MODELLING VITRIFIED GLASS VISCOSITY IN A NUCLEAR FUEL REPROCESSING PLANT USING NEURAL NETWORKS

Katy Ferguson[1], Jie Zhang[1], Carl Steele[2], Colin Clarke[3] and Julian Morris[1]
[1]*School of Chemical Engineering and Advanced Materials, Newcastle University, NE1 7RU, Newcastle Upon Tyne, U.K.*
[2]*Sellafield Ltd, Sellafield, Seascale, Cumbria, U.K.*
[3]*National Nuclear Laboratory, Preston, U.K.*

Keywords: Viscosity, Neural networks, Vitrification, Nuclear reprocessing, Modelling, Application.

Abstract This paper presents a study of using neural networks to model the viscosity of simulated vitrified highly active waste over a range of temperatures and compositions. Vitrification is the process of incorporating the highly active liquid waste into the glass by chemically changing the structure of the glass for nuclear fuel reprocessing. A methodology is needed to determine how the viscosity will change as a result of a new feed. Feed forward neural networks are used to model the viscosity of new product glasses. The results are very promising, with a Mean Squared Error (MSE) of 1.8×10^{-4} on the scaled unseen validation data, highlighting the high accuracy of the model. Sensitivity analysis of the developed model provides insight on the impact of composition on viscosity.

1 INTRODUCTION

The wastes from nuclear reactors are radioactive and need to be treated for long term storage. Vitrification is one of the nuclear waste treatment processes, which turns the highly active wastes into a glass which is much safer for storage. The understanding of what affects the nuclear waste glass viscosity has increased due to inactive simulant viscosity tests carried out at the National Nuclear Laboratory. It is essential to understand how the viscosity is affected by the composition using non-active trials.

The experiments provide an insight for plant engineers/operators on what may happen with particular feeds. Only a limited number can be performed due to cost. It is desirable that a model can be built to predict the viscosity saving the company a large amount of investment

Developing a detail mechanistic model has been shown to be difficult, time consuming and effort demanding. The full mechanism of how various compositions affect viscosity is not fully understood. Data based empirical modelling can be a very useful alternative in this case. Neural networks have been shown to be capable of approximating any continuous nonlinear functions (Cybenko, 1989); (Girosi and Poggio, 1990); (Park and Sandberg, 1991) and have been applied to nonlinear process modelling (Bhat and McAvoy, 1990); (Bulsari, 1995); (Duchesne et al., 2010); (Narendra and Parthasarathy, 1990); (Zhang et al., 1998).

This paper presents a study on using neural networks to model the viscosity of simulated vitrified highly active waste over a range of temperatures and compositions. Section 2 gives a brief description of the processes. Neural network modelling of glass viscosity is presented in Section 3. The obtained results are discussed in Section 4 and conclusions given in Section 5.

2 PROCESS DESCRIPTIONS

2.1 Waste Vitrification Plant

The Vitrification plant (Figure 1) is responsible for converting Highly Active Liquor (HAL) into a durable product glass. HAL, consisting of waste fission products in nitric acid, and other calcination additives are fed into a hot revolving furnace. The liquid is evaporated leaving dry particles.

The particles are dropped into a melter with Borosilicate glass frit and heated to high temperatures resulting in a new glass matrix being

Ferguson K., Zhang J., Steele C., Clarke C. and Morris J..
MODELLING VITRIFIED GLASS VISCOSITY IN A NUCLEAR FUEL REPROCESSING PLANT USING NEURAL NETWORKS.
DOI: 10.5220/0003654003220325
In *Proceedings of the International Conference on Neural Computation Theory and Applications* (NCTA-2011), pages 322-325
ISBN: 978-989-8425-84-3
Copyright © 2011 SCITEPRESS (Science and Technology Publications, Lda.)

formed, different from waste encapsulating. The glass is poured into a container. The container is then cooled, lid welded on, decontaminated, monitored then taken for long term storage.

2.2 Inactive Laboratory Experiments

Experiments were carried out in order to test many properties of the glass (Steele et al., 2011.) One property of the glass produced is its viscosity, relating to the pour rate, which can be measured on the active plant.

There are 4 groups of experiments, in total consisting of 28 compositions. The simulant glasses were made up in the laboratory with similar chemical compositions to what is expected. The viscosity readings were from a high temperature rotating viscometer. Batches of the glasses were put into an alumina crucible. The glass was heated to 1200°C and held at this temperature for 45 minutes. The rotor was lowered 20mm into the glass and measurements taken every 10 seconds during the cooling period down to 900°C at a rate of 2°C per minute. The cleaning process will not be described.

3 VISCOSITY MODELLING

By using the estimated compositions and a range of temperature, a viscosity curve can be predicted. Multiple Linear Regression is used to show that the data is non-linear. Neural Networks will be used to model viscosity due to the high complexity and time consuming nature of mechanistic models.

3.1 Multiple Linear Regression

Multiple Linear Regression (MLR) is a linear technique used to predict (Montgomery, 2006) and takes the following form:

$$y_i = b_0 + b_1 x_{i,1} + b_2 x_{i,2} + b_M x_{i,M} + e_i \quad (1)$$

where b_0 is the regression coefficient, b_M is the coefficient of the M^{th} predictor, $x_{i,M}$ is the M^{th} predictor at time i, e_i is the error at time i and y_i is the value of the variable at time i. This method will not be explained further, as the MLR approach does not predict viscosity. Figure 2 shows the relationship between measured and predicted viscosity with the line measured viscosity equals predicted viscosity shown. It shows that the data is highly non-linear and therefore is not the correct approach to use.

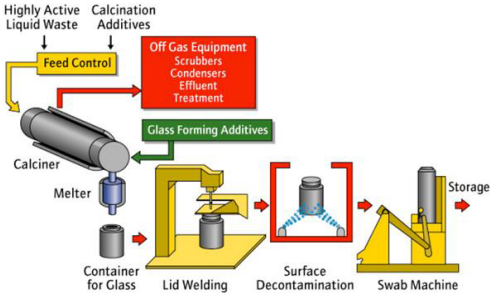

Figure 1: A Waste Vitrification Plant.

3.2 Neural Networks

In this paper, Neural Networks are analysed as a methodology to model viscosity using composition and temperature. The most commonly used is the multi-layer feed forward neural network. Inputs are presented at the input layer, I_i. The data is propagated through the network through complex connections. The hidden layer structure defines the topology of a feed forward network. It is possible to have more than one hidden layers and is proven to approximate any continuous non linear function with sufficient number of hidden neurons (Cybenko, 1989). Single hidden layer neural networks are used in nonlinear system modelling (Pham and Liu, 1995; Lennox et al., 1998).

Each interconnection has a scalar weight, w_i which modifies the signal strength. The neurons within the hidden layer: sum the weighted inputs and pass through a non-linear activation function as well as a bias, b. The output of a hidden neuron, O known as the sigmoidal neuron activation function and its output is in the range (0, 1).

$$S = \sum_{i=1}^{nh} (b + w_i I_i) \quad (2)$$

$$O = \frac{1}{1 + \exp(-S)} \quad (3)$$

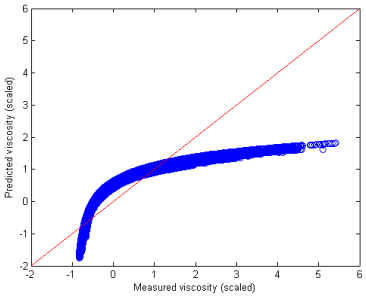

Figure 2: Measured against predicted viscosity using MLR.

Table 1: Mean squared errors (scaled) of neural network model predictions.

Data set	MSE
Training	1.0879e-4
Testing	2.9992e-4
Validation	1.8430e-4

Network weights are such trained so that the sum of squared network prediction errors, Eq(4) is minimised.

$$J = \frac{1}{N}\sum_{t=1}^{N}(\hat{y}(t) - y(t))^2 \quad (4)$$

where N is the number of training data points, $\hat{y}$ is the network prediction, y in the target value, and t is an index of the training data. The most commonly used network training method is the back propagation training method (Rumelhart et al., 1986), where network weights are adjusted as follows.

$$\Delta W(k+1) = \alpha \Delta W(k) - \eta \frac{\partial J}{\partial W(k)} \quad (5)$$

$$W(k+1) = W(k) + \Delta W(k+1) \quad (6)$$

$W(k)$ and $\Delta W(k)$ are the weight and weight adaptation at the training step k respectively, α is the momentum coefficient, and η is the learning rate. Training can be terminated when the error gradient is less than a pre-specified value, e.g. 10^{-6} or by a cross validation based stopping criterion. When using the latter, data for building a neural network model is divided into a training data set and a testing data set. The network prediction error on the testing data is continuously monitored and terminated when the testing error stops decreasing.

3.3 Neural Networks Viscosity Prediction

The glass contains 24 different components, C_i and will be in terms of number of Moles. The temperature, T, ranges from 1200 to 900°C in the experiment. The output is the viscosity, V which is of the form:

$$V = f(T, C_1,...,C_{24}) \quad (7)$$

The data set was split up into training (20%), testing (40%), and validation data sets (40%). The data were trained using the Levenberg-Marquardt optimisation algorithm (Marquardt, 1963) with regularisation and early stopping. The purpose of regularisation and early stopping is to avoid over-fitting noise in the training data. A regularisation term is added to the training objective function to penalise large network weights. A feed forward neural network with 25 inputs, 7 hidden nodes in a single hidden layer and 1 output was created.

4 RESULTS

4.1 Neural Network Predictions

The model proved to be successful at modelling viscosity using composition and temperature. Figure 3 shows a plot of temperature against viscosity. The blue line represents the true values while the red lines represents the model predictions. The error appears to be small. Table 1 gives the mean squared errors (MSE) of the neural network model predictions on the training, testing, and unseen validation data sets.

The MSE given in Table 1 are for scaled data, i.e. scaled to zero mean and unit variance. The very low MSE values in Table 1 indicate that the developed neural network model is very accurate.

4.2 Sensitivity Analysis of the Neural Network Model

Initial work has been carried out to investigate the model sensitivity to various model inputs. This study uses a numerical method where a small perturbation is added to one of the inputs while keeping other inputs unchanged. The ratio between the resulting change in model output and the applied model input is the model sensitivity to that input:

$$\text{Sensitivity} = \frac{\delta V}{\delta C_i} \quad (8)$$

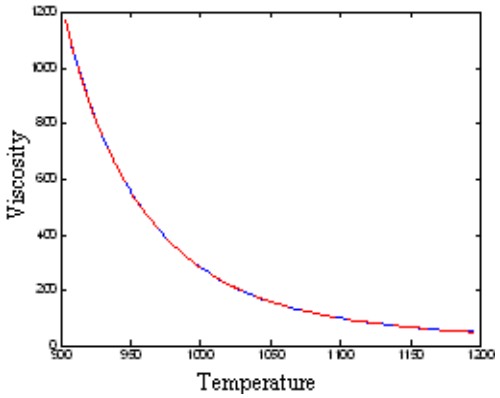

Figure 3: Graph showing actual (blue) and predicted viscosity curve over time (red).

The sensitivity changes, due to the non-linearity. If it changes considerably with only small changes in the component, then this suggests that the component significantly affects the viscosity.

Figure 4 shows the sensitivity curve when a variables is perturbed. Above 0.035Moles, there is an increase in viscosity, highlighting that variable has a big effect on viscosity.

5 CONCLUSIONS

This paper has used a well proven technique, multilayer feed forward neural networks, to predict the viscosity over a range of temperatures and different glass compositions. The prediction error (MSE) of the model for this range of feed was found to be 1.84×10^{-4} for the scaled validation data set which highlights the model's accuracy at predicting viscosity.

The model is only valid over a certain range for each variable, but in future work the model will be adapted for further different compositions and feeds. The work carried out so far has provided encouraging predictions for a larger range of compositions. This will be developed into a user tool for a greater understanding of how the composition will affect the viscosity.

ACKNOWLEDGEMENTS

The author would like to thank Northern way and Technology Strategy Board for part funding the Knowledge Transfer Partnership. The author would also like to thank Barbara Dunnett, National Nuclear Laboratory for the initial guidance on this study.

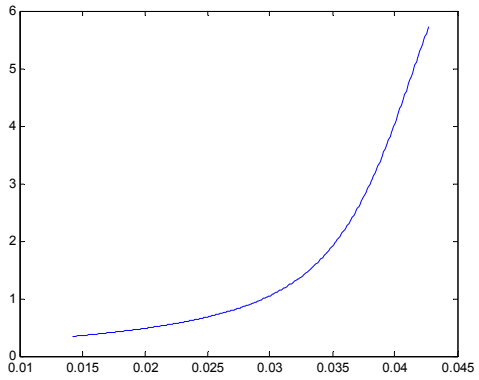

Figure 4: Sensitivity graph for variable 25.

REFERENCES

Bhat, N. V. and T. J. McAvoy (1990) "Use of neural nets for dynamical modelling and control of chemical process systems", *Computers & Chemical Engineering*, 14, 573-583.

Bishop, C. (1995) *Neural Networks for Pattern Recognition*. Oxford University Press: Oxford.

Bulsari, A. B., (Ed), (1995) *Computer-Aided Chemical Engineering, Vol.6, Neural Networks for Chemical Engineers*, Elsevier: Amsterdam.

Cybenko, G. (1989) "Approximation by superposition of a sigmoidal function", *Math. Control Signal Systems*, 2, 303-314.

Duchesne, M. A., Macchi, A., Lu, D.Y., Hughes, R. W., McCalden, D., J. Anthony, E. J., (2010) Artificial neural network model to predict slag viscosity over a broad range of temperatures and slag compositions - *Fuel Processing Technology*, *Volume 91, Issue 8, Pages 831-836*

Girosi, F. and T. Poggio (1990) "Networks and the best approximation property", *Biological* Cybernetics, 63, 169-179.

Lennox, B., Rutherford, P., Montague, G. A., Haughin, C., (1998) Case study investigating the application of neural networks for process modelling and condition monitoring – *Computers & Chemical Enginerring, Volume 22*, Issue 11, Pages 1573-1579

Marquardt, D. (1963) "An algorithm for least squares estimation of nonlinear parameters", *SIAM J. Appl. Math*., 11, 431-441.

Montgomery, D. C., Peck, E. A., Vining, G. G., *Introduction to Linear Regression Analysis*, Wiley Series in Probability and Statistics

Narendra, K. S. and K., Parthasarathy (1990) "Identification and control of dynamical systems using neural networks", *IEEE Transactions on Neural Networks*, 1, 4-27.

Park, J. and I. W. Sandberg (1991) "Universal approximation using radial basis function networks", *Neural Computation*, 3, 246-257.

Pham, D. T., Liu, X., (1995) Neural Networks for Identification, Prediction and Control. Springer-Verlag London Limited. 4th edition

Rumelhart, D. E., G. E. Hinton, and R. J. Williams, "Learning internal representations by error propagation", in *Parallel Distributed Processing*, (Eds) D. E. Rumelhart and J. L. McClelland, MIT Press, 1986.

Steele, C. J., Dunnet, B., Riley, A. D., Ferguson, K., Gribble, N., Short, R., (2011) Viscosity of simulated nuclear waste vitrified product *International Conference on the Chemistry of Glasses and Glass-Forming Melts, To be published*

Zhang, J., Morris, A. J., Martin, E. B., Kiparissides, C., (1998) Prediction of polymer quality in batch polymerisation reactors using robust neural networks *Chemical Engineering Journal 69, Pages 135-143*