

## **A Systematic Approach for Synthesis of Optimal Polymer Films for Radioactive Decontamination and Waste Reduction**

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### **Abstract**

In this work, an effective and systematic modelling technique is devised to generate optimal formulations for explicit polymer film engineering applications. These methods aim at developing quantitative values for not only intrinsic properties, but qualitative characteristics are developed in order to simultaneously optimize the formulation subject to the product specifications. The predictive modelling framework developed is comprised of material and energy balances, constraint parameters, constitutive equations, design/optimization variables and possible polymer synthesis techniques. A set of user defined design constraints produces a subset of different optimization formulations comprised of different polymer blends, molecular weights, hydrolyzation extents, solvents, and additives. This contribution illustrates a novel way to evaluate a wide range of polymeric film compounds and mixtures with fewer testing iterations.

**Keywords:** Formulation synthesis, material development

### **1. Introduction**

Formulation of new products and improvement of existing merchandise is practiced in many different industries including paints and dyes, polymers and plastics, foods, personal care, detergents, pharmaceuticals and specialty chemical development. Current trends in the engineering design community have moved towards the development of quantitative integrated solution strategies for simultaneous consideration of multiple product characteristics. The optimization variables are most often determined by qualitative attributes, stochastic variables, visual observations and/or design experience. The effectiveness of these approaches is limited by available data, bias towards specific solutions, reproducibility, and experimental error. Model insight is required for development of fast, reliable and systematic screening methods capable of identifying optimal formulations and reducing the number of subsequent laboratory trials.

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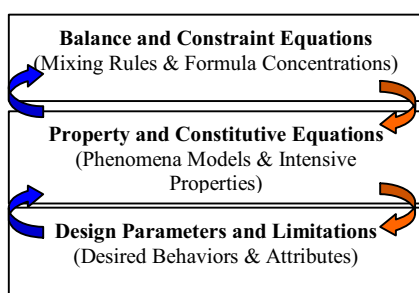
## 2. Model Development Methodology

In order for the product to exhibit the desired performance, a combination of discrete constraints must be fulfilled. Identification of an optimal formulation that is suitable for the desired system requires integration of all the interlacing behaviours of the product constituents. These characteristics include the constituents used for construction as well as their inherent properties. This is accomplished by using a combination of novel modelling techniques.

### 2.1 Property integration

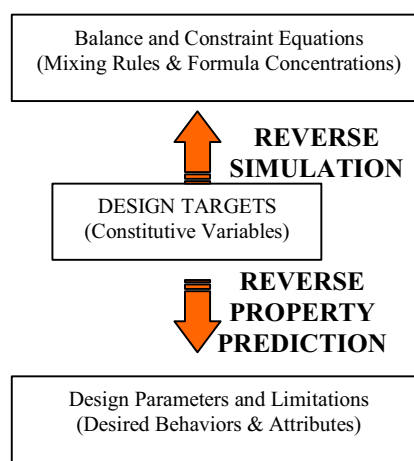
This method consists of tracking functionalities instead of chemical species in order to represent the synthesis problem from a properties perspective. The conventional approach to product formulation development is selecting constituents that exhibit desired produced properties and optimizing the mixing ratios. In order to model the product characteristics, these pre-determined candidate components are required as inputs to the design algorithm. These inputs are based on qualitative process knowledge and/or design experience, which can exclude solutions involving other possible raw material sources. In this work, the concept of property integration for process development introduced by El-Halwagi et al. (2004) is applied to product synthesis. This modelling approach allows for solution of many different engineering problems to be conducted on a property only basis. This method allows for identifying optimized solutions to specified chemical engineering problems by determining the desired output and solving backwards for the constituents and compositions. In the case study to be described in the next section, these techniques are used in conjunction with a model decomposition technique to allow for formulation of reverse property prediction problems.

#### Conventional Model Structure



(a)

#### Decoupled Model Structure



(b)

Figure 1. Decoupling of constitutive equations for reverse problem formulation

## **2.2 Model decomposition and reverse property prediction**

These modelling techniques are useful tools to reduce the complexity involved when trying to simultaneously optimize balance and constraint equations with constitutive and property models. The development of these novel techniques has been described by Eden et al (2004). Although these procedures were created for process development, minor changes allow for application to product design. The main objective of the method is to separate the balance and constraint equations from the often complex and non-linear constitutive property relations. Figure 1 illustrates this decomposition principle by showing how the overall model (a) is divided into two separate models by defining target property variables (b). These target variables are a set of solutions to both the formula balance model and the constitutive property design model. Each mathematical system is solved independent of the other until valid sets of solutions are found that satisfies both networks. This technique is illustrated in the following case study.

## **3. Case Study – Polymer Film for Nuclear Applications**

The desire to decontaminate surfaces inside nuclear power plants has been addressed with a number of different products. The implementation of latex-based peelable films has been used for many years. The coating serves to initially “fix” the contaminants in place for containment and ultimate removal. However, power plants have discontinued the use of these products because of their long drying times and expensive disposal costs. In the place of these products, protocol has turned to the use of steam jets to remove the radioactive particles and clean the exposed surface. This method has proven to be ineffective due to a build up of contaminants that, through molecular transport, become airborne and contaminate larger areas. The purpose of this work is to develop an effective and systematic model to synthesize a formulation of a water soluble polymer film coating for radioactive decontamination and waste reduction. This material development involves the use of a polymer matrix that is applied to surfaces as part of the decontamination system in place of the past latex products. Upon mechanical entrapment and removal, the polymer coating containing the radioactive isotopes can be dissolved in a solvent processor, where separation of the radioactive metallic particles occurs. Ultimately, only the collection of filtered solids must be disposed of as nuclear waste. The ability to identify such a product creates an attractive alternative to direct land filling or incineration. In order for the polymeric film to be a viable candidate, it must exhibit the desired performance that previous coatings are unable to. These characteristics include, drying time, storage constraints, decontamination ability, removal behaviour, application technique, coating strength and dissolvability processes.

### **3.1 Property integration of polymer coating model**

Identification of an optimized formulation that is suitable for this entire decontamination system requires integration of all the interlacing characteristics of the coating composition that affect the film behaviour. In order to accomplish this, an accurate representation of the system must be developed in order to solve the design parameters in terms of properties only. The representation of the design parameters along with the interactions between them and the overall formula behaviour is given in

figure 2. This model could be solved as a reverse simulation problem using the final coating characteristics as input variables and the final polymer, solvent, and additive selections established as output solutions. The intricacy here is producing an accurate model, as the inherent non-linearity of the property relationships in conjunction with the complex formulation balance equations makes acquiring viable solutions difficult. In order to overcome these obstacles, model decomposition is employed.

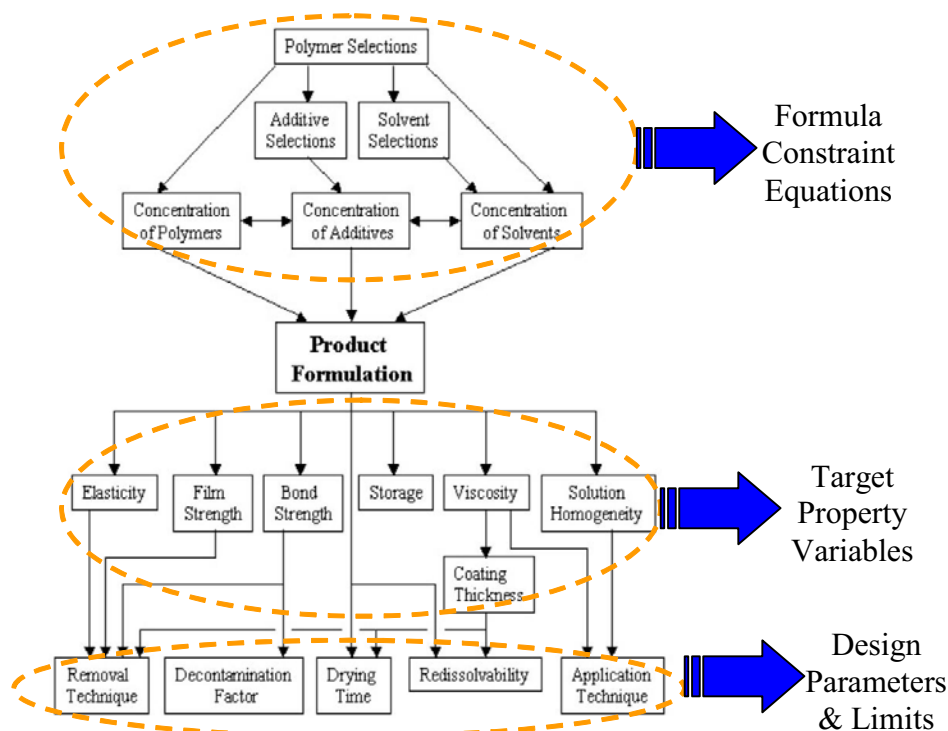


Figure 2. Schematic representation for polymer film design & model decomposition

### 3.2 Decomposition of polymer film design problem

The problem schematic shown in figure 2 is decomposed into three separate parts in order to reduce the complexity of the solution procedure. These subparts are comprised of formula equations, design parameters, and target property values.

#### 3.2.1 Formula constraint equations

The formula balance equations are separated into a reverse simulation problem that includes polymer, additive and solvent choices. Among these selections are available synthesis variables that affect polymeric properties such as molecular weight and extent of hydrolysis. With this information included in the model, not only can different polymer chains be compared, but also different variations of the same polymer and polymer blends. The ability to optimize the polymer synthesis as well as the film composition increases the possible formula combinations and improves the chances of acquiring an acceptable optimized formula. The additive options include components that enhance the desired film properties in order to fulfil the necessary constraints from

the target property variables. This assortment of compounds contains wetting agents, surface tension reducers, biocides, cross-linking agents, elastomers, resin hardeners, dyes, pigments and dispersants. The choice of solvents is limited not only by the polymer selection, but also by the application. The list and amounts of volatile solvents allowed to be used on a nuclear power plant floor is extremely limited. The initial concentration of solvent present in the coating is the primary driving force involved with drying time. It is imperative for this part of the overall model to simultaneously optimize the formulation so that target properties are exhibited and the overall film behaviour is superior to current competitor products.

### *3.2.2 Design parameters*

The design parameters and limitations represent a compilation of attributes that the final product must exhibit. Because this formulation is intended to fulfil a market niche that already exists, the final formula characteristics are well known. The primary design parameters are the decontamination ability, drying time and redissolvability. The ability for the film to remove contaminants is measured by the ratio of radiation detected divided by the radiation present before the film removal. This numeric value is known as the decontamination factor and is a major selling point that must be equivalent or better than other possible decontamination products and processes. Another parameter where the new formulation must outperform the competing processes is drying time. Nuclear power plant outages are very costly and the schedule is optimized to minimize profit losses. By producing an optimized formula with the customer's major objectives in mind increases the marketability of the product and improves possible sales. The issue of redissolvability mostly pertains to the manner in which the film is disposed of. The current operations in nuclear plants involve the use of many different polymer based products that are sent to processing stations for redissolving and filtering. It is desired that the film can be disposed of by utilizing these same processing procedures. Other constraints include a simple and effective means to apply the coating to the walls and surfaces inside the plant as well as removal techniques. The model's main objective is to determine what intrinsic properties govern the desired performance variables and develop a dynamic set of target properties.

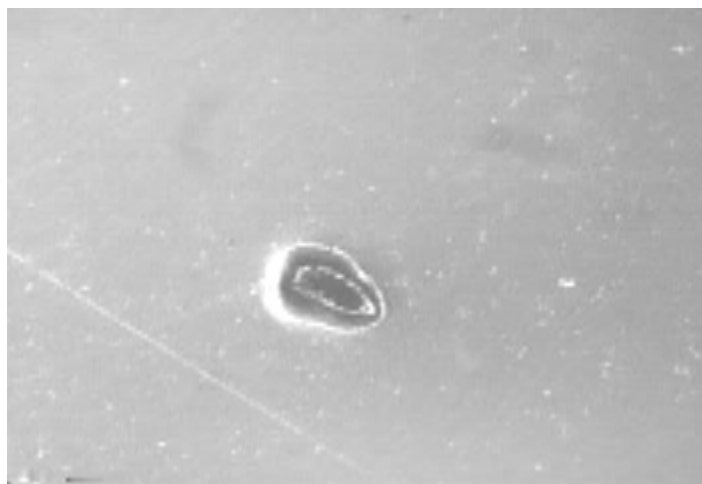
### *3.2.3 Target property variables*

The development of a set of target properties allows this model to utilize reverse property prediction to identify the design alternatives. This is accomplished through experimentation to determine what property ranges equate to final film behaviour. In order to illustrate the modelling techniques presented in section 2, we can simplify the system by assuming that the only major target property in figure 2 is viscosity. This seemingly simple model is decoupled into two separate systems, which is illustrated by figure 1; the chemical makeup equations that produce a given viscosity and the behavioural models which predict how the viscosity affects the design parameters and limitations. By conducting laboratory tests and simulation studies, the optimum fluid viscosity that produces adequate application behaviour can be determined to be a given value, 4000 centipoises for example. This value becomes the viscosity design target of the qualitative prediction model. By implementing the reverse simulation of mixing rules and formula concentration models, a set of viable product formulations that meet the 4000cps design target are determined. These techniques seem unnecessary when

considering only one target property, but when numerous targets are set, these simplification processes are extremely advantageous.

#### 4. Results

The ultimate result of this model aided in the development of a product that increases the removal rate of radioactive contaminants by 69% while attaining a 33% reduction in drying time over the current marketed competitors. The finalized product formula will be available through the Orex Technologies Catalogue in Fall of 2005.



*Figure 4. Carbon particle encapsulated in the final polymer film formula*

#### 5. Conclusions

By employing novel model development techniques such as property integration and model decomposition; a complex product formulation development process has been simplified. In this work, these methods were illustrated by addressing a problematic phenomenon in the nuclear power industry. The utilization of these modelling techniques took an industrial idea to full scale testing and production in under 18 months by reducing the number of subsequent laboratory trials.

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