

## Investigating Waste/Binder Interactions By Neural Network Analysis

C. D. Hills, J. A. Stegemann, N. R. Buenfeld

Imperial College of Science, Technology and Medicine, London, SW7 2BU, UK

### ABSTRACT

Solidifying cement/waste mixtures comprise highly complex chemical environments. The difficulty in predicting either positive or detrimental interactions between different components hampers the design of solidification treatment processes and can result in either environmentally unacceptable products which require expensive remediation, or unnecessary overdosing of additives. Development of a reliable diagnostic and predictive system for assessing treatability of wastes by solidification would therefore be a great benefit to users. No efforts in this regard have yet been undertaken because of the enormous number of variables involved. However, a preliminary exploration of the concept of using neural networks for this purpose has shown that it is possible to train a network to predict solidified waste properties to an acceptable degree of accuracy. The present work reviews factors that influence the properties of stabilised/solidified wastes, presents research objectives designed to improve waste solidification technology and preliminary results from neural network modeling studies.

### 1. INTRODUCTION

Solidification systems employing cementitious and pozzolanic binders rely upon hydration reactions to physically and chemically bind waste species to form products that are more stable in the environment. The fundamental principle behind this technology appears relatively simple and involves the controlled application of hydraulic binders, such as ordinary Portland cement. Correctly applied, these binders minimize the solubility of contaminants and entrap them in the structure of a solidified product by sorption, lattice inclusion<sup>1</sup> and enclosure in the matrix porosity. Solidification is used to significantly improve the handling and facilitate the safe disposal of a wide range of hazardous waste materials and contaminated land<sup>2</sup>.

In practice, the wide choice of solidification systems coupled with the fact that wastes from different processes, and with differing characteristics, can not all be treated alike means that choosing an appropriate binder for a given waste stream is a complex undertaking. Inappropriate solidification formulations can result in handling difficulties during processing and placement, failure of the solidified material to set and harden into a durable monolith, physical degradation of the matrix with time, and/or provision of a chemical environment in which contaminants are not fully immobilized. All of these results indicate an inadequate protection of the environment. Reports of environmental problems associated with considerable quantities of commercially solidified wastes<sup>3,4,5,6</sup>, despite the application of testing regimes required by UK regulatory authorities, suggest that there are limitations on the use of hydraulic cements in this application, which are not yet defined.

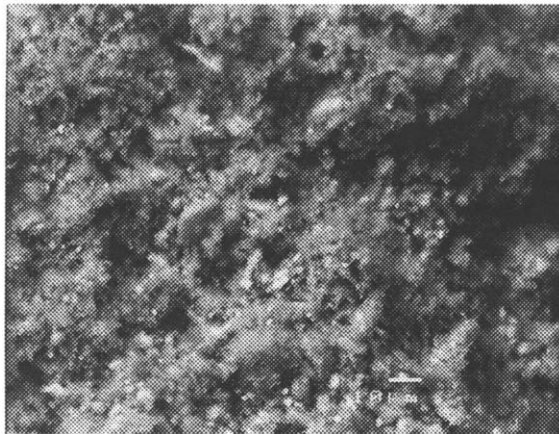
This work explores some of the factors responsible for effective waste solidification. Its purpose is to address the potential of neural network analysis for contributing to a better understanding of the relationship between waste and binder and variables of importance to solidified product properties in both the short and longer term.

## 2. INTERFERENCES WITH CEMENT HYDRATION

In preparing concrete, 10 to 20% of hydraulic binder is blended with inert aggregate to yield a high-strength building material. Small amounts of chemical admixtures (<2% by weight of cement) can have a significant impact upon the properties of concrete <sup>7</sup>. During solidification, small quantities of cementitious binder can be exposed to large quantities of chemicals. For example, a waste containing 45% by wet weight of solids might be treated with as little as 10% by weight of hydraulic cement. A considerable number of compounds are now known to affect hydration processes <sup>8,9,10,11</sup>, causing effects such as: acceleration or retardation of set (which, in the extreme, can result in flash set or complete inhibition of hydration), false set, increased water consumption, and matrix disruption by expansive reactions <sup>12,13,10</sup>.

Figure 1 shows an example electron photomicrograph of a Portland cement bound solidified product about 1 year of age. The solidified product contained 40% blended neutralised heavy metal waste and 15% binder, on a wet weight basis. The microstructure is devoid of the crystalline phases which are common in conventional cement and concrete and is typical of the evidence of persistent interference with hydration.

Figure 1. Micrograph showing typical solidified product microstructure (1 year old solidified inorganic waste).



Both organic and inorganic waste species are capable of interference with hydration mechanisms by hindering transport processes which occur into and out of cement particles during normal hydration <sup>14,15</sup>, and consequently impair microstructural development <sup>16</sup>.

Nonpolar organic compounds, such as polychlorinated biphenyls and many polynuclear aromatic compounds, tend to be water insoluble and will tend to partition with any solid phase in preference to water <sup>17</sup>. Although this may have the effect of reducing their leachability, sorption of even small quantities of organics onto the solid binder can interfere with hydration. Many soluble polar organics do not tend to react with cement, and their leachability is therefore not reduced by solidification; organic compounds which do produce salts, complexes or precipitates may also remain mobile by compromising hydration reactions.

Inorganic species are generally considered most amenable to cement-based treatment, because they tend to be reactive with hydraulic binders. However, it is this very reactivity which also

affects cement hydration reactions. The severity of retardation effects for a variety of anions and cations has been ranked by several authors e.g. <sup>18,9</sup>, but to a large extent this is an oversimplified approach, and only possible when working with pure compounds, rather than blended species as they are found in real wastes. In real wastes there are complicating factors which prevent simple ranking, for instance there may be confounding effects resulting from a particular combination of cation and anion, the concentration of each, the presence of other compounds in the waste, and curing conditions.

By way of a simple example, less than approximately 1% of  $\text{CaCl}_2$  retards the set of Portland cement, greater amounts cause acceleration <sup>19</sup>, and more than approximately 4% causes matrix disruption <sup>20</sup>. Similar effects have been observed for actual solidified products; use of Portland cement to solidify an electric arc furnace dust resulted in initial brief retardation followed by rapid acceleration of hydration at 30% dry weight electric arc furnace dust, but only a strong inhibition of hydration at 60% electric arc furnace dust <sup>21</sup>.

Figure 2 illustrates the synergistic effect of the addition of interfering agents on the heat of hydration of OPC. Compounds found in waste materials were added to OPC paste as metal hydroxides and sodium salts in single and multiple additions <sup>12</sup>. Whereas most additions caused either retardation or acceleration, a combination of metal hydroxides was found to cause indefinite retardation of hydration reactions.

### 3. INFLUENCE OF BINDER CHEMISTRY

The chemistry of a hydraulic binder system can be tailored by the inclusion of other cementitious materials, such as blast furnace slag, and/or pozzolanic mineral admixtures, such as coal fly ash. Alternative binders to Portland cement can sometimes help overcome deleterious effects induced by a waste <sup>22</sup>. For instance, in the example of the solidified electric arc furnace discussed earlier, use of an activated blast furnace slag formulation in place of Portland cement resulted in a consistent mild retardation, which simplified control of the process <sup>21</sup>.

Figure 3 shows the effect of binder type on the amount of chromium leached from solidified waste forms containing a zinc and chromium rich plating waste extracted with distilled water for 24h, by end over end tumbling according to the modified DIN 38414 test <sup>23</sup>. Particularly at high waste additions, the concentration of chromium in the leachates was far higher for blended ordinary Portland cement (OPC) than for calcium aluminate cements (CACs) with the individual differences between pozzolan additions being of less importance. The relatively high chromium concentration leached from some samples indicated that chromium was speciated as  $\text{Cr}^{6+}$  in this waste. As the solubility of  $\text{Cr}^{6+}$  is not pH sensitive, the observed differences in leachability imply a change in speciation or uptake of the ion into the cement hydration products.

### 4. OTHER FACTORS

In general, wastes presented for solidification have undergone pretreatment in some way. Appropriate pretreatment can be important to maximize efficient use of binder. For example, pre-neutralization saves using valuable cement just to neutralize acidity and can help reduce interferences by taking interfering agents out of solution <sup>24</sup>. Precipitated species are incorporated in the solidified waste matrix as sludge particles, and species which were not precipitated remain available for incorporation in cement hydration products.

Curing conditions are another factor to which lamentably little attention is paid. The effect of some interferences is temperature dependent <sup>25</sup>, which suggests that use of an appropriate curing

temperature is important in obtaining the desired final properties in a waste form. For solidified wastes, as for concrete, freezing, drying and physical disturbance of a setting mixture have detrimental effects. Therefore, protection from freezing conditions, high humidity and placement prior to set are all essential for obtaining a high quality solidified product.

Figure 2. Effect of additions on the heat of hydration of OPC (after Hills et al. 12)

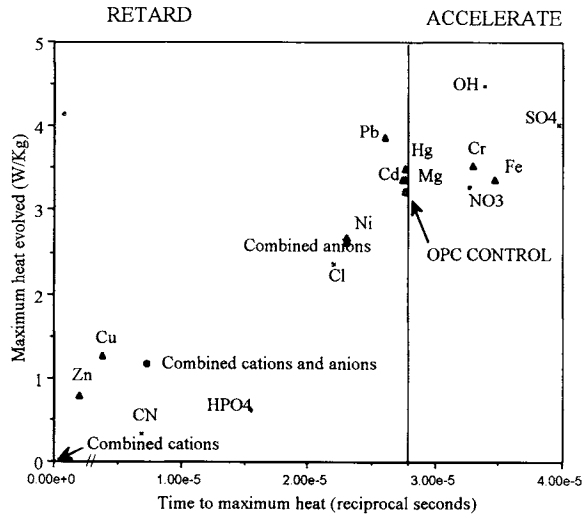
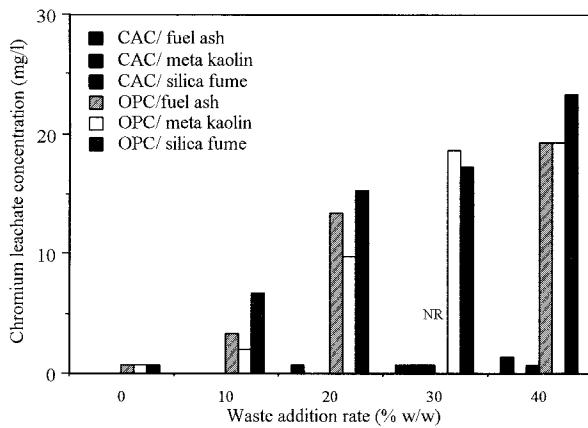


Figure 3. Chromium leached from blended OPC and CAC binders with increasing waste contents



Finally, it is worth noting here that other factors such as the method and efficiency of blending waste and binder and other aspects of processing such as mix design will all exert some influence on solidified product properties.

## 5. APPLICATION OF NEURAL NETWORKS

The many and complex potential interactions between the variables in a solidified product are not possible to evaluate by simple means. The aforementioned discussion of the deleterious interactions between waste and binder has concluded that a considerable number of materials are involved and that synergistic relationships are implicated. With this in mind, alternative means of identifying the relationships of importance are required and could incorporate an examination of existing data.

To-date little work has been carried out on evaluating existing data in order to aid future solidification practices. The reason for this may lie in the difficulty of approaching such a complex and clearly nonlinear problem with conventional data analysis techniques. However, it may be possible to address the problem using neural network analysis.

Neural network analysis is capable of processing of a variety of inputs, and therefore has advantages over conventional techniques for finding relationships between variables in complex systems. A neural network consists of a number of interconnected processors which are arranged in layers: are capable of receiving incoming information from several sources, weighting the information, and transmitting it in a single output to the next layer of the network. Learning by a neural network consists of changing the weights on the information transferred through the processors in a systematic fashion based on training. When enough training data is available neural networks can generalise to predict combinations of inputs not previously encountered, enabling them to be used to identify patterns in large data sets of many variables <sup>26</sup>.

An inevitable consequence of neural network analysis is that any relationships identified between variables in a dataset are non-mechanistic. While this might appear to be an obvious drawback, the advantage is that relationships can be identified without a preliminary mechanistic hypothesis. Thus, it may be possible to develop new insight into mechanisms, based on identification of new relationships through neural network analysis.

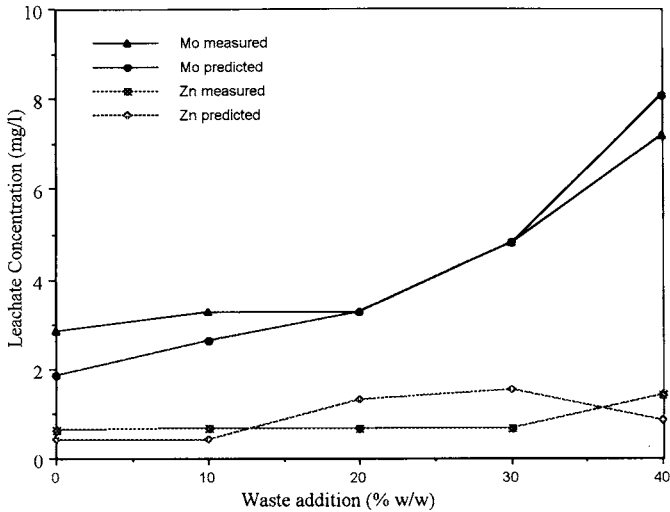
The application of neural network analysis to cementitious systems has been demonstrated elsewhere e.g. <sup>26,27,28</sup>. Glass et al. <sup>29</sup> have quantified a wide range of factors influencing the binding of chlorides in concrete from published data.

Preliminary experiments concerning the application of neural networks to analysis of laboratory solidification data, have been positive and encouraging. When applied to the data from a laboratory program involving 150 mixes produced from different waste/binder combinations to predict setting times, strength development and leachate metals composition an average accuracy of greater than 75% was achieved <sup>30</sup>.

Table 1 summarises the networks constructed and the results obtained. Figure 4, shows the predicted vs measured leaching of molybdenum and zinc. The close correspondence between measured and predicted concentrations clearly illustrates the potential of this approach.

Additional preliminary neural networks were constructed using specific information concerning the metals of interest, such as ionic radius, atomic weight, and valency and this has enabled other relationships to be generalised, however, further work is required before these results can be verified.

Figure 4. Predicted vs measured leaching values of molybdenum and zinc from laboratory solidified wastes (after Hills et al., 1997)



There is an urgent need to improve our knowledge concerning waste fixation through both short and long term research. Three main areas of research and development are required to be addressed as shown in Table 2.

## 6. OBJECTIVES FOR FUTURE DEVELOPMENT

Neural network analysis is a potential tool in several of these areas and could lead to a knowledge-based diagnostic and predictive system for predicting interactions between components of solidified waste, and final product properties. This would be a great benefit to users of the technology by: providing insight into the most important parameters in design of durable formulations of low leachability, eliminating or reducing the time and expense of treatability studies, streamlining performance testing enabling process refinement during full-scale solidification based on feedback of quality analysis and control data, and reducing the requirement for and subjectivity associated with human expertise. Table 3 lists parameters which can be found in the existing data and could be used to train and test neural networks to examine the relationships between solidified waste properties.

The relationships expected to be of particular interest are those between:

- waste/binder dosages, the presence of agents which normally interfere with cement hydration, and engineering properties.
- different measures of the transport characteristics of the solidified waste matrix (e.g., water absorption, and leachability in ANSI/ANS 16.1, and
- waste and binder composition and pH and acid neutralization capacity of the solidified product.

Table 1 Summary of data from neural network analysis of laboratory solidified wastes (after Hills et al. <sup>30</sup>)

INPUTS	NN1	NN2	NN3	NN4	NN5
Cement type (5 types)	YES				
Pozzolan (3 types)	YES				
Waste Type Content	YES				
Setting time (mins)	NO	If measured	NO	YES	Measured and predicted from NN3
Strength (kPa)	NO	If measured	YES	NO	Measured and predicted from NN4
Leachate metal (5 metals)	YES		NO		YES
Hidden neurons	12	22	14	15	14
OUTPUT	Leachate concentration (mg/l)		Setting time (mins)	Strength (kPa)	Leachate concentration (mg/l)
Average accuracy	63.2%	56.8%	76.8%	74.2%	75.3%

Table 2 Research and development effort for improving solidification technology

RESEARCH AREA	OBJECTIVE
Basic Science	elucidating waste binder interactions improving /optimising binders developing novel systems
Applied Science	improving materials processing characterising solidified product performance
Regulation	developing long-term monitoring framework specifying characterisation procedures developing guidelines for safe containment structures

## 7. CONCLUSIONS

When Portland cement is used to solidify waste materials the complex interactions that result cannot be easily characterised. Consequently, design of solidification formulations is complicated, and entails a high risk of failure.

Neural network analysis can be used to identify complex relationships between large numbers of variables and it may be possible to elucidate factors of importance to solidification from existing data.

Preliminary studies have been carried out by neural network analysis with the result that contaminant leachate concentrations of laboratory-prepared solidified wastes were predicted to an acceptable degree of accuracy. In addition, a number of other relationships were generalised indicating that this method may have considerable potential to aid our understanding of the important variables involved.

Table 3 Example of variables of potential importance for solidification

COMPONENT	VARIABLE
Binder	type composition mineralogy acid neutralisation capacity
Waste	type composition, mineralogy acid neutralisation capacity
Combined (Solidified Product)	setting time strength development moisture content specific gravity age water adsorption hydraulic conductivity acid neutralisation capacity pore water composition monolithic leachability (e.g. ANSI/ANS 16.1) leachability (distilled water/ acid batch extraction etc.) freeze thaw resistance wet/dry weathering resistance

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