

DESIGN OF FUEL-ADDITIVES USING HYBRID NEURAL NETWORKS AND EVOLUTIONARY ALGORITHMS

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Abstract

Fuel-additives play an important role in deposit reduction on the valves and combustion chamber of the automobile. They reduce cold-start problems, emissions and improve fuel-efficiency. The testing and design of fuel-additives is an expensive and lengthy procedure. A hybrid first-principles neural-network model for fuel-additive performance prediction was developed in this effort. This model predicts additive performance directly from the structure of the additive and outperformed existing models based on basic structural descriptors. The design of fuel-additive structures was accomplished using an evolutionary algorithm with problem specific representation and genetic operators. Some results from the model and the design algorithm are discussed in this paper.

Keywords

Computer-aided design of materials, Fuel-additives, Hybrid phenomenological neural-network models, Evolutionary algorithms

Introduction

An additive is a substance added to another in small quantities to improve current performance or provide for a desired effect. Additives are added to gasoline to provide improved performance or to correct deficiencies. Gasoline additives have been historically used as combustion modifiers, anti-oxidants, corrosion inhibitors, anti-icing components as well as deposit control detergents (Gibbs, 1990). The primary focus of this paper is on the class of fuel-additives that act as deposit controllers in the automobile. Specifically, this effort is on fuel-additives that control the deposit formation on the intake-valves of the automobile. Figure 1 shows the schematic of the intake-valve and its surrounding components in an

automobile. The intake-valve forms the opening into the combustion chamber. The fuel-injection nozzles spray gasoline directly on the intake-valve. When the valve opens, it draws in a mixture of fuel and air into the combustion chamber where it is burned to supply power to the automobile. Over a period of time, both during and shortly after engine operation, deposits tend to form on the surface of the intake-valve (Kalghatgi, 1990; Lacey et al., 1997). These deposits have been documented to affect driveability, cold-start efficiency, knock characteristics and emissions (Grant and Mason, 1992; Graham and Evans, 1992; Houser and Crosby, 1995).

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The US Environmental Protection Agency (1996) has adopted a standard test to determine the deposit forming tendency of fuel package (gasoline + additives) before approving commercialization of the package. This is the ASTM Standard (1995) BMW-IVD test. The test uses a 4-cylinder 1985 BMW vehicle, operated over the road for a total of 16,093 km. The daily test cycle consists of 10% city, 20% suburban and 70% highway mileage with an overall average speed of 45 mph. A fuel package must produce an average deposit of less than 100mg/valve for certification (Lacey et al., 1997). This is the performance measure of a fuel-additive.

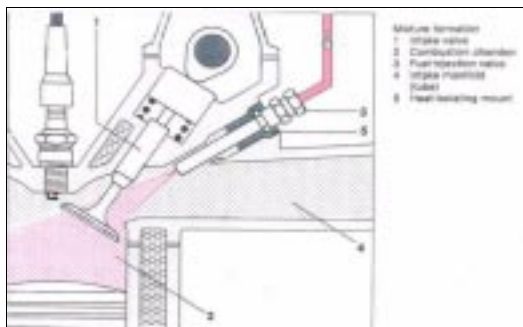


Figure 2 Intake-Valve and manifold

The basis of any product design problem is the forward problem that involves the prediction of product performance from its components (Venkatasubramanian et al., 1995). The main function of the fuel-additive is the prevention of deposition on the valve by scavenging the deposit forming pre-cursors of the fuel. The mechanism of intake-valve deposit (IVD) formation is a complex process with a variety of influential factors such as additive and fuel chemistry, operating conditions and flow properties of the additive, fuel and oil. Most of these parameters are not measured. This makes a purely first-principles model difficult if not impossible to determine. The large noise in the typical BMW test data (Arters et al., 1997) makes a purely statistical approach inaccurate and physically unreliable. A hybrid approach that combines the best possible phenomenological description of fuel-additive performance with the non-linear capabilities of neural-networks is proposed for additive performance prediction.

Hybrid Model for Fuel-Additive Performance Prediction

The hybrid model was developed along the following major steps

- (i) Determination of a functional description of the fuel-additive in terms of key performance imparting components in the structure.
- (ii) Development of a kinetic model that determines quantitatively the dynamic degradation of the functional components under given operating conditions.

(iii) Prediction of the time dependent "activity" of the additive in the fuel, in terms of its ability to sustain its deposit removal abilities.

(iv) Prediction of the IVD performance of the additive using a neural-network model to correlate the "amount of active additive" at different points in time to the experimental engine test data.

The main functional components of the fuel-additive structure were: (a) a polar core component that reactively bound to the deposit forming pre-cursors of the fuel (b) several branches of functional groups that enhanced the deposit removal activity of the additive (c) a transitional component that lended stability to the structure and held the core to the branches of the additive. A kinetic model for the degradation of the different components in the additive was developed and is given by Equation (1).

$$d\mathbf{X}/d\tau = \mathbf{K} \cdot \mathbf{X} \quad (1)$$

Here, \mathbf{K} is the matrix of rate constants describing the first-order irreversible reactions for additive degradation and \mathbf{X} is the vector of concentrations of additives at different levels of degradation. Using this model, the distribution of the structure of the additive was determined as a function of time. A group contribution method was employed to determine the cohesive energy density of the additive in the fuel from its structure and the given set of fuel characteristics. In combination with the degradative model, this gave a time varying, quantitative description of the activity of the additive in the fuel. This information was then used as input to a neural-network model whose output was the IVD performance of the given additive in the given fuel. This model is shown in Figure 2.

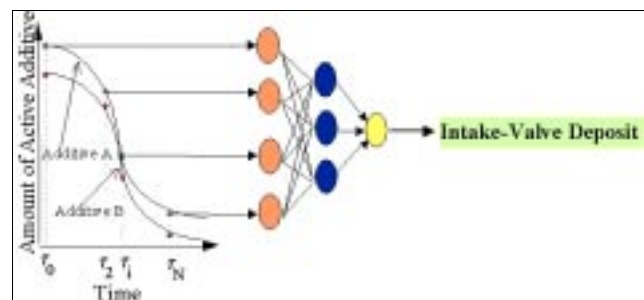


Figure 2 Hybrid model for IVD prediction

This hybrid model was applied to the BMW engine test database provided by the Lubrizol Corporation. The data consisted of 92 engine-test results along with the structures of the additives and the fuel characteristics used in each test. The results from different models are compared in Figure 3. The phenomenological model for IVD prediction used just a single descriptor (the activity of the additive at time = 0) and outperformed all other multivariate statistical models that were based on only the structural features of the additives. These descriptors for example, included the counts of different functional groups. This difference in

performance was clearly due to the mechanistic description of additive degradation and its interaction with the fuel characteristics, included in the phenomenological model.

Design of High-performance Fuel Additives

The hybrid model predicted the IVD performance of a fuel-additive given its structure and operating conditions such as the fuel characteristics and temperature. The design problem involves the construction of optimal fuel-additive molecules given desired IVD requirements. This is a non-linear combinatorial optimization problem involving the use of a black-box objective function (due to the use of the NN). Widely used techniques such as knowledge-based enumeration (Gani and Fredenslund, 1993), mathematical programming (Maranas, 1996) and graph theoretic approaches (Skvortsova et al., 1993) will not work for this scenario, because of the above problem.

No. of Input Descriptors	Projections (PLS/PCA/None)	NN Architecture Transfer Function [No of Hidden Neurons]	RMSE (mg) in Testing (CV)
36	None	Tan-Sigmoid [7]	273 [*]
4	PLS	Log-Sigmoid [3]	105 [†]
3 [†]	None	Tan-Sigmoid [3]	142
7	PCA	Tan-Sigmoid [3]	200
6 ^{††}	None	Tan-Sigmoid [3]	172
1 ^{**}	None	Radial Basis [4]	124

*Large Standard Deviation; [†]Descriptors extracted from PLS factors

^{††}Descriptors extracted from PCA factors

**Solubility Descriptor extracted at time $\tau = 0$

Figure 3 Model Comparison for IVD prediction

In this approach genetic algorithms(GAs) (Holland, 1975;Goldberg, 1989) which are search procedures based on Darwin's evolutionary model were adapted to the fuel-additive domain. Previous work in computer-aided molecular design has demonstrated GAs to be flexible in capturing the rich underlying chemistry (Venkatasubramanian et al., 1996). Moreover, they are robust to nonlinearities and hence powerful procedures for global search. The strategies for the forward and inverse phases for fuel-additives design are shown in Figure 4. The following were the main components of the evolutionary algorithm.

Representation: A fuel-additive was primarily composed of a group of functional components described previously. Differences between additive structures stemmed from differences in the chemical composition (in terms of the functional groups) and the topology of the functional components. Both of these affected the activity of the additive. Hence an additive was represented in an object-based fashion, containing the functional components themselves as the underlying objects. The representation also included information on their connectivity.

Genetic Operators: There were rules concerning the connectivity and topology of the fuel-additive structure. These were chemical valence or connectivity constraints

on specific components that preclude chemical and synthetic infeasibilities in the additive molecule. Moreover, every additive consisted of at least one of all three functional components in it. From a search perspective, genetic operators defined the moves that navigate the space of solutions that consisted of feasible fuel-additive molecules.

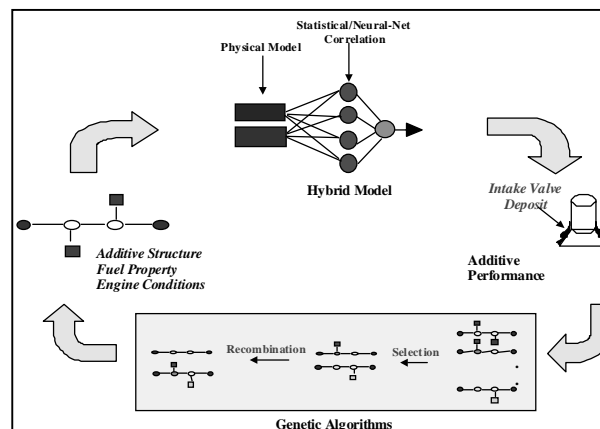


Figure 4 Designing for optimal fuel-additives

Crossover and mutation are the typical genetic operators for evolutionary algorithms. Crossover involves large scale but random exchange of information across two members of the current set of solution candidates. Mutation involves a random but small change to a solution candidate. To avoid the creation of infeasible fuel-additive structures from feasible ones, these operators were customized to obey rules of feasibility. Four different constrained operators were employed in both crossover and mutation depending upon the functional component to which it was applied. A feasible mutation operation in the transitional component that connects the core of the additive to the branches is depicted in Figure 5. In this operation, the components connected to the mutated region were re-arranged into a feasible configuration such that none of the feasibility constraints were violated. Other operators were implemented in a similar fashion.

Initialization and evolution: The algorithm was initialized with a single structure that was randomly but feasibly constructed from a given set of functional components. A desired IVD objective was set as the goal. The initial population consisted of members that were copies of this structure. Each structure was evaluated using the hybrid model to determine its IVD performance and assigned a fitness value between zero and one quantifying its degree of desirability. Parents were chosen from the population based on their fitness and allowed to undergo crossover and mutation operations stochastically and according to a pre-determined frequency. The resulting offspring formed the next generation and the process of fitness evaluation, selection and recombination was continued. The evolutionary algorithm was employed to search a space of

solutions that was roughly about a million in combinatorial size. This was used as a case study to examine the efficiency of the algorithm. With function evaluations at a premium, this efficiency was crucial. The evolutionary algorithm stepped through 25 generations each containing a pool of 25 fuel-additive structures. A crossover frequency of 0.60 and a mutation frequency of 0.40 were used. The objective was to design fuel-additive structures that, based on the hybrid forward model, were predicted to accumulate of less than 10 mg of IVD on a standard engine test.

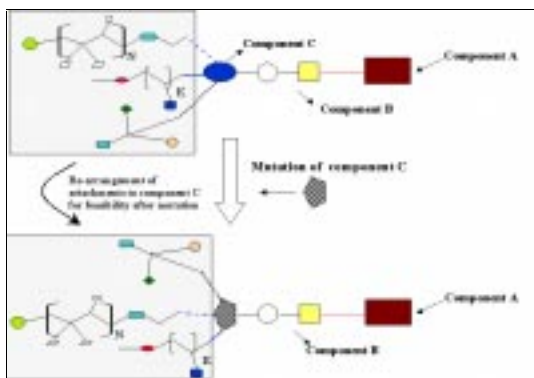


Figure 5 Mutation of a transitional component

The neural-network model was trained from a set of HONDA generator engine tests with a predicted RMSE error on cross-validation of 30 mg. This was quite close to the quality of the engine tests whose repeats differed on an average of about 24mgs. The fitness function used is given by Equation (2)

$$F = 1.0; \text{IVD}_{\text{predicted}} < \text{IVD}_{\text{limit}}$$

$$F = \exp(-\alpha \{ (\text{IVD}_{\text{predicted}} - \text{IVD}_{\text{limit}}) / \text{IVD}_{\text{limit}} \}^2); \text{ otherwise} \quad (2)$$

All additive structures were assumed to be present at the same dosage initially. The results of three different runs of the evolutionary algorithm are summarized in Figure 6.

Run	Rank/Identifier	Fitness	Predicted IVD (Hybrid-NN Model)	Structural Description
I	1, I-1	0.997	11.4 mg	Novel Structure. Rarely used components
	2, I-2	0.996	11.5 mg	Novel Structure. Similar to best structure, different core
	6, I-6	0.993	12.8 mg	Variant of structure found in the SIVAF database
II	1, II-1	0.999	90.1 mg	Novel Structure. Different from I-1. Infrequently used transitional component.
	2, II-2	0.998	12.6 mg	Slight variant of additive structure found in SIVAF and HONDA databases
	4, II-4	0.993	13.2 mg	Minor variation of structure II-2 above. Slight modification of the core
III	1, III-1	1.08	8.9 mg	Novel Structure. Different from I-1 and II-2. Commonly used components
	2, III-2	0.994	11.9 mg	Variant of III-1. One of the transition and branch components different
	3, III-3	0.993	12.1 mg	Variant of structure III-2 above. Slight modification of core. Contains an additional branch

Figure 6 Results from three different runs of the evolutionary algorithm.

The evolutionary method was successful in constructing several known good candidates as well as novel ones. The fitness and predicted IVD performance of the different additive structures are shown in Figure 6. In the figure, a structure is recognized as novel when all the functional components comprising it had never occurred in combination in any of the databases used in training. Structures, some of whose components had been examined in combination in the databases were characterized as variants of known structures. Even with a small sampling of the search space (~625 out of a one million), the evolutionary algorithm was successful in identifying diverse structures that met or were close to meeting the set objectives. Indeed, most of the best structures found in each of the runs were never encountered before. One fuel-additive molecule (III-1) in the Figure 6, comprised completely of frequently used components but never before tried in combination. This was not only a novel structure but was also seen to possess good synthesis potential.

Conclusions

A new hybrid first-principles neural network framework for fuel-additive performance prediction was developed and demonstrated to perform accurately even with limited and noisy data. Evolutionary algorithms with customized constrained operators were developed for the inverse design problem and proved to be successful in identifying novel, optimal candidates that also possess characteristics desirable in synthesis.

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