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Inverse modelling and model complexity in computational pyrolysis

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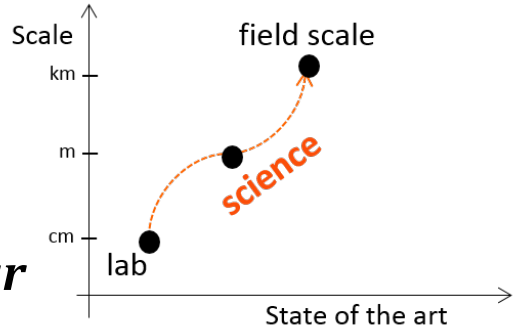
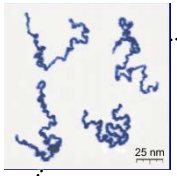
I have a dream:
The Up-scale Path
from fundamentals to real behaviour

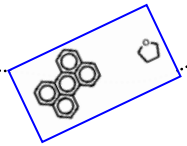


Fire Behaviour

Product

!



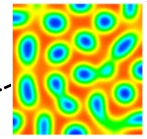


$$\text{solid} \rightarrow \text{gas}$$
$$\text{solid} \rightarrow \beta\text{ - solid}$$
$$\beta\text{ - solid} \rightarrow \text{gas}$$

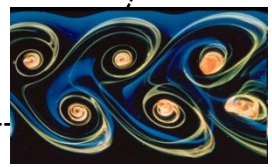
Elemental Reaction

Q

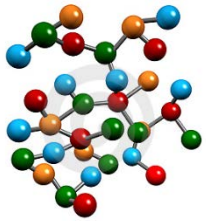
Fundamental Physics



$$\frac{\partial T}{\partial t} = \alpha \nabla \cdot T$$



Fundamental Chemistry



Why is Pyrolysis of interest?

- Onset and evolution of material degradation.
- A driver of ignition and flame spread.
- Burning with flame, or without flame.

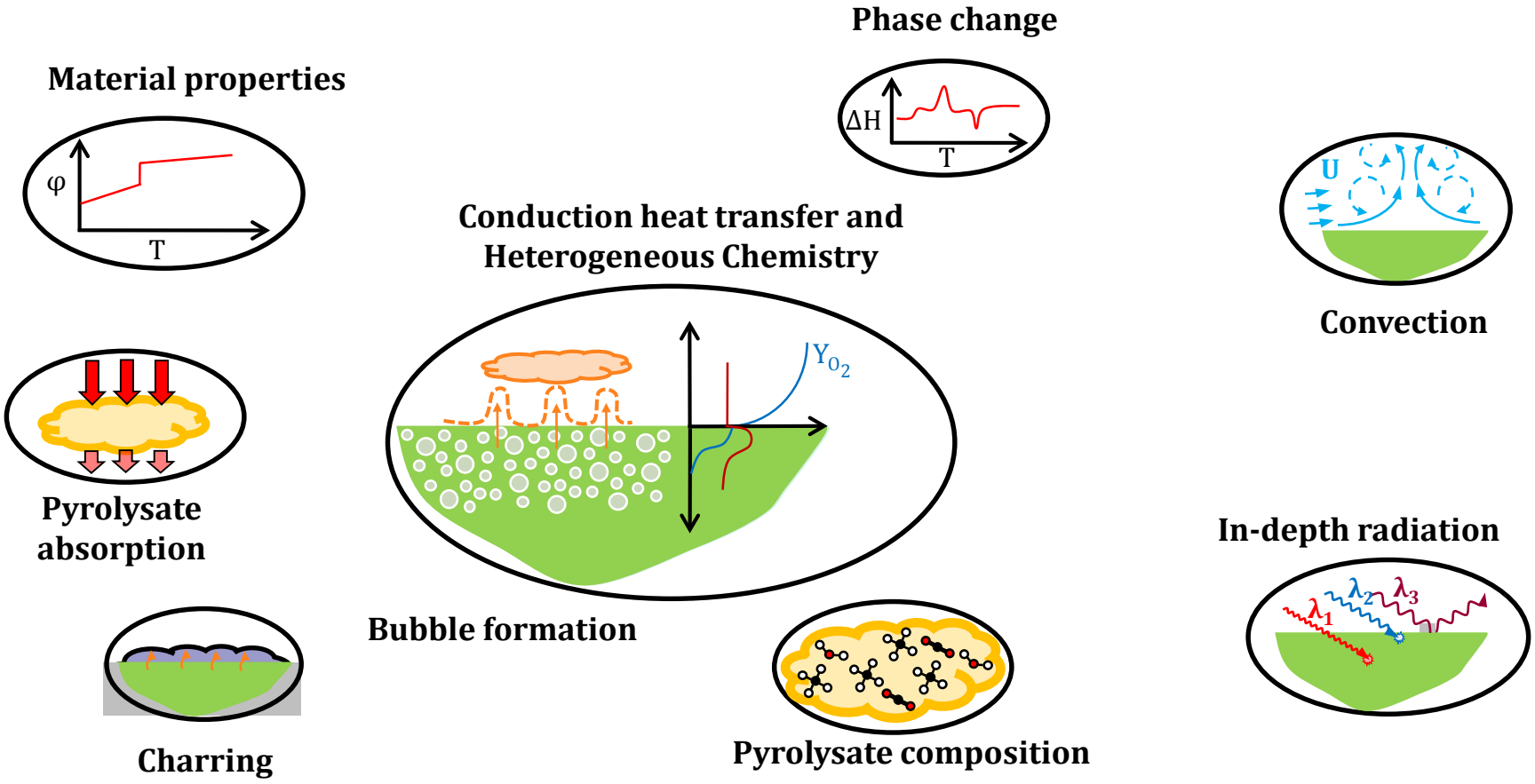
"…22nd century to advance knowledge in chemistry and physics to the state that most required fire knowledge could be computed from first principles…"

Prof. Howard Emmons, Harvard, 1984

- He predicted that turbulence will be solved before pyrolysis.
- Note: Prof. Emmons is the founding father of *Fire Science* and also of *Turbulence*.
- Given that the historical ratio of #researchers working on pyrolysis per #researchers working on turbulence is 1/500, we are making sure he is right.

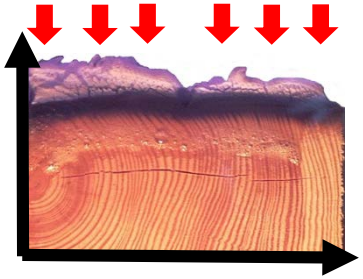
Emmons, **History of Further Fire Science**, Fire Technology, 1984

Pyrolysis: multiphysics problem



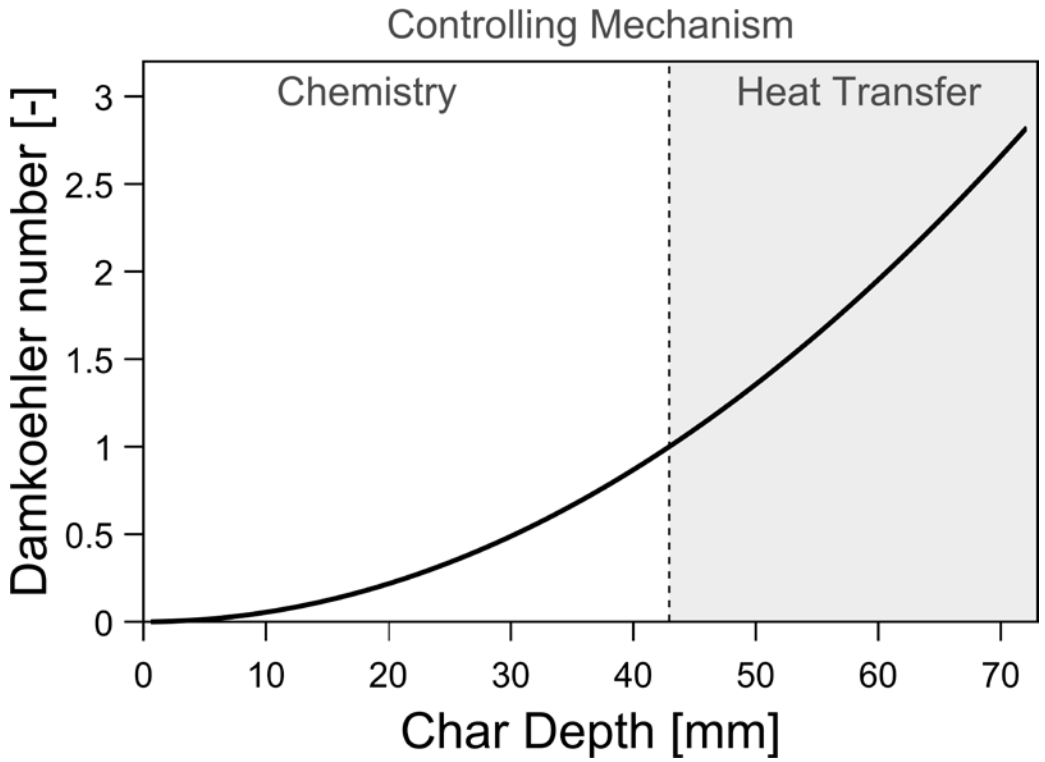
Pyrolysis: the simultaneous chemical decomposition and phase change that provide the gaseous fuel feeding the flame burning over a solid. Controlled by heat transfer and condensed-phase kinetics

Chemistry vs. Heat Transfer



$$Da = \frac{\rho c L^2 \dot{\omega}}{k} = \text{Ratio of chemical to physical times}$$

$$Bi = \frac{hL}{k} = \text{Relative thermal thickness}$$



Transport parameters:

W. C. Park, A. Atreya, H. R. Baum, Determination of pyrolysis temperature for charring materials. *Proc. Combust. Inst.* **32 II**, 2471–2479 (2009).

Kinetic parameters:

Y.-C. Lin, J. Cho, G. a. Tompsett, P. R. Westmoreland, G. W. Huber, Kinetics and Mechanism of Cellulose Pyrolysis. *J. Phys. Chem. C*. **113**, 20097–20107 (2009).

Charring rate:

P. B. Cachim, J. M. Franssen, Assessment of Eurocode 5 charring rate calculation methods. *Fire Technol.* **46**, 169–181 (2010).

Richter and Rein, in *8th European Combustion Meeting* (2017).

The following material, up to the conclusions, is extracted from these two journal papers:

Bal and Rein, *Fire Safety Journal* , 2013
http://dx.doi.org/10.1016/j.firesaf.2013.08.015

Fire Safety Journal 61 (2013) 36–44

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Relevant model complexity for non-charring polymer pyrolysis

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ABSTRACT

The choice of the heat, mass and chemical mechanisms included in a pyrolysis model is often subjective, and detailed justifications of the inclusion or exclusion of the different mechanisms are infrequent. The implicit assumption that models with a higher number of mechanisms reproduce more accurately the reality has led to the recent growth of complexity in pyrolysis modelling seen in the literature. However, as we show in this work, the comparison of several conceptual models predicting the same experimental results does not support this assumption, but reveals the presence of unnecessary complexity and multiple sources of uncertainty. Using a novel approach corresponding to a mechanism sensitivity, the influence of the heat, mass and chemical mechanisms on the transient predictions of surface temperature and mass loss rate (non-flaming conditions) for PolyMethylMethAcrylate (PMMA) samples is investigated. While a small change in the chemical degradation mechanism has a large effect on the predictions of the mass loss rate, the surface temperature is not affected. The heat transfer mechanisms appear to have however a significant effect on both quantities of interest. This study demonstrates that the use of complex chemical mechanisms (e.g. multi-step reaction scheme or oxidative reaction) is not justified if the mechanisms of the heat transfer are kept simple. It is therefore recommended to use consistent levels of crudeness dictated by the heat transfer.

1. Introduction

While the choice of a conceptual[1] model is a major step in the prediction of a phenomenon, its justification is often subjective, and detailed justifications of inclusion or exclusion of the different mechanisms are infrequent. Assumptions and simplifications decrease the complexity of a model and define somehow its limitations. The implicit hypothesis that models with a higher number of mechanisms reproduce more accurately the reality has led computational modelling in general and fire modelling in particular to undergo recently a large growth in complexity [2–4].

Chwif et al. [5] have listed some of the reasons for this growth in complexity. Among the non-technical reasons, they highlight the "include all" syndrome and the "possibility" factor. The former is the consequence of the inexperience of the modellers who might feel insecure and include the maximum number of mechanisms just in case. The latter is due to the increasing computational power available which makes possible to include a significantly higher number of mechanisms without increasing the running time. One example of the technical reasons listed is the willingness of gathering the needs of several users, thus increasing the scope of the model and the number of mechanisms.

However, as the global level of complexity increases in models, the number of input parameters required increases as well. These parameters could be, for example, physical properties (or effective properties), mathematical constants, experimental constants or calibration factors, and all carry some degree of uncertainty. Their respective uncertainty accumulates in the model and contributes to the global uncertainty associated with the numerical predictions. The discrepancy between the experiments and the predictions is a combination of errors due to the lack of important mechanisms (continuous line in Fig. 1) and the parameter uncertainty (dashed line in Fig. 1) [6,7].

An equilibrium is therefore required between the error related to the simplicity of the model equations and the prediction uncertainty in order to find an appropriate level of model complexity as shown in Fig. 1. The parameter uncertainty can be reduced by a calibration process, decreasing the resulting

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[1] Note the difference between a conceptual model and its programmed code version. According to the ISO standard 16730:2008 [1] a conceptual model is the "description of all the information, mathematical modelling data and mathematical equations that describe the physical system or process of interest". It is called model hereafter for simplicity.

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Bal and Rein, Fire Safety Journal , 2015
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On the effect of inverse modelling and compensation effects in computational pyrolysis for fire scenarios

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ABSTRACT

Polymer pyrolysis is a key phenomenon in solid ignition, flame spread and fire growth. It is therefore an essential part in the understanding of fire behaviour. Advances in computational pyrolysis during the last decade have mainly resulted in an increase of the number of physical and chemical mechanisms implemented in the models. This stems from the implicit assumption that models with a higher level of complexity should be more accurate. However, a direct consequence of this growth in complexity is the addition of new parameters and the accumulation of modelling uncertainty born from the lack of knowledge of their values. The large number of parameters and the difficulty to quantify their values from direct measurements often oblige modellers to solve an inverse problem to perform the calibration of their models. By doing inverse modelling, the equations and the experimental data are consequently coupled to the parameter values found. This coupling and its consequences, which are most often ignored, are investigated here using different levels of model complexity for the simulation in Gpyro of transient pyrolysis of PolyMethylMethAcrylate in non-burning conditions. Among the wide range of possible model complexities, five models with a number of parameters ranging from 3 to 30 are considered. It is observed that models of different complexities (i.e. number of mechanisms and associated assumptions) can achieve similar levels of accuracy by virtue of using different parameters values. The results show the strong presence of multiple compensation effects between implemented mechanisms (e.g. chemistry or heat transfer), and that an increase of model complexity can induce a large scatter in the parameters values found. We recommend the use of larger data sets from different experimental procedures (e.g. different boundary conditions) and of different nature (e.g. in-depth temperature profile instead of only surface temperature) to break down the compensation effects found in this study.

1. Introduction

Despite the extensive use and constant development of fire modelling tools, the current state of the art is still not capable of predicting fire growth rate from first principles. The pyrolysis process of the condensed phase represents one of the main challenges related to this problem. It is a key phenomenon in solid ignition, flame spread and therefore in the global understanding of fire behaviour. Advances in pyrolysis modelling during the last decade have mainly resulted in an increase of the number of physical and chemical mechanisms implemented in the models. This stems from the implicit assumption that models with a higher level of complexity should be more accurate. However, a direct consequence of this growth in complexity is the addition of new parameters. The uncertainty associated with each of these parameters is propagated to the output via their sensitivity [1]. Examples are available in Refs. [2,3]. The growth of complexity might influence therefore the global modelling uncertainty.

The predictive capabilities of acomputational model isfunction on three main components: the equations of the model, the input parameters, and the experimental data used to validate the model. The equations, directly function of the assumptions performed, describe mathematically the physical and chemical mechanisms which are then solved in time based on the assumed boundary conditions. The input parameters are a set of values required to perform a simulation. This set is composed of material properties (e.g. kinetic triplet, attenuation coefficient), initial and boundary conditions (e.g. incident heat flux, sample thickness, convective cooling) and mathematical artefacts (e.g. grid size and time steps). The experimental data is made of the measurements to which the model predictions are to be compared (e.g. mass loss rate, surface temperature and in-depth temperature profile).

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Computational Pyrolysis

Energy conservation: $$\rho c \frac{\partial T(y,t)}{\partial t} = k \frac{\partial^2 T(y,t)}{\partial y^2} + (1-\phi)(1-r)(1-\gamma)\dot{q}''_e \kappa e^{-(\kappa y)}$$

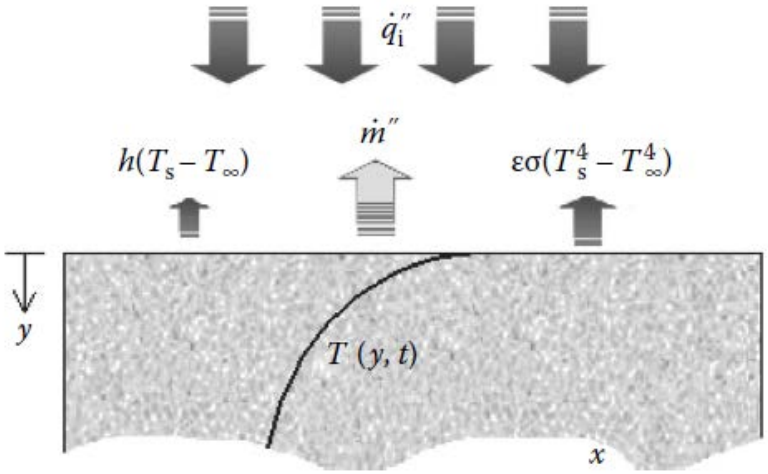
Multi-step reaction scheme:

$$\mathrm{PMMA} \rightarrow \beta\mathrm{PMMA}$$

$$\beta\mathrm{PMMA} \rightarrow \mathrm{GAS}$$

$$\beta\mathrm{PMMA} + \upsilon_{O_2} O_2 \rightarrow \upsilon_{GAS}\mathrm{GAS}$$

Properties
k [W/m K]
c [J/kg K]
ρ [kg/m3]
κ [1/m]
ε [dimensionless]
log (A) [1/s]
E_a [kJ/mol]
n [dimensionless]
ΔH [kJ/g]



Boundary conditions:

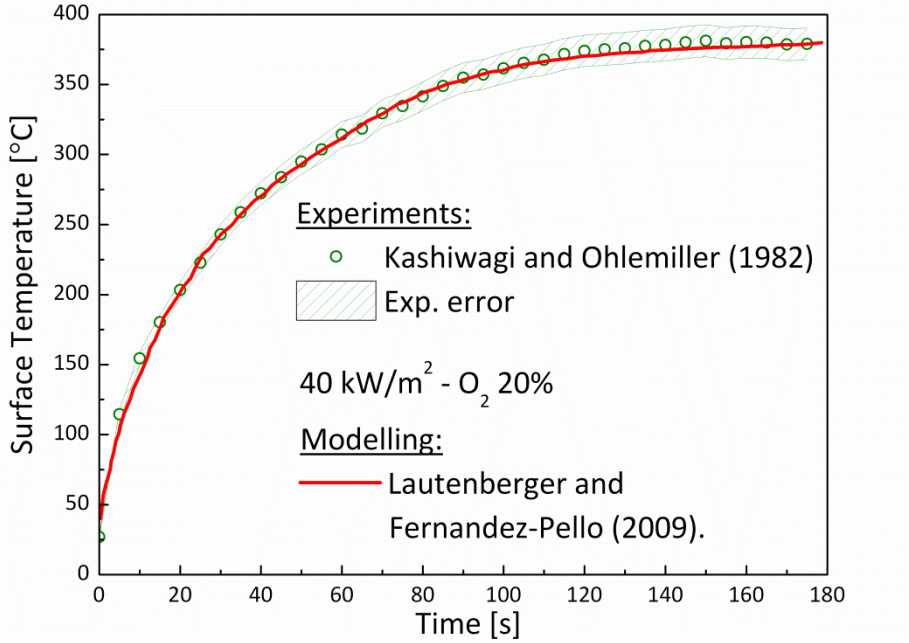
$$\forall t; -k\left.\frac{\partial T}{\partial y}\right|_{y=0} = (1-\phi)(1-r)\gamma\dot{q}''_e - h(T_S - T_0) - \sigma\varepsilon(T_S^4 - T_0^4)$$

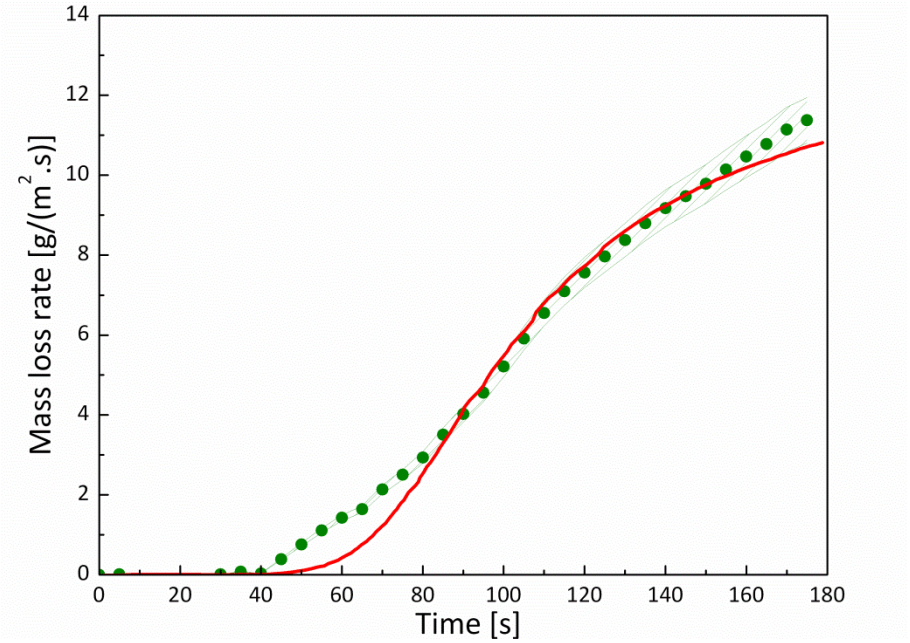
$$\forall t; \left.\frac{\partial T}{\partial y}\right|_{y=L} = 0$$

$$\forall y; T(y, t=0) = T_0$$

Bal and Rein, *Fire Safety Journal* , 2013 http://dx.doi.org/10.1016/j.firesaf.2013.08.015

Measurements vs. Predictions





- Clear PMMA
- Non-flaming conditions
- Vertical exposition
- Small scale apparatus

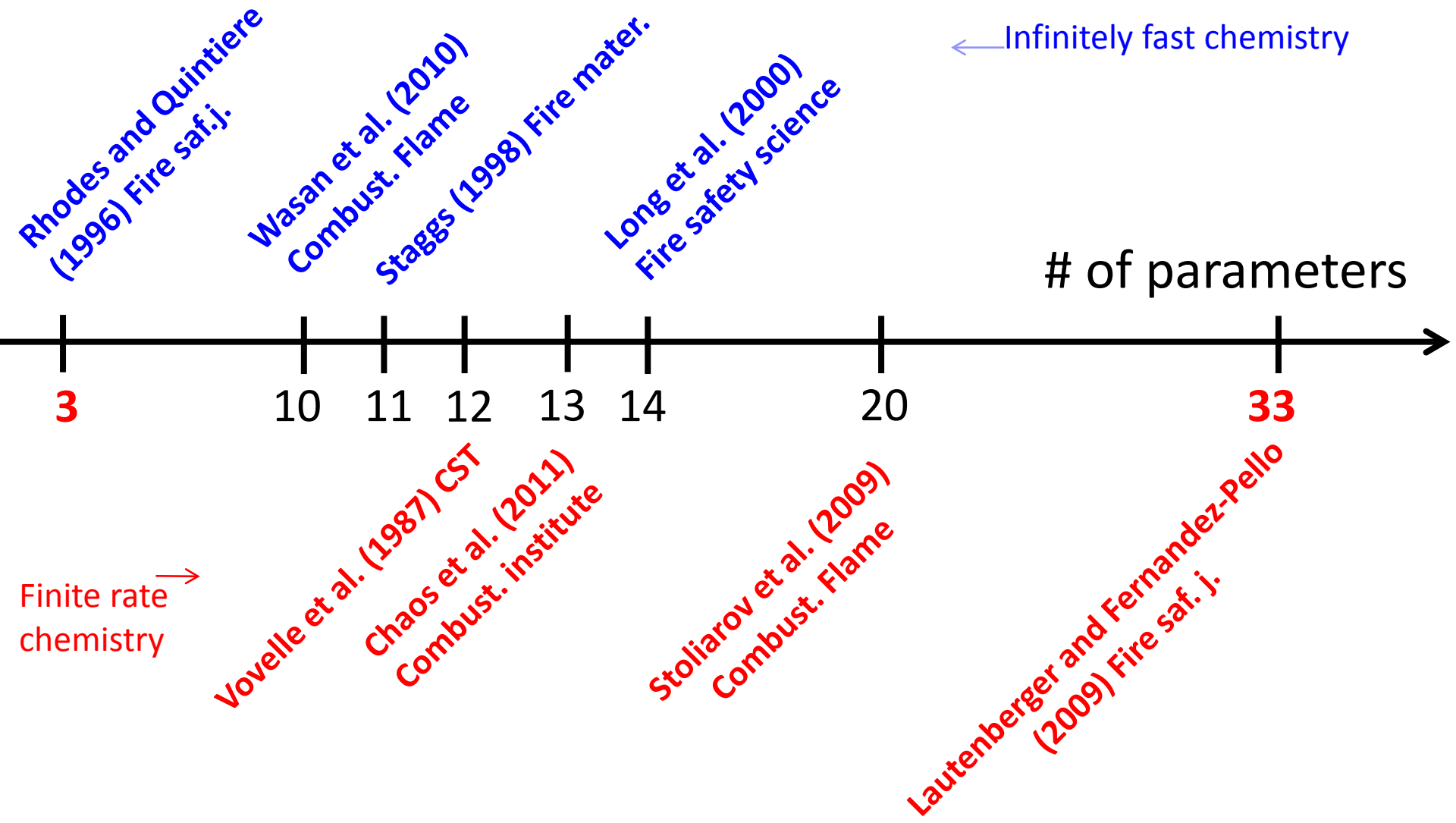
Kashiwagi and Ohlemiller, Proceedings of the Combustion Institute, 1982

Lautenberger and Fernandez-Pello, Fire Safety Journal 2009

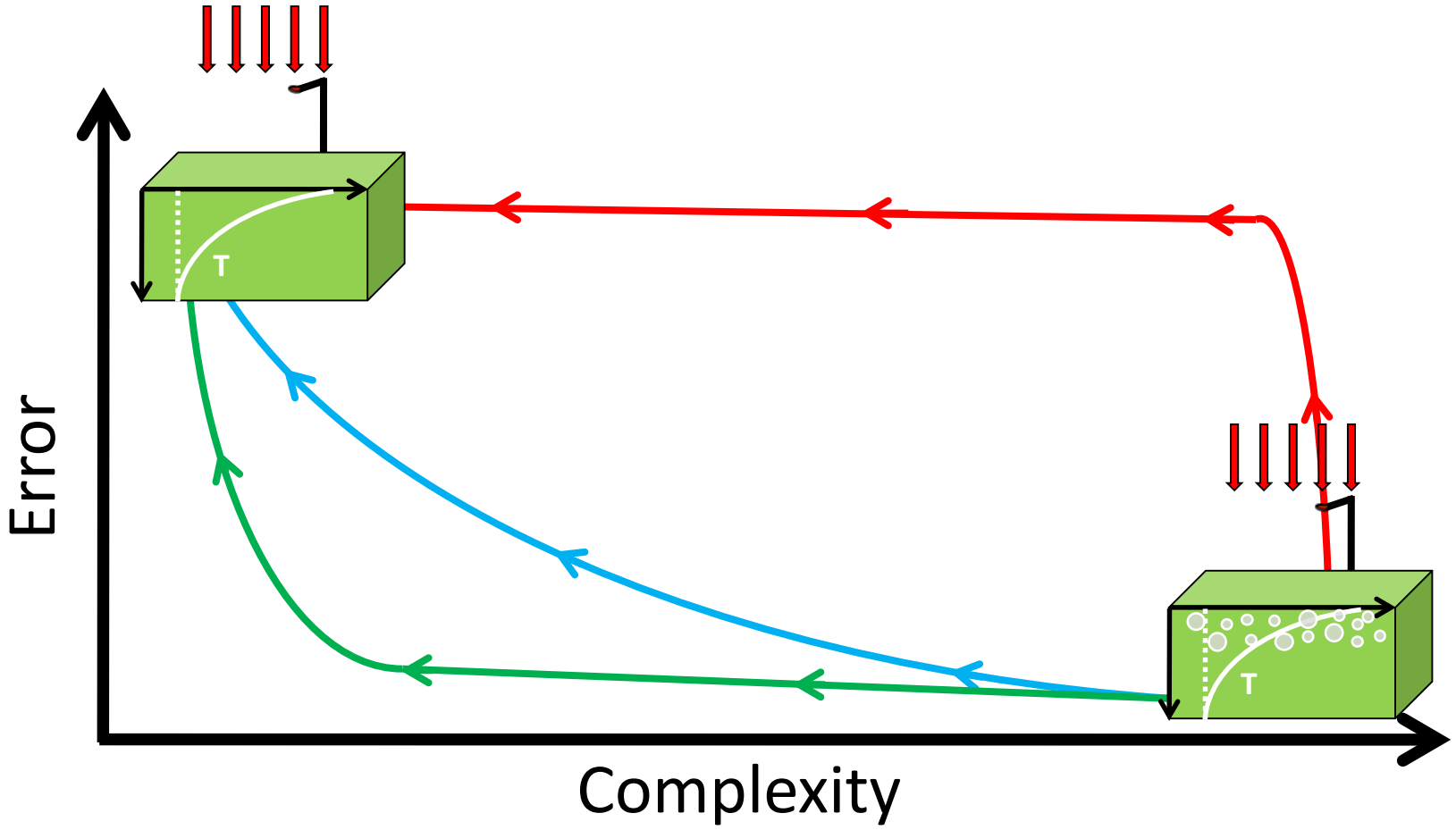
Bal and Rein, *Fire Safety Journal* , 2013 http://dx.doi.org/10.1016/j.firesaf.2013.08.015

Complexity Growth

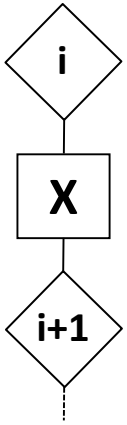
Models for PMMA:



Error due to incomplete mechanisms



Mechanism sensitivity: methodology



Model M_i

Assumption X

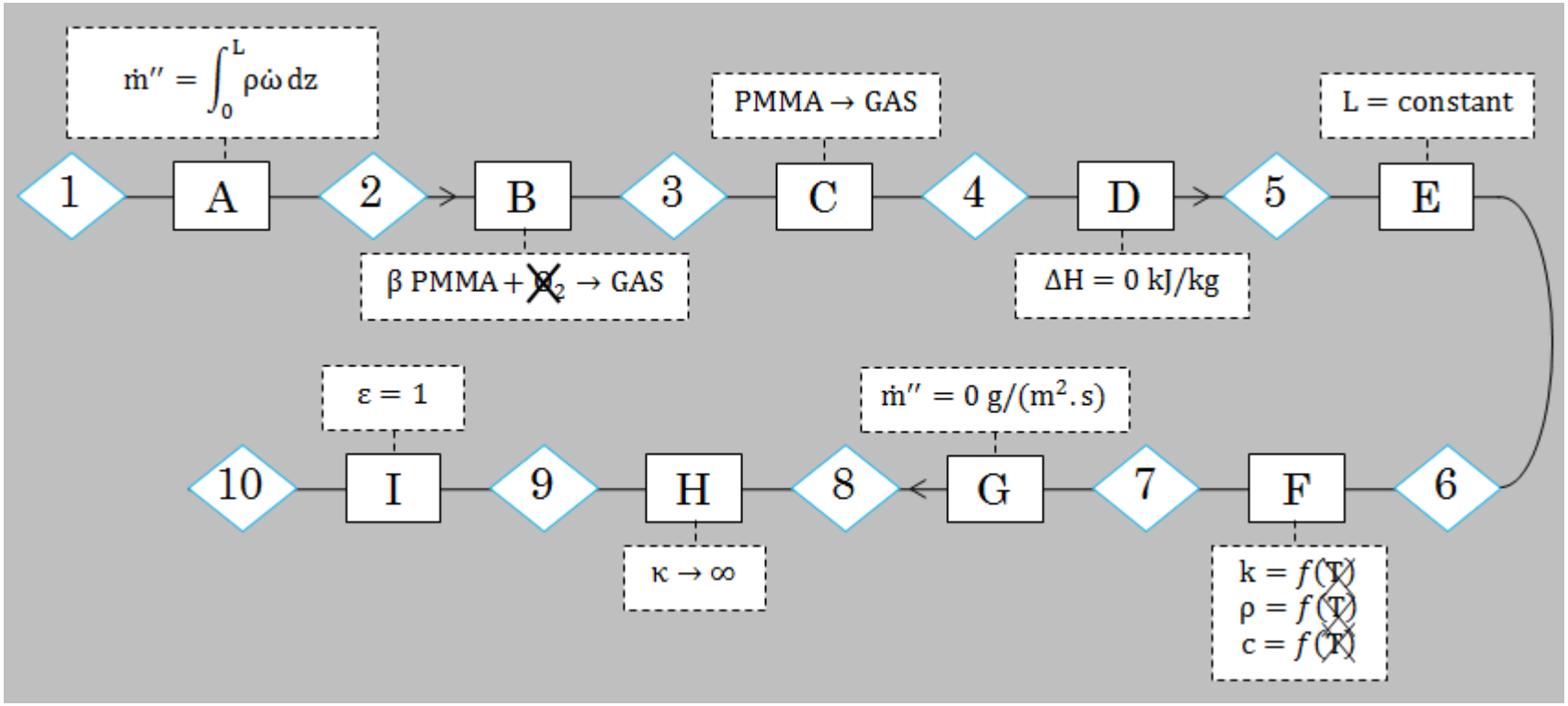
Model M_{i+1}

Mainly two types of assumptions:

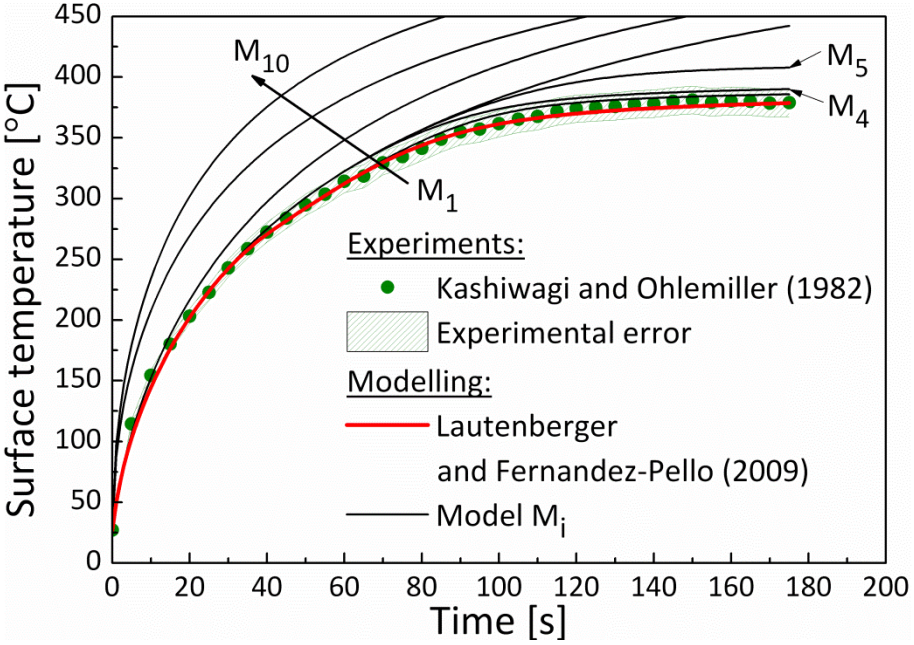
Heat transfer assumptions

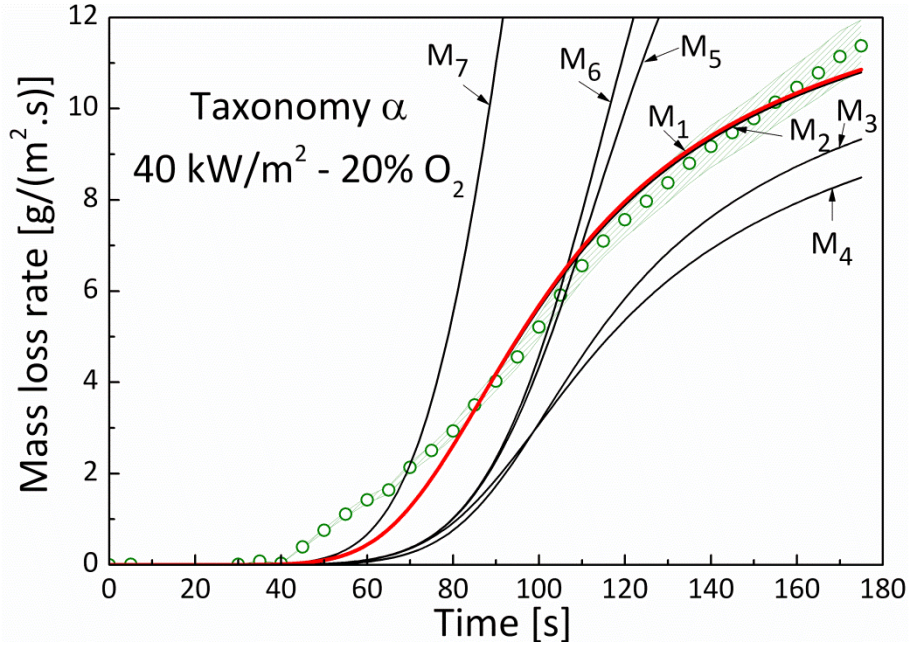
Chemistry assomptions

Taxonomy α



Mechanism sensitivity: qualitative results





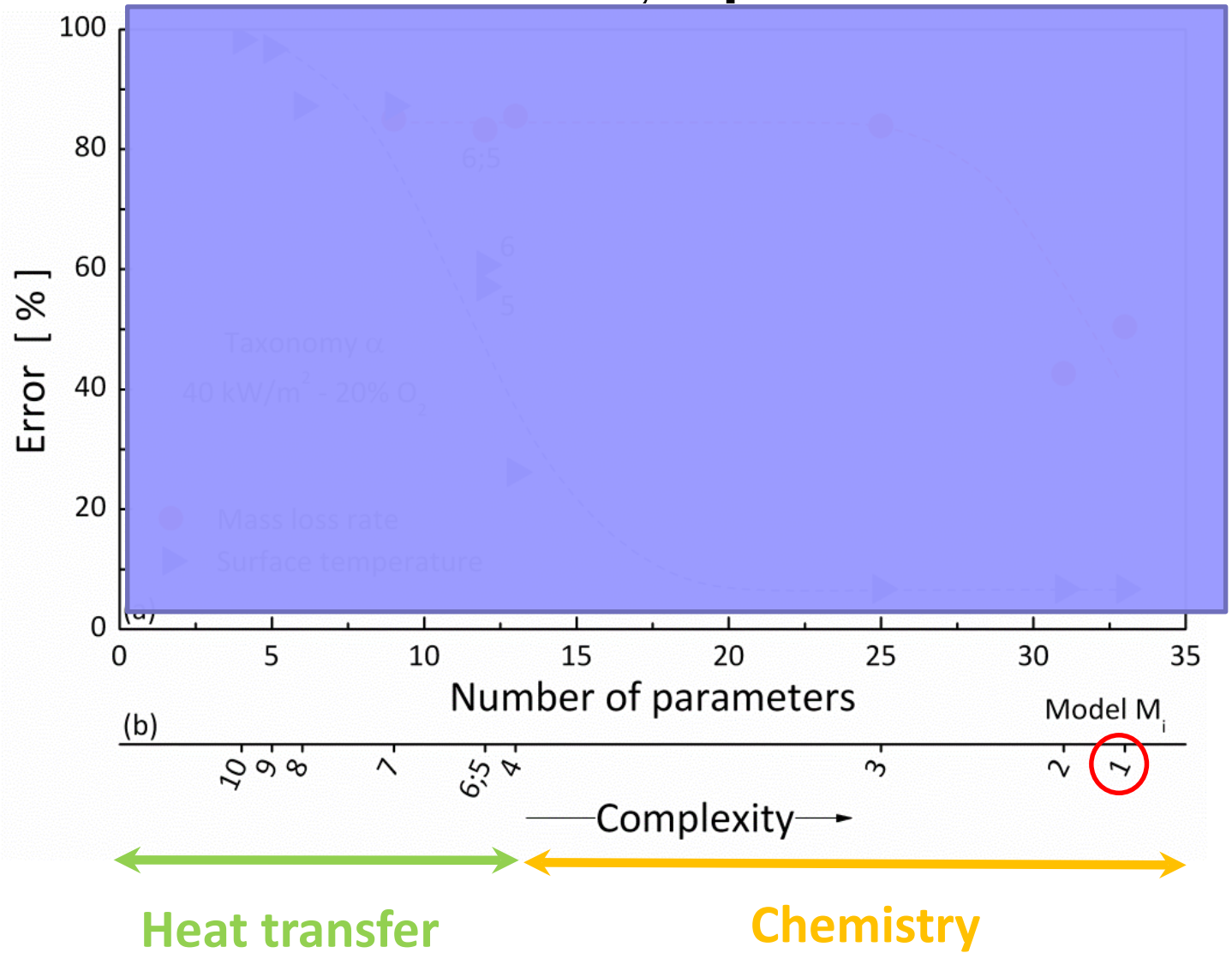
Surface temperature:

- T_S not affected up to M_4 ($M_5 \equiv [\Delta H=0]$)
- Heat transfer assumptions induce substantial over-estimations

Mass loss rate:

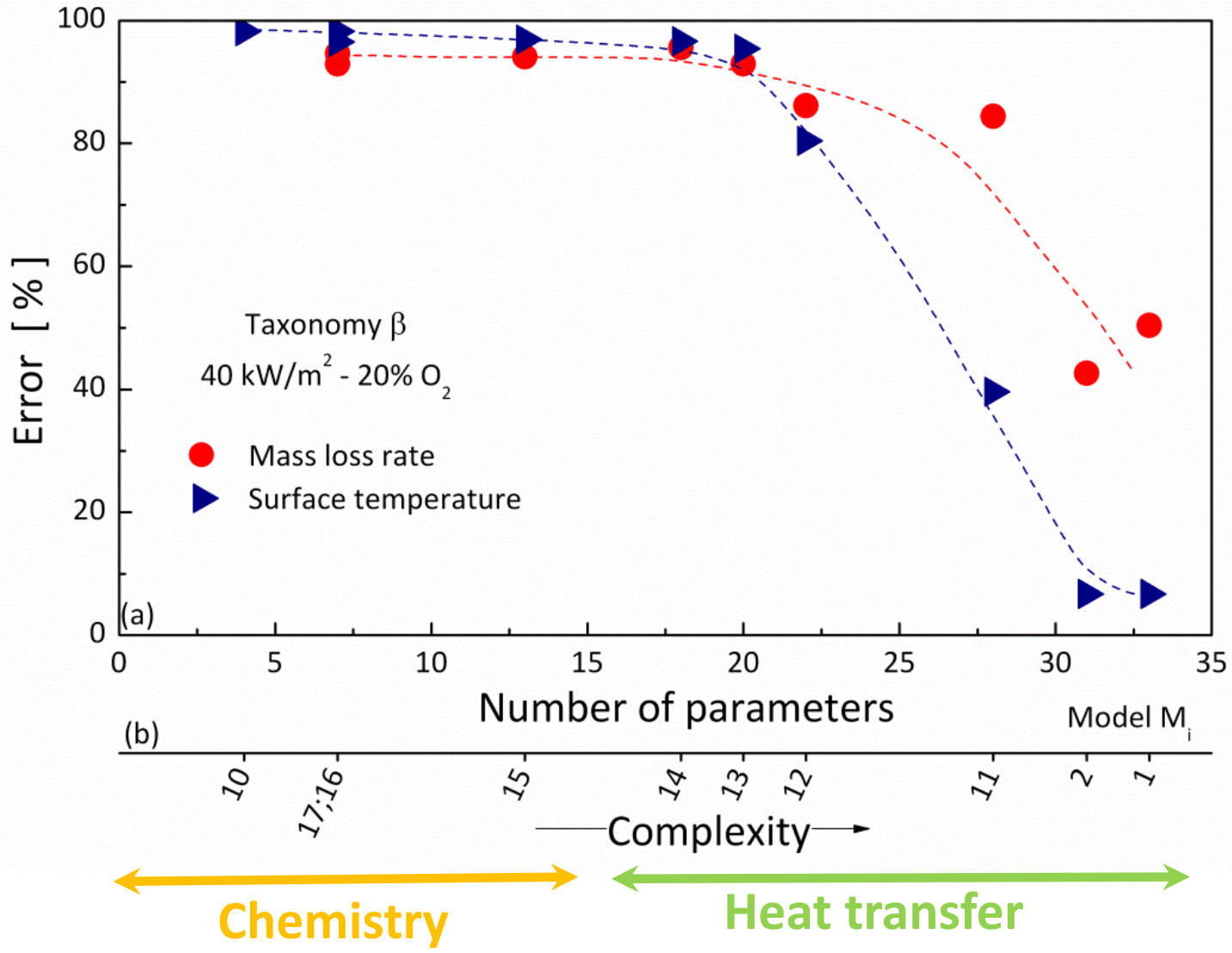
- MLR affected as soon as the reaction scheme is changed (M_3)
- MLR shape drastically changed for $M_5 \equiv [\Delta H=0]$.

Mechanism sensitivity: quantitative results



This taxonomy does not allow to evaluate influence of heat transfer on MLR

Mechanism sensitivity: new taxonomy



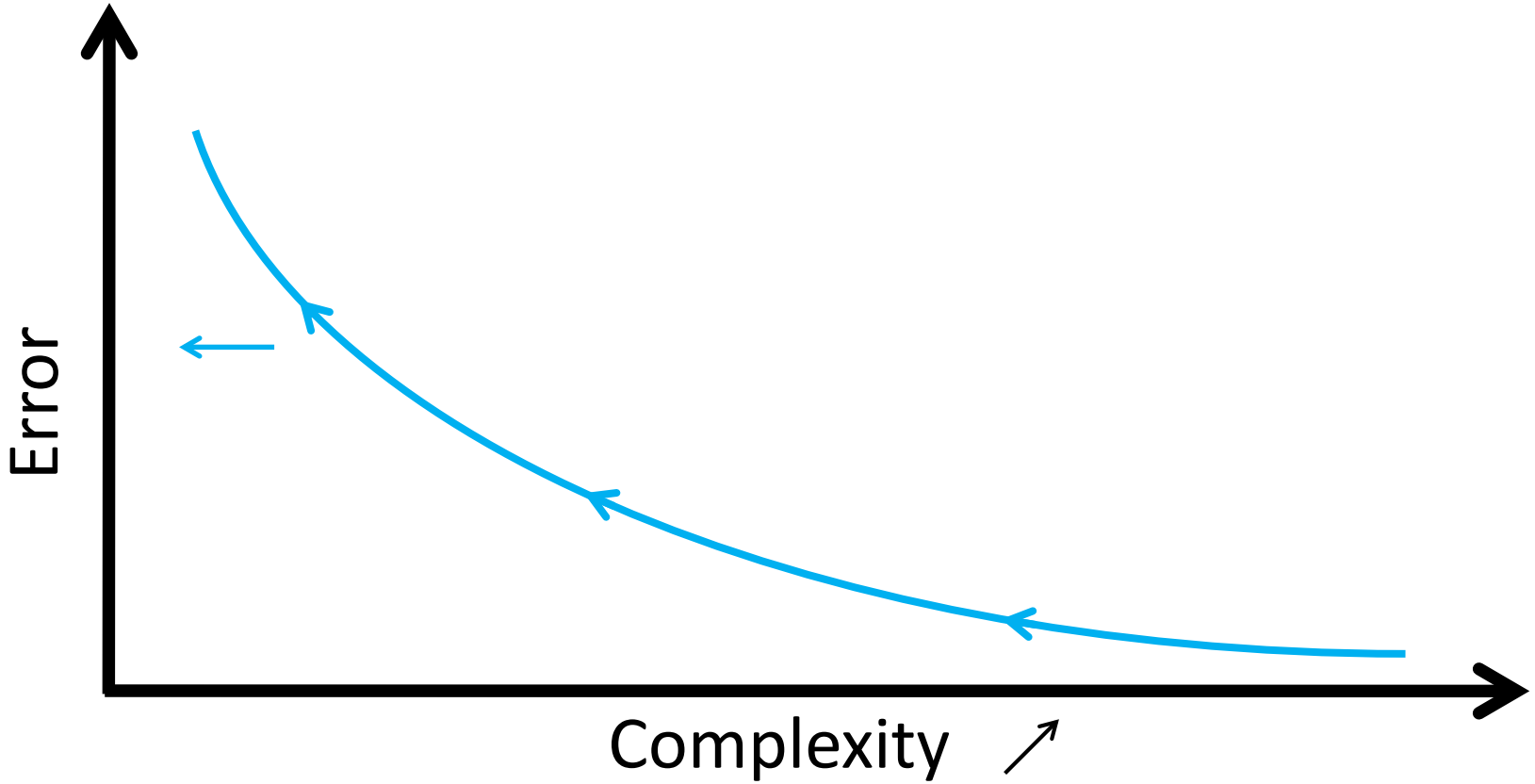
New Taxonomy β: Heat transfer assomptions made prior to Chemitry assumptions

With full chemistry, error on T_S increases to 100% with heat transfer assumptions alone.

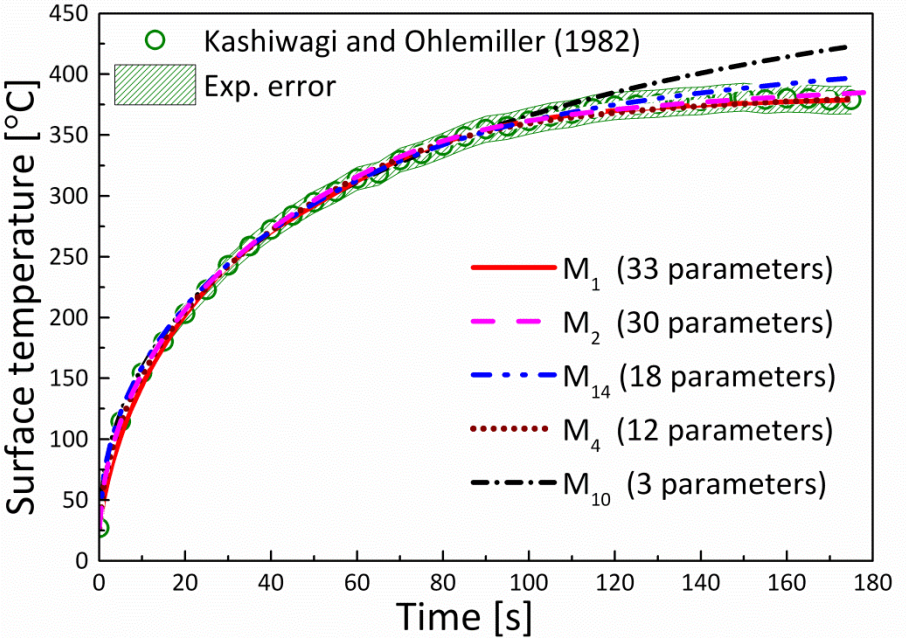
Accuracy of the predictions related to the crudeness of heat transfer mechanisms

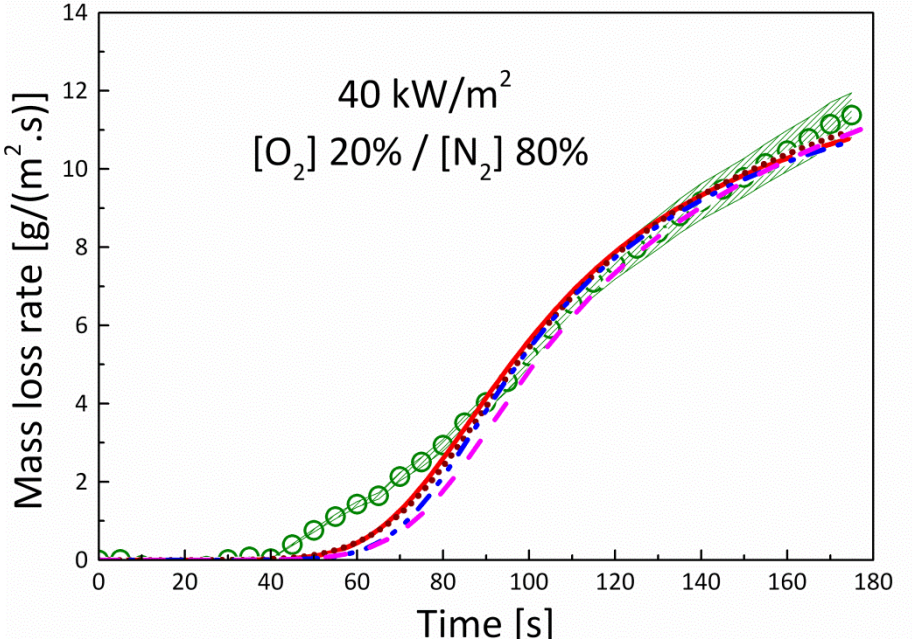
Balance between model complexity and uncertainty

For low level of complexity, the prediction accuracy is controlled by the lack of important mechanisms.



Calibration by inverse modelling: results





- For M_1, M_2 and M_4, different sets of values provide similar prediction of T_s and MLR.
  → For different level of complexity, the same accuracy can be obtained (compensation effects).

- The best fit of M14 and M10 do not manage to predict Ts >370 °C.
  → The calibration cannot always reduce the prediction error.

Calibration by inverse modelling

Assumption invoked	Models				
	M_1	M_2	M_{14}	M_4	M_{10}
Gases released instantaneously: no momentum equation		✓	✓	✓	✓
Heat of pyrolysis negligible: $\Delta H = 0$ kJ/kg			✓		✓
1-step chemical degradation: PMMA → GAS				✓	
Inert solid: no kinetics, no mass loss					✓
Constant thermo-physical parameters: $\gamma_\varphi = 0$			✓		✓
Radiation absorption at the surface: $\varepsilon = 1, \kappa \to \infty$			✓		✓
Number of input parameters	33	30	18	12	3

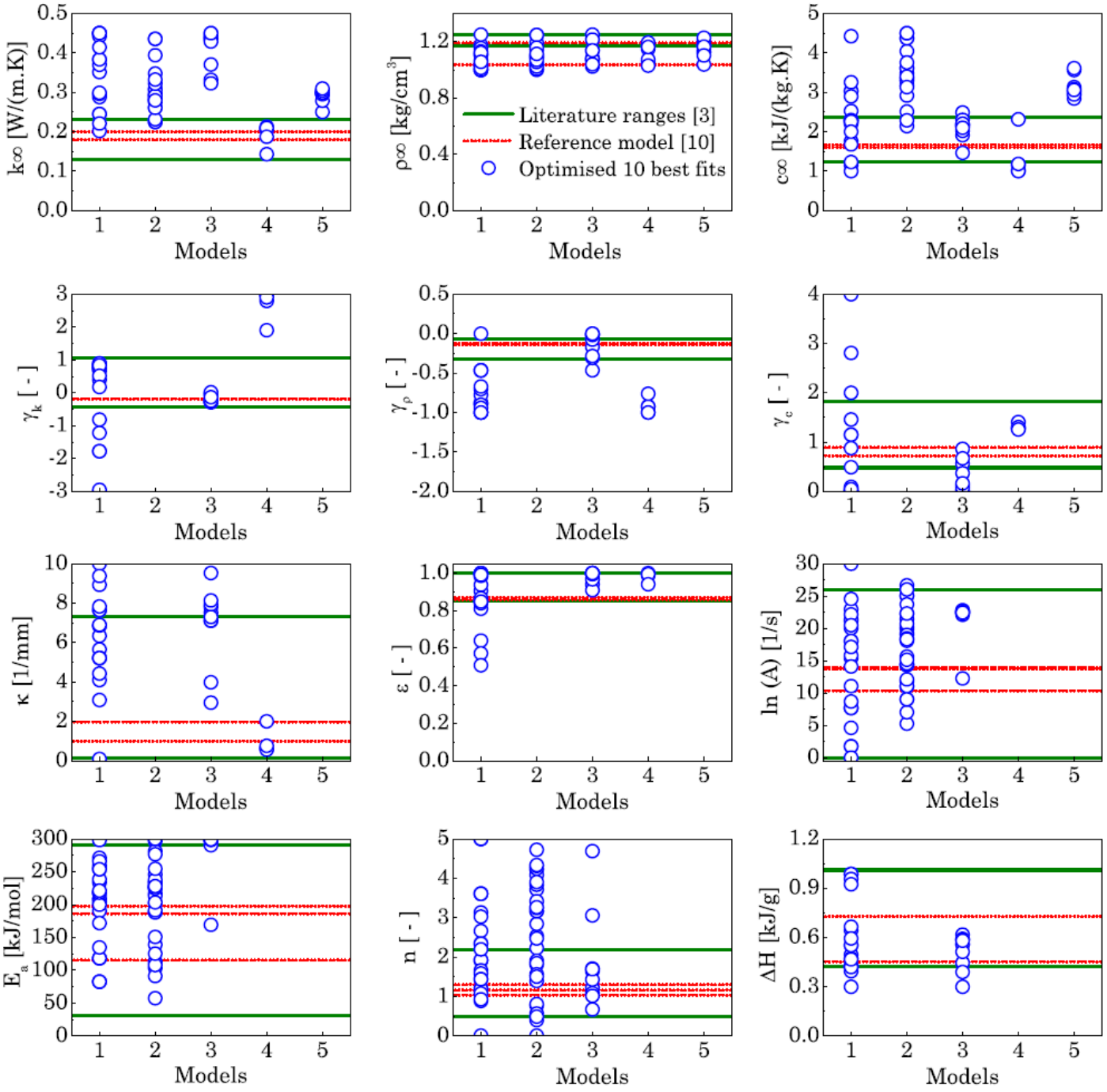
M_1 Most complete model (Lautenberger and Fernandez-Pello, 2009 Fire Saf. J.)

M_2 = M_1 without momentum conservation

M_{14} = M_2 without detailed heat transfer

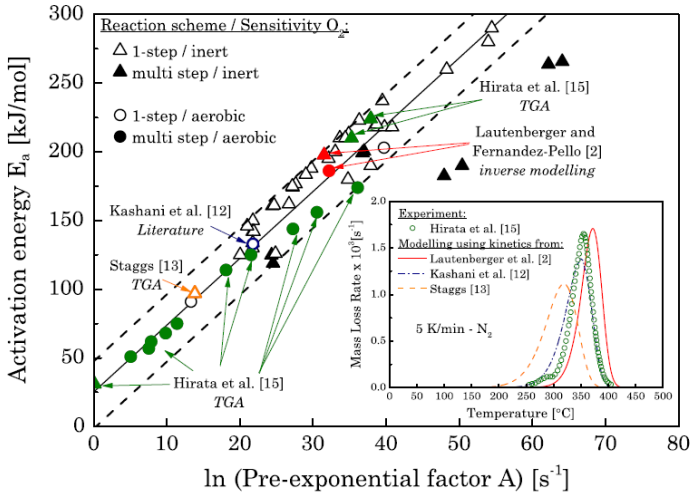
M_4 = M_2 with 1-step reaction scheme

M_{10} = inert solid without detailed heat transfer


k∞ [W/(m.K)]
ρ∞ [kg/cm³]
c∞ [kJ/(kg.K)]
γk [-]
γρ [-]
γc [-]
κ [1/mm]
ε [-]
ln (A) [1/s]
Ea [kJ/mol]
n [-]
ΔH [kJ/g]
Models
Literature ranges [3]
Reference model [10]
Optimised 10 best fits

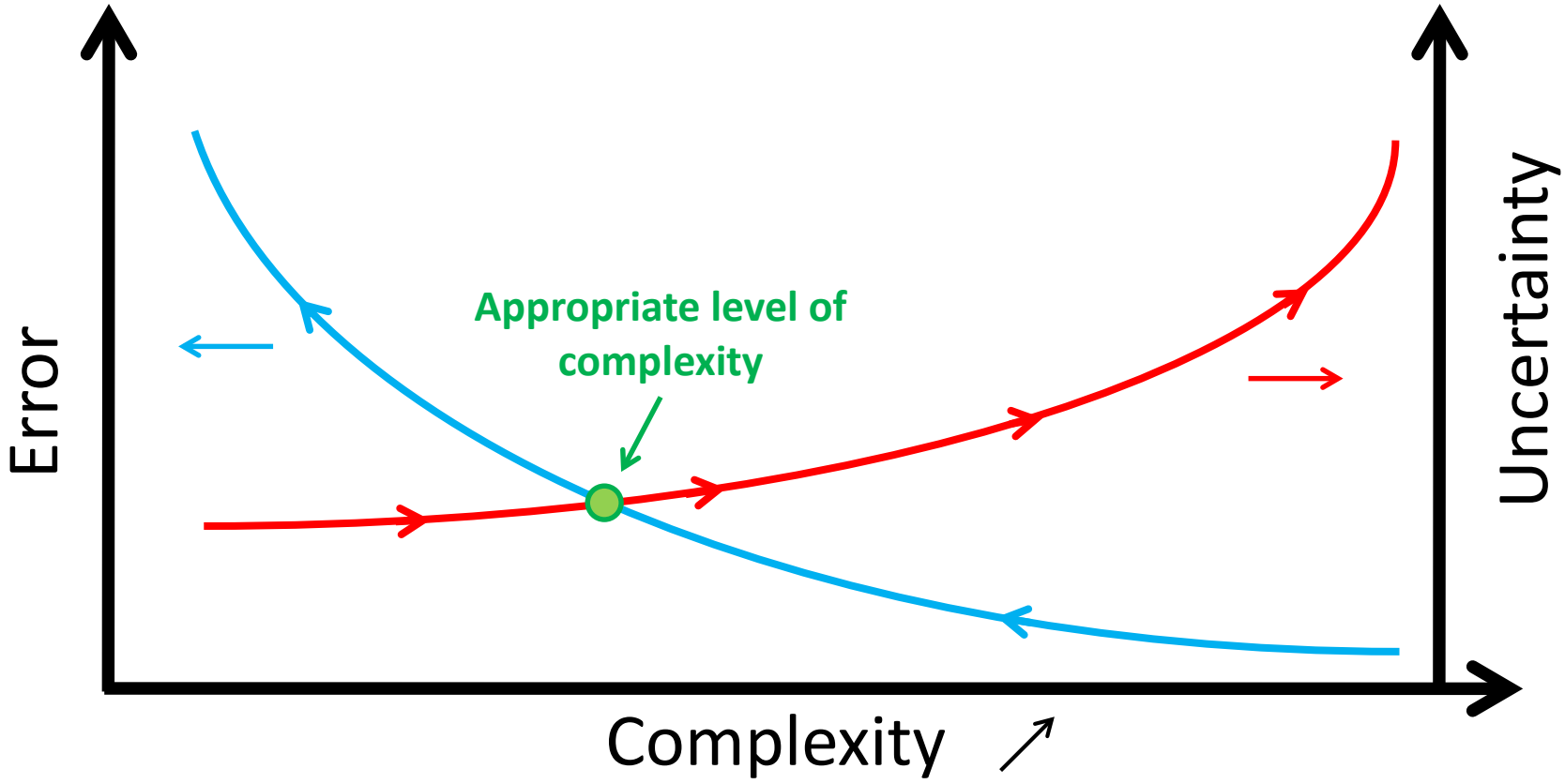
Balance between model complexity and uncertainty

For high level of complexity, the prediction accuracy is controlled by the input parameter uncertainty.



Balance between model complexity and uncertainty

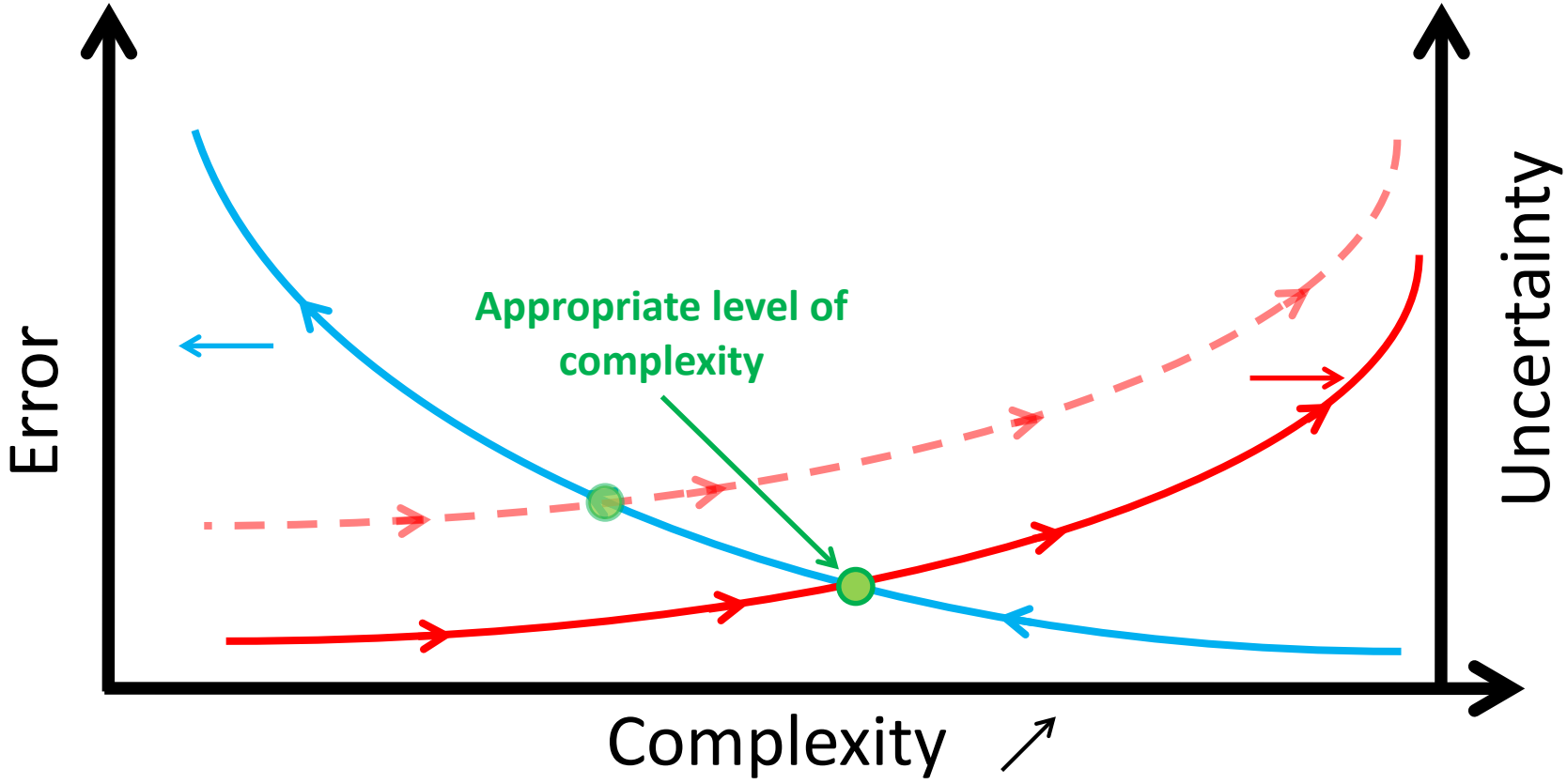
Here be appropriate level of complexity …



Balance between model complexity and uncertainty

Here be appropriate level of complexity …

which evolves with the size of experimental data set.



Concluding Remarks

- Pyrolysis is a function heat transfer and chemical kinetics. But accuracy of the predictions related to the crudeness of heat transfer. **Keep chemistry as simple as heat transfer**.
- Balance needed between **model complexity** and modelling uncertainty.
- This balance depends on the quantity and quality of the experimental data available.

Poor and Scarce data = only simple models are justified

Good and Abundant data = more complex models are justified

- Corollary: we **need better understanding** of pyrolysis so we can provide better predictive tools of fire.

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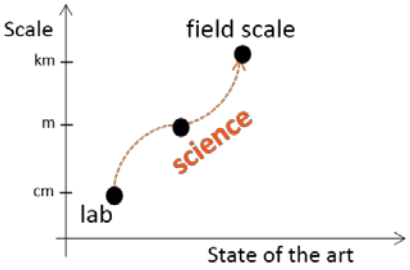

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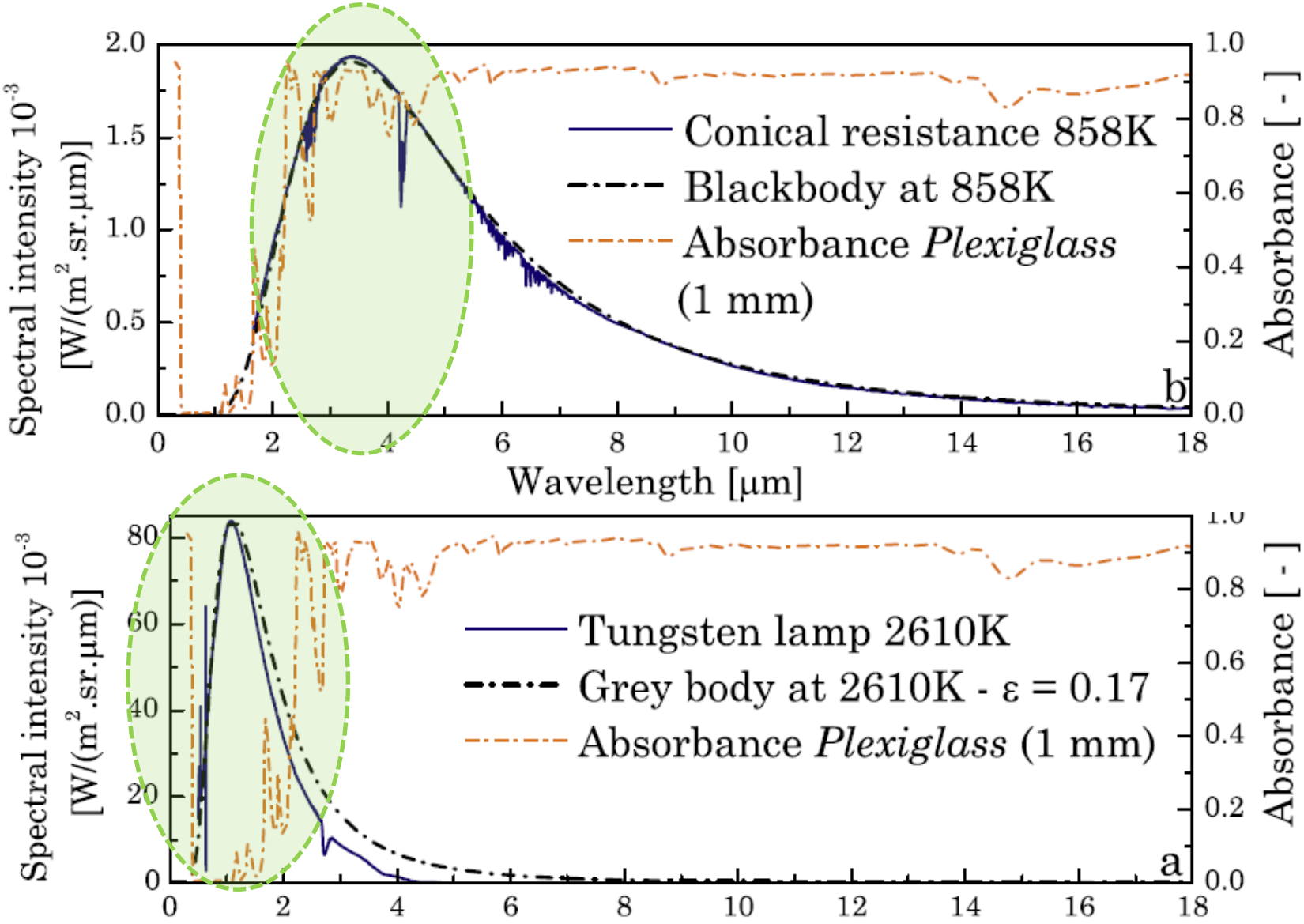
bre Met Office

Experimental Pyrolysis – Spectral sources

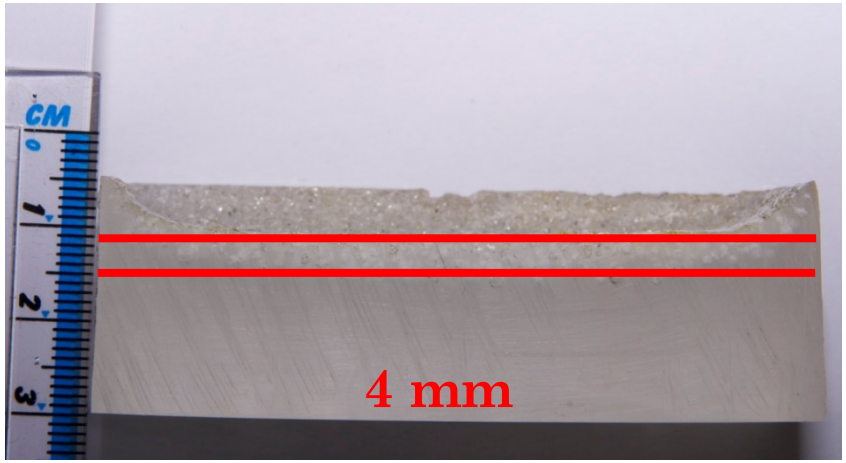


Bal, et al. , *International Journal of Heat and Mass Transfer* 61, pp. 742–748, 2013.
http://dx.doi.org/10.1016/j.ijheatmasstransfer.2013.02.017

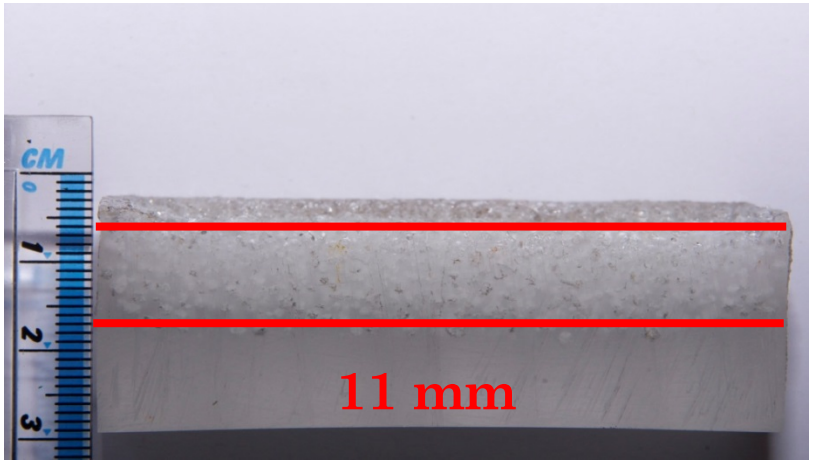
Experimental Pyrolysis – PMMA Unexpected slower ignition when radiation source is changed

According to the state of the art of fire science, the pyrolysis behaviour of the PMMA sample should had been exactly the same under the two heat sources.
What is causing this repeatable observation?

Exposed to 20kW/m^2 in **cone calorimeter**



Exposed to 20kW/m^2 **tungsten lamps**



Girods et al., *Fire Safety Science* 10: 889-901, 2011. http://dx.doi.org/10.3801/IAFSS.FSS.10-889