Prediction Results of Compound-Formation Tendency

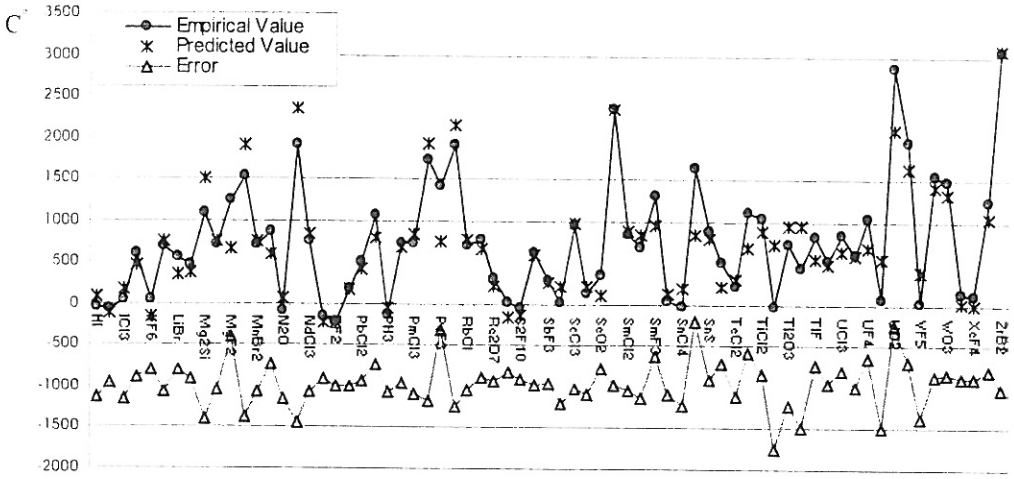


Fig. 5. Prediction Results of Melting Points of Binary Compounds

In a next step, we used the procedure to predict the compound-formation tendency of binary, ternary and quaternary materials systems. In that exercise, neural networks were used for learning mappings between the Basic Concepts representation of multinary materials systems and the category membership of compound-formation. The predicted estimates of compound-formation tendency were of good accuracy. A summary of the results is contained in Figure 4.

As test cases, we set out to estimate the density and the melting points of various binary compounds. Mappings were learned between the Basic Concepts representation of binary compounds and the property values of density or melting points. The results of melting point prediction for some binary compounds are shown in Figure 5. A bias of −1000 was added to the error in order to exhibit it clearly in the picture. The results are inline with the experimental data and would be useful for some circumstances.

4.ISSUES OF REPRESENTATION AND ACCURACY

Although the theme of Basic Concepts is simple and essentially correct, there are details which need to be addressed in practice. These are discussed briefly in this section.

First of all, not all the Basic Concepts features are of equal weight. This is because the absolute

magnitudes of ordinary specified feature values maybe valid in a relative sense in intra-feature manner but not necessarily in inter-feature manner. A change in the value of one feature need not have the same effect on output values as that produced by a corresponding change in another feature. The issue is that of ensuring appropriate inter-feature scaling. In practice a near optimal relative weighting can be established through optimization of predicted elemental properties. In our work the relative weighting is 1 to 0.22. Namely, GN was given a weight of 1 while QN was only given 0.22. Secondly, there is a question of stochiometry. One could either treat a binary system A_2B as a pseudo ternary system A_A_B or use the stochiometry as additional weights applied to the input features. We decided that a rational approach would be to use stochiometry as additional input 'Basic Concept', an additional feature. But this third feature also had to be scaled, by a factor of 0.1 in our work. This is a matter of augmenting the representation and making sure that the additional feature is also scaled properly.

Finally, there is the issue of prediction accuracy in cases where either input is out of domain or output is out of range. Such cases usually happen when the training data are confined to certain regions of input space, or are distributed sparsely. Neural networks learned from such data are not adequate for prediction in the entire input space of interest. However, neural network models can be adapted in a successive manner to improve the prediction accuracy. As shown in Figure 6, the estimation error of melting point (M_t) for binary compound Ta_2C (M_t=3880C°) is unacceptably large initially in step 1, using a model trained with 215 experimental data points, with property values in the range from −208.5C° to 2525C°. The error decreased quickly when more exemplars with high melting points (2714C° ~ 3390C°) were added to the training data. Steps 2-5 in Figure 6 show the results of prediction after addition of one more exemplar. It is remarkable that with only 4 patterns added to provide information about the high melting point compounds, the prediction accuracy with the adapted model was improved dramatically. In this case, even though the M_t value to be predicted is still out of the range of previously encountered exemplars, the adapted neural net prediction is approximately 92% accurate. The point of this discussion is to suggest that this practice can indicate what new materials might be synthesized. The neural network model incorporating the additional information would yield predictions of enhanced accuracy.

5.SEARCH FOR COMPOUNDS WITH CERTAIN REQUIRED PROPERTIES

The Basic Concepts approach not only can be used for predicting property values given specific compound compositions, but also can be used in the search for likely compound compositions with predefined property values. The later is actually an inverse problem of the former, and can be accomplished by a combined technique of guided stochastic search and neural network mapping.

The combined procedure of guided search and neural network estimation is illustrated in Figure 7.

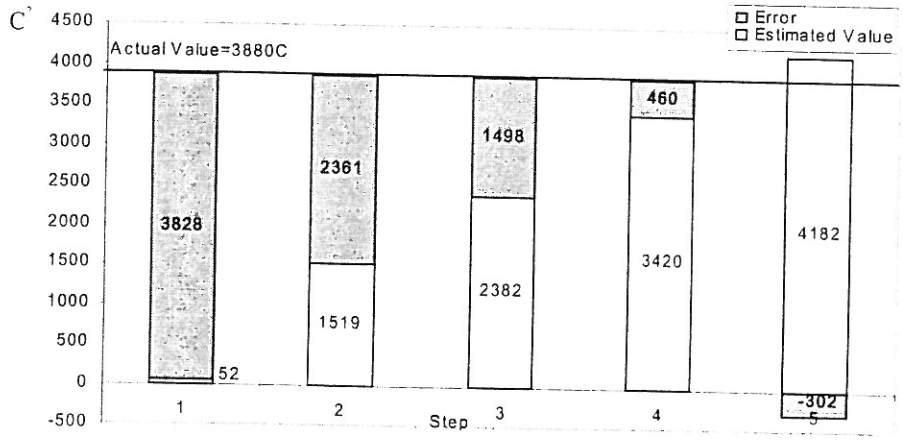


Fig. 6. Estimation Results for Ta_2C with Adapted Model

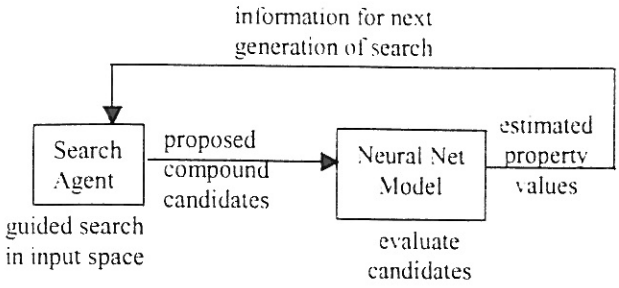


Fig. 7. Guided Search with Neural Network for Compound Composition

The search agent in Figure 7 is implemented using the GESA algorithm (Yip and Pao, 1995), which controls the search resource allocation and the region of search in the input space. It provides the neural network model with a set of candidates, and the model evaluates their property values. The estimated property values are compared with the desired value and are sent back to the search agent as information for the next generation of search. Locations with better performances in the current generation are assigned more resources for searching in the next generation

For example, if the objective is to find the compound composition with a density value of 13.9, the above procedure found a binary compound TaC_2 with estimated density value of 13.3. Actually it is known that compound TaC has a density of 13.9, and the result obtained is very close to it! In practice there are occasions where several compounds have property values close to each other. In that situation, different solutions might be obtained depending on the start point of the search. In addition, noise in the neural net model might prevent a candidate being identified even if its real value is exactly that of the required value.

6.SUMMARY

This paper reports on research directed towards developing a computational approach to materials design and discovery. A Basic Concepts approach is described in this paper. Predictions of compound formation tendency and of compound properties are investigated and the results are encouraging. The main advantage of the approach lies in that the details of the interactions of the constituent elemental species are captured automatically in the neural net mapping during the learning process, even if the 'Basic Concepts' only describe the elemental atomic constituent species individually. The approach can also lessen the burden of collecting qualified data and is computationally efficient.

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