Hybrid and Neural network based devolatilisation modelling in biomass fuel advanced systems

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Mathematical modelling techniques are often based on simplified assumptions which fall short of the close approximations levels required for good prediction of combustion phenomena. The complexities involved during combustion such as turbulence and emissions formation, as yet have not been fully explained and modelled on mathematical grounds.
The process of devolatilisation is very important in terms of NOx formation and flame stability, especially when using coal blends, biomass, pine or wood as the fuel. The lack of knowledge regarding devolatilisation while co-firing hampers the efforts to device efficient, stable and environmentally friendly combustion. The effect of devolatilisation on flame stability and emission formation has been thoroughly investigated by the researchers. The rapid devolatilisation is an important feature that establishes flame stability.
All the devolatilisation models used for Computational Fluid Dynamics (CFD) combustion codes are over simplified and does not represent the actual behaviour of volatile release such as single reaction models.
The advanced forms of devolatilisation models that include multi-step reaction models were thought to resolve the aforementioned problems in predictions. These models have posed other problems. The multi-step models are very complicated and contribute to make the system very slow in terms of their convergence rate. In recent years researchers have made many attempts to introduce further advanced devolatilisation models, that include FLASHCHAIN, CPD, FG-DVC. These models are based on micro-molecular knowledge of the chemical bondage within the coal structure. They are being used off-line and not as a direct part of the CFD simulation.
Neural and hybrid model could be efficient methods to overcome this problem. Neural networks are fast, reliable and simple methods developed to solve complex modelling and control problems. The strength of neural networks lies in the fact that they can be built using example by example training thus effectively exploiting the experimental data.
The artificial neural network (ANN) model could be coupled with the CFD: in this way, the complicated information that the CFD needs to entirely describe the problem, is being acquired on-line from the neural network model. The artificial neural network (ANN) model could be coupled with the CFD: in this way, the complicated information that the CFD needs to entirely describe the problem, is being acquired on-line from the neural network model. ANN model has been developed on the database "BIOMASSDEVO" [ ] which collects the data from about 130 different experiments, on 115 different type of biomasses and residues, reported in current literatures, from the 1982 to the 1999.
Also hybrid models are part of this study; these particular models are the result of the interaction of parametrical structures (first principles models) and non parametrical ones (ANN).
In particular in this report two different approaches are investigated.
The first is the ANN model that predicts directly the rate of devolatilisation of the biomass and residue by the chemical composition and operating conditions.

Furthermore, the structure of the hybrid model is based on the following evidences:

- devolatilisation of biomasses is well described by lumped parameter models, with key component fractions as input (as described in 2)
- EDTGA and TGA tests on reference coals and biomasses showed that devolatilization cannot be described as a single rate, but a sub model for devolatilization is needed which can be based on

the activation energy distribution (DAEM) and/or lumped parameter with key components. The models, using parameters from TGA analysis, were tested and proved to be suitable to describe devolatilization of coal and biomass particles in a wide temperature and heating rate range.

- it is possible to define a simple first order reaction model *(SFOR)* which well describes devolatilisation only in a limited range of heating rates. This can be more suitable than a DAEM, due to a comparable precision and a lower complexity, to be used in comprehensive codes.
- *Ultimate Analysis* of a residue or a biomass is commonly available; this is not the case of chemical analysis, either by direct or indirect methods.

The second consists of hybrid models that try to add important physical information to the devolatilisation model. The structure of kinetic expressions tested for devolatilisation is included in the physical part of the model, in order to consider an Arrhenius-like dependence on temperature and degree of conversion. On the other hand, the relations between the ultimate analysis and the chemical components liable of the kinetics of the process have been also included in the hybrid approach.

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1. INTRODUCTION

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The process of devolatilisation is very important in terms of NOx formation and flame stability, especially when using coal blends, biomass, pine or wood as the fuel. The lack of knowledge regarding devolatilisation while co-firing hampers the efforts to device efficient, stable and environmentally friendly combustion. The effect of devolatilisation on flame stability and emission formation has been thoroughly investigated by the researchers. The rapid devolatilisation is an important feature that establishes flame stability.
All the devolatilisation models used for Computational Fluid Dynamics (CFD) combustion codes are over simplified and does not represent the actual behaviour of volatile release such as single reaction models.
The advanced forms of devolatilisation models that include multi-step reaction models were thought to resolve the aforementioned problems in predictions. These models have posed other problems. The multi-step models are very complicated and contribute to make the system very slow in terms of their convergence rate. In recent years researchers have made many attempts to introduce further advanced devolatilisation models, that include FLASHCHAIN, CPD, FG-DVC. These models are based on micro-molecular knowledge of the chemical bondage within the coal structure. They are being used off-line and not as a direct part of the CFD simulation.
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Also hybrid models are part of this study; these particular models are the result of the interaction of parametrical structures (first principles models) and non parametrical ones (ANN).
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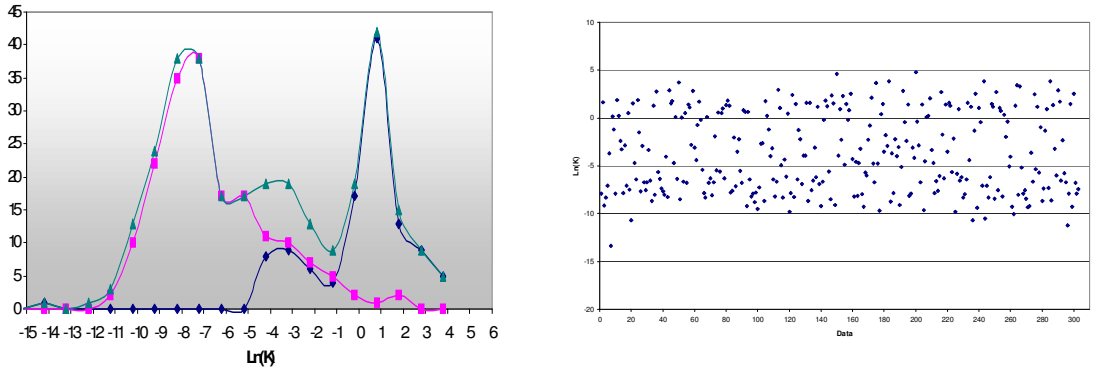
ANN MODEL
Artificial neural networks

The artificial neural networks were developed with the aim of reproducing, in a simplified way, the brain structure so that they can learn from external data and interpret new data. According to the human brain, the ANN are composed of several processors, called neurons, linked each other by connections, to which weights are assigned. The common ANN structure is the *feed-forward* and its elements are three different layers: the input layer which receives the process information, the hidden layer which works out information, and the output layer which gives the calculated value.
ANN model has been developed on the database "BIOMASSDEVO" [ ] which collects the data from about 130 different experiments, on 115 different type of biomasses and residues, reported in current literatures, from the 1982 to the 1999.
The database was built up not only recording the row data found in the articles data, but also trying to evaluate kinetic parameters, thermal history of the samples and, where possible, verifying the actual heating rate of the experimental runs. According to the experimental technique used in the experiments, it is possible to summarise the following main parameters,

Equipment	**Temperature (°C)**	**Holding/Residence time (sec)**	**Heating rate (°C/sec)**
TGA	50-800	100-200	0.083-1.33
Fluidized bed	250-750	0.1-1	$<1*10^3$
Wire mesh reactor	250-750	10-50	$1*10^3$-$1*10^5$
Pyroprobe	< 1400	0.4-100	$1*10^3$-$1*10^5$
Screen heater	250-1300	0.1-1	$1*10^3$-$1*10^5$

The devolatisation rate varies from a maximum of 120 (mg/mg sec) to a minimum of 1.6 10^{-6} (mg/mg sec), according to the temperature, the heating rate and the composition of the biomass. In



the figure 3 the natural logarithm of the kinetic constant for each experiment is reported.

g. 3

The natural logarithm of the rate of devolatilisation varies from 6 to –14: it is relevant to show the normal distribution of that parameter to understand its variation with the operating condintions.
The heating rate is one of the parameter that controls the value of the rate of devolatilisation, so it is possible to identify two different data sets by dividing all the values into two categories: devolatilisation at high heating rate (>10°C/sec), and devolatilisation at low heating rate (<10°C/sec). Those sets correspond to different kind of apparatus or experimental runs as reported in the table above. The figure 5 shows this two set. The starting material can be identified, on a chemical point of view, by the content of different *key components* , that have been *Cellulose,*

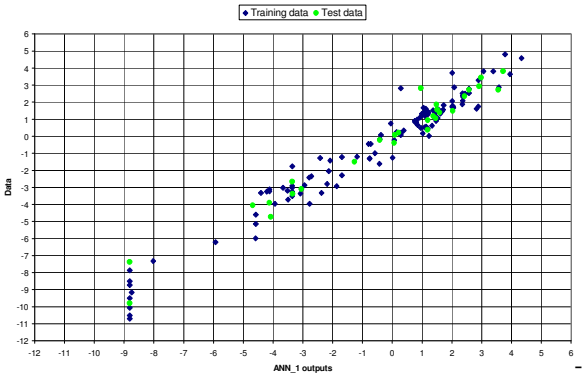
Lignin, Hemicellulose for biomasses, with the addition of *plastics (Polypropylene, Polyetilene, PET)* for residues. The other parameters that determines the devolatilisation conditions are the *size* of the biomass particle, the *temperature* and the *holding/residence time*.
All these parameters are the inputs of the neural network that was trained to predict the value of the rate of devolatilisation. The output are then plotted in form of an Arrhenius plot, in order to describe the variation of the rate of devolatilisation as a function of the temperature.
The neural model consists of two feed-forward neural networks, ANN-1 and ANN-2, with one hidden layer for each, table 1. The number of inputs are seven and the output is one, the hidden layer has a different number of neurons, because of the difference in the two data sets.

	Set	**Type**	**Size**
ANN 1	**High heating rate**	FF	7x6x1
ANN 2	**Low heating rate**	FF	7x6x1

The way of processing the information is depicted in the following diagram.

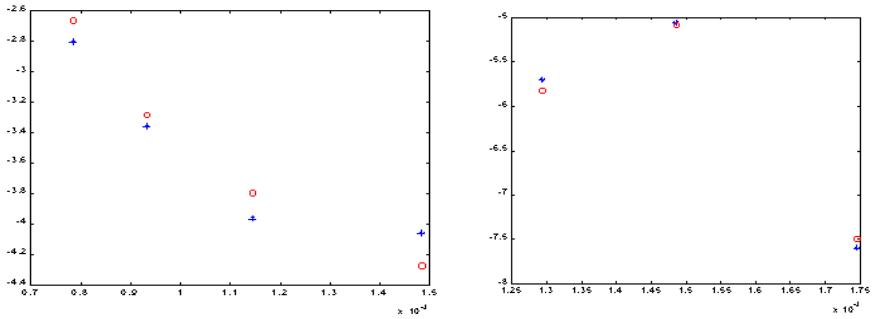
BIOMASSDEVO ⇒ Particle size, Cellulose %, Lignin %, Hemicellulose %, Heating rate, Temperature, Holding/Residence time ⇒ ANN ⇒ Rate of devolatilisation (expressed as Ln(K))

The activation functions of the neurons are sigmoidal. The optimisation algorithm is related to the FMINU function which is a part of the Matlab® software, used as base for the neural network implementation. The default algorithm is the BFGS Quasi-Newton method, with a quadratic and cubic line search procedure.
During the training step, the error between the real set and the validation set is calculated and its value is used to update the weights of the neurons.
One of the most important parameter of the neural model is the number of the neurones of the hidden layer. To find the right number of neurones, it is useful to train the net on more than one set of data. The set should be divided in two part: the training set and the test set. Then, for every set of data we have recorded the value of the parameters of the ANN performance. In particular it is important to consider the different value of the neural model on the training set and on the test set.
Another important parameter is the number of iteration made to find the optimisation of the ANN learning algorithm.
The figure xx refer to the **ANN1** (high heating rate). Similar results were obtained for the **ANN2** (low heating rate).



In the following examples in Arrhenius plot form are reported on some data sets from the database (the number indicates the identification on the database; *; experimental data; o ANN-1 predictions).

Almond shells (size 372 micron) Entrained Bed reactor
Fig. 8.3: **96-4** Cellulose CDS intruments model Pyroprobe 1000



HYBRID MODEL

Detailed first principle models and "black-box" models have complementary advantages and disadvantages. The former, as shown in section 2, require in many case a large number of physical and chemical parameters, which may be difficult to obtain, and are usually computationally heavy; at the same time they offer a precise description of the process and have the ability to extrapolate, i.e. to supply reasonable data and information for operating conditions for which experimental data are not available.
The latter, as reported in section 3, do not require kinetic parameters and involve only easy calculations to estimate the process outputs from process inputs; however, they may be obtained only if a very large number of data are available and they give completely meaningless results for operating conditions which are very different from the ones on which they have been built.
A "hybrid" model has been developed, with the aim of retaining the advantages and eliminating the disadvantages of the two approaches.
There are two main different methods to combine the two models, the first called “series model”[1] and the second “parallel model”[2]. In the first approach the neural model estimates the process parameters needed by the physical model, which finally gives the outputs. The advantages of this approach is that the output behaviour is guaranteed.
In the second approach the neural and the physical models are put in parallel and the neural model compensates for discrepancies between data and predictions of the parametric model.
Moreover the simplifications in the first principle model have to be chosen so that neither many parameters nor too heavy computations will be required.
Furthermore, the structure of the hybrid model is based on the following evidences:

- devolatilisation of biomasses is well described by lumped parameter models, with key component fractions as input (as described in 2)
- EDTGA and TGA tests on reference coals and biomasses showed that devolatilization cannot be described as a single rate, but a sub model for devolatilization is needed which can be based on the activation energy distribution (DAEM) and/or lumped parameter with key components. The models, using parameters from TGA analysis, were tested and proved to be suitable to describe devolatilization of coal and biomass particles in a wide temperature and heating rate range.
- it is possible to define a simple first order reaction model *(SFOR)* which well describes devolatilisation only in a limited range of heating rates. This can be more suitable than a DAEM, due to a comparable precision and a lower complexity, to be used in comprehensive codes.

- *Ultimate Analysis* of a residue or a biomass is commonly available; this is not the case of chemical analysis, either by direct or indirect methods.

On the basis of these considerations an hybrid scheme has been defined in which a first ANN (H1) determines the amount of key components on the basis of ultimate analysis and other available information on chemical nature of the material. Then, a second ANN (H2) evaluates the kinetic constant and the global amount of volatile releasable, V∞ . Finally the SFOR model calculates the volatile yield.
The scheme of the hybrid model is described in figure 10:

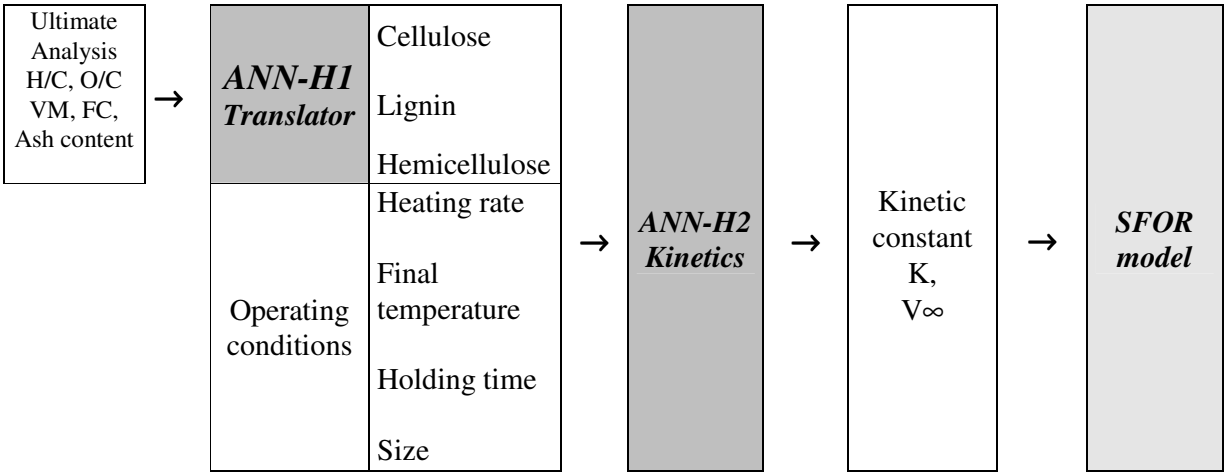


Fig. 10

All the steps of the hybrid model, ***ANN-H1 - ANN-H2 – SFOR*****,** are described in the following.
The chemical analysis is probably the most significant way of characterising the biomass and residue from a kinetic point of view.
The first attempt has been to train a neural model that could correlate the ultimate and proximate analysis with the chemical one. The important parameters that would give the information of how the carbon, oxygen and hydrogen are linked to form the various molecules of cellulose, hemicellulose and lignin are the following:

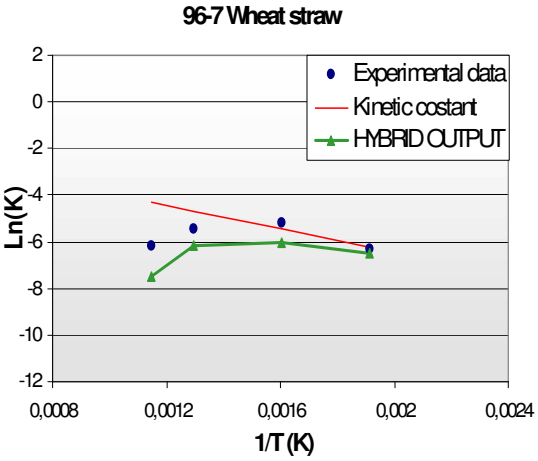
- Ultimate analysis
- Ratio H/C and O/C
- Volatile matter
- Fixed carbon
- Ash content

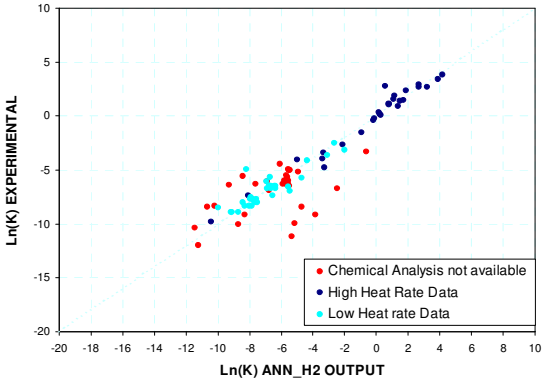
All these parameter are the input for a the ANN-H1, with the scope to create a model that gives significant information on the amount of cellulose, lignin and hemicellulose of a generic residue or biomass.
The chemical analysis is probably the most significant way of characterising the biomass and residue from a kinetic point of view.
Table 3 shows the range of variation the elementary composition of biomass.

Ultimate analysis

Carbon wt%	17-64
Hydrogen wt%	2-8
Oxygen wt%	3-52

Furthermore, ratios **H/C** and **O/C** can represent another way to individuate the nature of biomass or residues: the H/C - O/C plot for the available data is reported in appendix **A**. The relative position of the data points in the plot suggests which is the most relevant chemical component of the biomass. For examples in the bottom left there are the coals, the point (0,0) represent elementary carbon, while in the upper right "pure" celluloses are located, with the largest amount of oxygenated molecules. The woods and the agricultural residues are placed in the middle, because of the presence of lignin. An AN approach seems to be appropriated for our purposes. The applicability of this approach is currently tested.





As previously reported in section 3, ANN model well predicts the kinetic constant for devolatilisation in different operating conditions.

Another artificial neural network (H2) can then predict the rate of devolatilisation K [mg/mg*sec] using both the information given by the ANN-H1, the chemical composition, and the other given directly as outer inputs as the heating rate, size, temperature, holding time.

In order to take into account the effect of conversion on devolatilisation rate and the different amount of volatiles potentially released by a biomass under different severities of treatment, the term $\left(V^{\infty} - V \right)$ should be considered in the simple first order model. Thus H2 outputs, K and V∞, are the inputs parameters for the kinetic expression that describes the volatile yield:

$$\frac{dV}{dt} = K * \left(V^{\infty} - V \right)$$

By this model it is possible to describe the complex process of devolatilisation of biomass and residue in comprehensive codes, applying a simple first order reaction model. This is due to the specific parameter calculated both from the physical relations and from the neural network. This model is currently under training.

REFERENCES

[1] D.C.Psichogios and L.H. Ungar, A hybrid neural network-first principles approach to process modelling, *AIChE Journal*, October 1992 vol.38, No. 10

[2] M.L.Thompson and M.A.Kramer, Modelling chemical processes using prior knowledge and neural networks, *AIChE Journal*, August 1994 vol.40, No. 8

[3] V. Cozzani; A. Lucchesi; G. Stoppato; G. Maschio, A New Method to determine the composition of biomass by thermogravimetric analysis, *Canadian Journal of Chem. Eng.*, v.75 n°1 pp. 127-133, Feb. 1997.

[4] J. Simitzis, High temperature pyrolysis of novolac resin biomass composites, *Journal of Analytical and Applied Pyrolysis*, v30 (1994) pp 161-171.

[5] ASTM, D 1104-56, Standard method of test for holocellulose in wood, 1956, 846-847.

ASTM, D 1106-56, Standard method of test for lignin in wood, 1956, 848-850.

[6] C.A. Koufopanos; G. Maschio; A. Lucchesi, Kinetic modelling of the pyrolysis of biomass and biomass component, *Canadian Journal of Chem. Eng.*, v.67 pp. 75-84, Feb. 1989.

[7] F.A. Agblevor; R.J. Evans; K.D. Johnson, Molecular-Beam Mass-Spectrometric analysis of lignocellulosic materials, *Journal of Analytical and Applied Pyrolysis*, v30 (1994) pp 125-144.

[8] NREL Research Institute 1999, *http://www.nrel.org*

[9] "BIOMASSDEVO" Department of Chemical Engineering ,Univerity of Pisa 1999