An Imprecise Bayesian Approach to Thermal Runaway Probability

Marc Fischer
Mines de Saint-Étienne, CNRS, UMR 5307 LGF, Centre SPIN, F-42023 Saint-Étienne, Université de Lyon, France

Alexis Vignes
INERIS, Parc Technologique ALATA, B.P. 2, F-60550 Verneuil-en-Halatte, France

Abstract

In this pioneering work, an assessment of thermal runaway probability based on simplified chemical kinetics has been performed with imprecise Bayesian methods relying on several priors. The physical phenomenon is governed by two chemical kinetic parameters $A$ and $E_a$. We suppose that their values are considerably uncertain but also that we know the experimental profiles of a chemical species corresponding to their true values, thereby allowing us to compute likelihoods and posteriors corresponding to different levels of information. We are interested in the critical delay time $t_c$ beyond which an explosion will certainly occur. The use of several priors allows us to see when the data truly dominate the prior with respect to the probability distribution of $t_c$. It does not appear possible to do so in an orthodox precise Bayesian framework that reduces all forms of uncertainty to a single probability distribution.

Keywords: Robust Bayesianism, explosions, chemical kinetics, principle of indifference

1. Introduction

Explosions remain a very serious threat in the industrial world (Atkar and Jabbari, 2013; Skob et al., 2020; Ahmed et al., 2012). From a general point of view, an explosion can be defined as a sudden increase in pressure and temperature stemming from an oxidation or other exothermic reactions. They are a complex phenomenon emerging from the interplay of chemistry, heat transfers and fluid dynamics. To make predictions, it is possible to rely either on phenomenological approaches (Proust, 2005) or on CFD (Computational Fluid Dynamics) tools (Ferrara et al., 2006). These CFD tools require a fundamental knowledge of the combustion characteristics of the substances. However, these characteristics are often highly uncertain because of the lack of relevant experimental data and also the absence of standardised experimental approaches to determining fundamental parameters (e.g. laminar flame speeds, oxidation kinetics at high temperatures). Another difficulty which arises when assessing explosion probabilities consists of the factors the thermal runaway process depends upon.

In order to estimate the probability of an explosion in a given situation, we must take into account both the probability distributions of the initial and boundary conditions (aleatory uncertainty) and the uncertainties of the physical and chemical parameters of the model (epistemic uncertainty). For that sake, classical (precise) Bayesian methods are increasingly being employed but they suffer from their inability to properly consider the difference between aleatory and epistemic uncertainty (Schöbi and Sudret, 2019; Ferson and Oberkampf, 2009; Mathon et al., 2010). Routinely, explosion hazards are assessed through the determination of explosion safety parameters according to various standards (e.g. EN 1839:2017 (EN et al., 2017), ISO 10156:207 for gases and vapours (Zakel et al., 2019)).

However, when it comes to determining explosion probabilities in a well-defined hazardous scenario by measuring ignition-sensitive parameters (e.g. minimum ignition energy, auto-ignition temperature), it should be noted that the risk and safety engineer can only access values which correspond to a maximised explosion probability. The explosion probability can thus be potentially considerably overestimated. In order to estimate more realistic probabilities, we would either have to perform costly tests or to use our more fundamental knowledge of oxidation kinetics from low to high temperatures. The advantage of such a chemical kinetic approach is that it can be applied to a wide range of industrial scenarios (Warnatz et al., 2017; Peters and Rogg, 1993).

Nevertheless, the chemical kinetic parameters are then the main source of epistemic uncertainty in explosion hazard assessments. They are generally unknown but constrained by physical bounds and a set of experimental data the complete model must be able to reproduce well enough (Shen et al., 2017; Fischer, 2019).

Up until now, most authors who sought to estimate explosion probabilities did not consider the chemical kinetic parameter uncertainties but designed their approach at a macro-level. Ronza et al. (Ronza et al., 2007) and Moosemiller (Moosemiller, 2011) used event trees based on historical data and expert knowledge to predict explosion probabilities. Many researchers apply Bayesian networks or fuzzy Bayesian networks to the assessment of explosion hazard by eliciting expert knowledge or by using the known
Applied to a continuous variable such as a pre-exponential factor $A$, the POI orders us to use a uniform prior $f_0(A)$. However, since we are equally ignorant about $1/A$, $\log_{10}(A)$ and $1/\log_{10}(A)$, and many other deterministic functions of $A$, we should also use priors which are flat with respect to these variables and highly non-uniform with respect to $A$. In their excellent work devoted to chemical parameter uncertainty propagation, (Frenklach et al., 2007) approvingly stated statisticians Box and Hunter’s recommendation for defining priors in chemical kinetics:

In considering a parameter like the specific rate [
constant] $\varphi$ which is essentially positive, it is probably most realistic to take $\theta = \ln \varphi, -\infty \leq \theta \leq 0$, as locally uniform a priori. This would mean, for example, that having guessed a value of $\varphi$, an experimenter would be about equally prepared to accept a value twice as big as he would to accept a value one-half as big.

At first glance, this advice would sound quite reasonable to most chemical kinetics. However, if the authors had also mentioned some logical implications of their approach such as: “In the absence of any kind of knowledge about $\varphi$ except its positivity, we should all feel completely confident that $\Pr(1 \leq \varphi \leq 100) \approx 467.51 \Pr(10,001 \leq \varphi \leq 10,100)$, even though both intervals have the same length.”, most practitioners would probably find that rule very strange and arbitrary. If we only know that $\varphi > 0$, how on earth can we deduce such a highly specific result?

The fundamental problem that uniform priors are no longer uniform upon reparametrisation has led Jeffreys to define a standard prior based on the Fisher information that remains the same for any other parameter that is a deterministic function of the first one (Kass and Wasserman, 1996). However, Jeffreys’ prior is usually bound to be (highly) non-uniform with respect to many parametrisations of the problem and it thus also illegitimately creates specific knowledge out of ignorance.

To overcome this problem, the field of imprecise Bayesianism (also called robust Bayesianism or Bayesian sensitivity analysis) chooses to describe genuine ignorance through a wide variety of priors that results in a wide variety of posteriors (Valley, 2000; Berger et al., 1994; Insua and Ruggeri, 2012). This approach is being employed in an increasing number of fields such as insurance risks (Bozart, 2008), climate science (Tomassini et al., 2007), cybersecurity (Hallgren and Turcotte, 2020), and clinical trials (Greenhouse and Waserman, 1995), to name but a few. However, as Fischer pointed out (Fischer, 2019), chemical kinetists almost always only use one single uniform prior and do not feel concerned about the fact that their posterior might not be data-dominated. As Kass and Wasserman pointed out (Kass and Wasserman, 1996), the reliance on a single so-called uninformative prior is particularly dangerous if the experimental data available are insufficient to constrain the values of the model parameters, and Fischer showed an example where relying on only one flat prior would lead one to reject the model closest to the measurements at hand whereas the use of several priors reveals that this is a spurious and invalid conclusion (Fischer, 2019).
In this paper, we want to compare the use of classical precise Bayesian methods with an imprecise Bayesian approach relying on six different priors for assessing a thermal runaway probability. In Section 2, our methodology is explained. In Section 3, our results are presented and discussed. The article ends with a conclusion and the outlook in Section 4.

2. Methodology

2.1. Computation of the Delay Time Distribution

Our detailed method can be read in Appendix A: Delay time distributions. We first designed a risk scenario. We consider a container filled with gaseous propane C<sub>3</sub>H<sub>8</sub> in a closed room with a constant volume initially at atmospheric pressure. We suppose that the room is adiabatic, which means that we neglect the heat losses. Through a leak, a stoichiometric mixture of air-propane is formed in the room. Because of some incident, the ambient temperature jumps from 293.15 K (20 °C) to an initial temperature following a normal distribution: T<sub>0</sub> ∼ N(μ<sub>T0</sub> = 524 K, σ<sub>T0</sub> = 20 K). We have p(0 < T<sub>0</sub> < 440 K) < 1E-04 and p(0 < T<sub>0</sub> > 600 K) < 1E-04. As a consequence, we can always consider that the initial temperature belongs to the interval [T<sub>0,min</sub> = 440 K; T<sub>0,max</sub> = 600 K]. We are interested in how quickly the mixture reaches the critical temperature (also called ignition temperature in a technical context) T<sub>c</sub> = 766 K beyond which it would be impossible for a technician to intervene to stop the explosion (Reed, 1986). Let t<sub>c</sub> be the critical delay time defined as T(t<sub>c</sub>) = T<sub>c</sub>. If that time is elapsed, an explosion will surely occur. As explained in Appendix A: Delay time distributions, for given values of A and Ea, log<sub>10</sub>(t<sub>c</sub>) can be very well approximated by a linear function of 1/T<sub>0</sub>:

\[
\log_{10}(t_{c,A,Ea}(T_0)) \approx a_{A,Ea} \frac{1}{T_0} + b_{A,Ea}
\]  

(1)

with

\[
a_{A,Ea} = \frac{\log_{10}(t_{c,A,Ea}(T_{0,min})) - \log_{10}(t_{c,A,Ea}(T_{0,max}))}{1/T_{0,min} - 1/T_{0,max}}
\]

(2)

and

\[
b_{A,Ea} = \log_{10}(t_{c,A,Ea}(T_{0,min})) - a_{A,Ea} \frac{1}{T_{0.min}}.
\]

(3)

log<sub>10</sub>(t<sub>c,A,Ea</sub>(T<sub>0,min</sub>)) and log<sub>10</sub>(t<sub>c,A,Ea</sub>(T<sub>0,max</sub>)) are themselves to a large extent bilinear functions of log<sub>10</sub>(A) and Ea so that they can be well approximated by a piecewise bilinear interpolation. For that sake, log<sub>10</sub>(t<sub>c,A,Ea</sub>(T<sub>0</sub>)) and log<sub>10</sub>(t<sub>c,A,Ea</sub>(T<sub>0</sub>)) were computed for 30*30 values of (A, Ea) numerically with Cantera in order to obtain a training set. For given values of A, Ea and T<sub>0</sub>, we can avoid calling Cantera by estimating log<sub>10</sub>(t<sub>c,A,Ea</sub>(T<sub>0</sub>)) through the piecewise linear interpolation and then approximate log<sub>10</sub>(t<sub>c,A,Ea</sub>(T<sub>0</sub>)) ≈ a<sub>A,Ea</sub> 1/T<sub>0</sub> + b<sub>A,Ea</sub> through the linear interpolation formula.

The quality of the model has been tested by generating 3000 random values of (A, Ea) and T<sub>0</sub> with A ∼ U(A<sub>min</sub>, A<sub>max</sub>), Ea ∼ U(E<sub>a</sub><sub>min</sub>, E<sub>a</sub><sub>max</sub>) and T<sub>0</sub> ∼ N(μ<sub>T0</sub>, σ<sub>T0</sub>). We systematically computed t<sub>c,A,Ea</sub>(T<sub>0</sub>) with Cantera and t<sub>c,pred,A,Ea</sub>(T<sub>0</sub>) by interpolation and then the relative difference

\[
r = \frac{t_{c,A,Ea}(T_0) - t_{c,pred,A,Ea}(T_0)}{\min(t_{c,A,Ea}(T_0), t_{c,pred,A,Ea}(T_0))}.
\]

(4)

For more than 99.3% of the points, we have r ≤ 5%. We have max(r) = 12.32% which is reached for a very high and utterly unproblematic delay time superior to 2E+08 s. Given the fact that we are not interested in numerical accuracy but in understanding the behaviour of probabilistic approaches to thermal runaway risks, we deemed that level of error to be acceptable.
for given A and Ea, the probability density of tc is given by

\[ f(tc|A, Ea) = \frac{a_{A,Ea}ln(10)}{tc(b_{A,Ea}ln(10) - ln(tc))^2}\phi\left(\frac{a_{A,Ea}}{log10(tc) - b_{A,Ea}}, \mu_0, \sigma_0\right). \]

This analytical formula has been compared with an empirical distribution obtained by generating 10,000 values of \( T_0 \sim N(\mu_0 = 524K, \sigma_0 = 20K) \) and computing \( tca, Ea(T_0) \) by using the linear approximation. As can be seen in Figure 1, the analytical distribution of \( tc \) corresponds very well to the empirical one.

### 2.2. Priors

Chemical kineticists often define the prior in such a way that A and Ea are stochastically independent (which might be a problematic assumption, see (Held et al., 2008) ) and that it is uniform with respect to \( log10(A) \) and \( Ea \) (Plessis, 2013; Hsu et al., 2009; Huan and Marzouk, 2013). Consequently, we defined our first prior \( f_0.1(\log10(A), Ea) \) as uniform with respect to \( log10(A) \) and \( Ea \). We then defined our second prior \( f_{0.2} \) in such a way that it is uniform with respect to A and Ea. Our third prior \( f_{0.3} \) is uniform with respect to \( 1/log10(A) \) and \( 1/Ea \). The fourth prior \( f_{0.4} \) is uniform with respect to A and \( 1/Ea \). The fifth prior \( f_{0.5} \) is uniform with respect to \( 1/\log10(A) \) and \( 1/Ea \). For the sake of the present study, we ignored the problem of the stochastic independence of \( \log10(A) \) and \( Ea \) but intend to explore this issue in future works. The prior distributions along with their analytical expressions can be seen in Appendix B. It is a well-known problem of robust Bayesian analysis that the set of priors must be large enough to avoid the introduction of spurious information that could dominate the experimental data (Walley, 2000) but not so wide as to lead to nearly vacuous posteriors that would make it very hard to draw any practical conclusion (Held et al., 2008).

From a pragmatic and pedagogical point of view, these six priors are a good way to present an imprecise framework to chemical kineticists as all priors are uniform with respect to some reformulation of the kinetic parameters so that it would be arbitrary to only rely on the results derived from one such prior.

The elicitation of priors through subjective means Daneshkhah et al. (2017) is outside the scope of the present study.

### 2.3. Measurements and Epistemic Situations

We created "experimental" data allowing us to determine the posterior probability distributions of \( \log10(A) \) and \( Ea \). We considered a constant-volume adiabatic reactor at atmospheric pressure with a very diluted mixture of propane and oxygen: \( X_{C3H8} = 1E-05, X_{O2} = 5E-05 \) and \( X_{N2} = 0.99994 \) so that the temperature \( T(t) \) remains nearly constant. Using the "true" values \( A_0 \approx 8.60E+11 \) and \( E_{A0} = 30.00 \text{ kcal/mol} \), we generated mole fraction profiles of propane at different temperatures. We then randomly chose several time points and generated normally distributed noise in such a way that \( X_{C3H8}(t) = X_{C3H8}(A_0, Ea_0, \epsilon_j) + \epsilon_j, A_0, Ea_0 \) and \( \epsilon_j, A_0, Ea_0 \sim N(0, \sigma_j, A_0, Ea_0) \) with the standard deviation \( \sigma_j, A_0, Ea_0 = \sigma_{j,X_{C3H8}(A_0, Ea_0, \epsilon_j)} \) where \( \sigma_r \) is the relative standard deviation that always remains constant during an experiment. In practice, the true parameters \( A_0 \) and \( Ea_0 \) are of course unknown. For each measurement \( j \) of a given experiment, we use the expression of the standard deviation \( \sigma_j, A_0, Ea_0 \) defined just above.

As explained in Appendix C, we distinguished four epistemic situations:

- **A:** we only know that \( A \in [6.0E+11 ; 8.0E+13] \) (mol, cm.s) and \( Ea \in [27; 46] \text{ kcal/mol} \).

- **B:** we have one profile of \( X_{C3H8} \) with 6 time points measured at 1845 K with \( \sigma_r = 25\% \).

- **C:** We have two profiles of \( X_{C3H8} \) with 6 time points measured at 1135 K and 2249 K with \( \sigma_r = 25\% \).

- **D:** We have four profiles of \( X_{C3H8} \) with 10 time points measured at 1135 K, 1478 K, 1845 K, and 2249 K with \( \sigma_r = 6\% \).

As an example, \( X_{C3H8} \) at 1845 K with \( \sigma_r = 25\% \) can be seen in Figure 2. The other "measurements" are shown in Appendix C. The log-likelihood can be expressed as...
An Imprecise Bayesian Approach to Thermal Runaway Probability

Figure 2: $X_{CMHR}$ at 1845 K with $\sigma_r = 25\%$

follows:

$$l(data|\log_{10}(A), Ea) = \sum_{i=1}^{m} \sum_{j=1}^{n_{i,j}} -\frac{1}{2} \ln(2\pi\sigma_{r,i,j}^2) - \frac{1}{2\sigma_{r,i,j}^2} \left(X_{CMHR,i}(t_j,A,Ea) - X_{CMHR,exp}(t_j)\right)^2$$

whereby $m$ is the number of experiments and $n_{i,j}$ is the number of time points for experiment $i$ and $\sigma_{r,i,j} = \sigma_r X_{CMHR,i,A_0,Ea_0}(t_j)$ as explained above. For a given prior $f_0(\log_{10}(A), Ea)$ and a set of experimental data, the joint posterior probability density of $\log_{10}(A)$ and $Ea$ can be expressed by Eq. 6

$$f(\log_{10}(A), Ea|Data) = \frac{L(Data|\log_{10}(A), Ea)f_0(\log_{10}(A), Ea)}{\int_{\log_{10}(A), Ea} L(Data|\log_{10}(A), Ea)f_0(\log_{10}(A), Ea)d\log_{10}(A)dEa}$$

The integration was carried out with a first-order Euler explicit method (Hoffman and Frankel, 2018). The posteriors in Situation B, C and D (see 2.3) obtained with the first prior and 300*300 values of ($A, Ea$) are shown in Figure 3, 4 and 5, respectively. It is very obvious that there was a strong reduction in uncertainty between Situation B (involving one imprecise experiment at one temperature) and Situation D (involving four precise experiments at four temperatures). The other posterior probability densities of $\log_{10}(A)$ and $Ea$ are displayed as contour-plots in fischer21c-supp.
We then numerically computed the three quartiles of $t_c$ where we have no measurements and only know that the true values obtained with $A$ are considerably more similar and closer to the measured at constant temperature (see 2.3). These stark differences have become much narrower and that they are very close to the true values obtained with $A_0$ and $EA_0$. However, the delay times are systematically under-predicted whereas $p_{\text{critical}}$ is systematically over-predicted. As can be seen in Figure 4, while situation C is a clear improvement over situation B, the parameter values are still considerably uncertain. This is plausibly due to the fact that in Situation C, the highest value of the likelihood function is obtained for $A_{\text{max},C} = 7.42E+11$ (mol, cm, s) and $EA_{\text{max},C} = 29.54$ kcal/mol instead of $A_0 = 8.60E+11$ (mol, cm, s) and $EA_0 = 30.00$ kcal/mol. This leads to a higher reaction rate and thus to shorter ignition delay times and a larger explosion probability which result in $t_{c25} = 22.14$ s, $t_{c50} = 41.48$ s, and $p_{\text{critical}} = 0.3639$ for $A_{\text{max},C}$ and $EA_{\text{max},C}$. The systematic under-prediction of the delay times could possibly be avoided by using a parametric family of prior probability distributions resulting in larger posterior probability intervals.

### 3. Results and Discussion

#### 3.1. Estimation of $f(t_c)$

For the four situations A, B, C, and D, we computed $f(t_c)$ and the four features mentioned in 2.4. All the results can be seen in Appendix D.

In Table 1, the features of $f(t_c)$ are shown in situation A where we have no measurements and only know that $A \in [6.0E+11; 8.0E+13]$ (mol, cm,s) and $EA \in [27; 46]$ kcal/mol. Our representation of our ignorance through the non-parametric Bayesian prior would not be able to step in. For $A_0$ and $EA_0$, we have $t_{c25} = 28.84$ s, $t_{c50} = 55.04$ s, $t_{c75} = 108.68$ s, and $p_{\text{critical}} = 0.2634$.

#### 2.4. Computation of $f(t_c)$ as a Function of $f(\log 10(A), Ea)$

For a joint probability density of $\log 10(A)$ and $EA$, $f(\log 10(A), Ea)$ (which could be either a prior or a posterior), the probability density of $t_c$ is given by Eq 7.

\[
 f(t_c) = \int_{\log 10(A) \in [\log 10(A_{\text{min}}),\log 10(A_{\text{max}})], EA \in [EA_{\text{min}},EA_{\text{max}}]} f(t_c|\log 10(A), Ea) f_0(\log 10(A), Ea)d\log 10(A)dEA
 \]

\[
 (7)
\]

We then numerically computed the three quartiles of $t_c$ ($t_{c25}$, $t_{c50}$, $t_{c75}$) and $p_{\text{critical}} = p(t_c \leq 30x)$ which is the probability that the temperature rises so fast that a technician would not be able to step in. For $A_0$ and $EA_0$, we have $t_{c25} = 28.84$ s, $t_{c50} = 55.04$ s, $t_{c75} = 108.68$ s, and $p_{\text{critical}} = 0.2634$.

### 3. Results and Discussion

#### 3.1. Estimation of $f(t_c)$

For the four situations A, B, C, and D, we computed $f(t_c)$ and the four features mentioned in 2.4. All the results can be seen in Appendix D. The results for situation C can be seen in Table 3. The posterior values are considerably more similar and closer to the true values obtained with $A_0$ and $EA_0$. However, the delay times are systematically under-predicted whereas $p_{\text{critical}}$ is systematically over-predicted. As can be seen in Figure 4, while situation C is a clear improvement over situation B, the parameter values are still considerably uncertain. This is plausibly due to the fact that in Situation C, the highest value of the likelihood function is obtained for $A_{\text{max},C} = 7.42E+11$ (mol, cm, s) and $EA_{\text{max},C} = 29.54$ kcal/mol instead of $A_0 = 8.60E+11$ (mol, cm, s) and $EA_0 = 30.00$ kcal/mol. This leads to a higher reaction rate and thus to shorter ignition delay times and a larger explosion probability which result in $t_{c25} = 22.14$ s, $t_{c50} = 41.48$ s, and $p_{\text{critical}} = 0.3639$ for $A_{\text{max},C}$ and $EA_{\text{max},C}$. The systematic under-prediction of the delay times could possibly be avoided by using a parametric family of prior probability distributions resulting in larger posterior probability intervals.

Finally, the results obtained by updating the priors with the data of four experiments with a higher accuracy are summarised in Table 4. One can see that the discrepancies have become much narrower and that they are very close to the true values of the variables. The small system-
The second problem is the inability of the orthodox precise Bayesian position to distinguish knowledge and ignorance. Let us consider the probability $p_{critical}$ that the thermal runaway gets out of hand. For an imprecise Bayesian, the distinction between knowledge and ignorance can be captured by the differences between the posteriors and the values derived out of them. In situation A where we only know the parameter bounds, $p_{critical,A}$ takes on values between 0.1585 and 0.4476 and we can see that the average delay times $t_{25}$, $t_{50}$ and $t_{75}$ differ by orders of magnitude. In situation B where we only have one experiment which does not allow us to separate $log10(A)$ and $Ea$, $p_{critical,B}$ takes on values between 3.4E-03 and 0.2183 and the average delay times again differ by orders of magnitude. In situation D where we have more accurate measurements from four experiments at different temperatures, $p_{critical,D}$ takes on values between 0.2664 and 0.2680, which is close to 0.2634 and the differences between the delay times is always smaller than 0.5 %. This corresponds to a situation of warranted knowledge. The main limitation of the very simple priors we have chosen can be seen in situation C: the relative difference between the delay times is smaller than 12 % but the lowest and highest values of $p_{critical,D}$ (0.3311 and 0.3689) are far from the true value (0.2634). This outcome could plausibly be avoided by using a family of near-ignorance priors in the exponential family (Benavoli and Zaffalon, 2015; Quaeghebeur and De Cooman, 2005).

While the crude priors we considered here do not allow us to discriminate between cases where the distribution of $tc$ is inaccurate, they do permit us to recognise situations where the data truly dominate the priors. Indeed, Bayesian convergence theorems (commonly known as "The priors wash out!") (Hawthorne, 1994) show that the different posteriors are bound to converge towards a singular probability density distribution equal to 1 for $A = A_0$ and $Ea = EA_0$ with a favourable rate if the chosen priors are not too extreme.

Now, how could a Bayesian who is, for some reason, allergic to the very idea of interval probability manage to capture the distinction between knowledge and ignorance whilst using only one prior distribution? One possible way to do this would be to rely on the difference between "the weight of the argument" and the balance of evidence that was first introduced by British economist John Maynard Keynes (Keynes, 1921) and was recently explored by (Hill, 2019). Let us suppose that this Bayesian chooses to use the prior which is uniform with respect to $log10(A)$ and $Ea$. In situation A, all pairs of $(log10(A), Ea)$ have the same probability density. This corresponds to a situation of extreme ignorance (or maximum ignorance given the parameter bounds). Thus in situation A, we have $p_{critical,A} = 0.2424$ but this value is extremely unreliable as the weight of the argument is equal to zero. Likewise, in situation B $p_{critical,B} = 0.06$ but this value is strongly unreliable because $log10(A)$ and $Ea$ are strongly correlated as can be visualised in Figure 3. $p_{critical,D} = 0.2672$ and this value is strongly reliable as shown by Figure 5 where the likely
values of \((\text{log}10(A), Ea)\) occupy a very small region. In situation C, \(p_{\text{critical}} = 0.3508\) but the degree of reliability (or weight of the argument) is weaker as a much larger range of values are equally probable.

The reliability of \(p_{\text{critical}}\). (Keynes’ weight of the argument) would also have obvious consequences for decision-making and actions. In situations D, we reliably know that the probability of an uncontrollable thermal runaway is way too high, which means we must absolutely store the propane in a room where this type of incident raising the initial temperature \(T_0\) is impossible. In situations A, B and C, we do not reliably know \(p_{\text{critical}}\) so that we need to collect more relevant experimental data in order to strongly narrow down the range of possible values of \((A, Ea)\).

Such a Bayesian approach could also be applied to the example of the coin mentioned in the introduction 1. If we know absolutely nothing about the coin (except that it cannot land on edges), we would believe that \(p(\text{heads}) = 0.5\) while also knowing that this value is extremely unreliable so that ambiguity-averse individuals would be unwilling to bet any amount of money on either outcome. If we saw the relative frequency of heads during 10,000 tosses oscillate in a very narrow interval around 0.5, we would believe that \(p(\text{heads}) = 0.5\) and that this value is strongly reliable so that ambiguity-averse individuals would have no problem participating in bets. As shown by (Feduzi, 2010), Keynes wanted both the weight of the argument and the balance of evidence (the precise probability value in his framework) to play a role in decision-making but was unsure about how to achieve this because of the stopping-rule problem.

If applied consistently, this variety of Bayesianism could in this specific situation account for the difference between ignorance and knowledge. It would also provide us with an explanation of Ellberg’s paradox (Ellsberg, 1961) that completely respects the intuitions of the betting agents. However, it deviates so strongly from the precise Bayesian orthodoxy that it deserves to be considered a form of imprecise probability which fully recognises that probability can have different degrees of reliability.

That being said, this solution (which basically relies on some sorts of second-order probabilities) would not work in other situations. As Walley (1996) showed, in problems involving a multinomial distribution (such as guessing the colour of the next marble drawn from a urn), inferences based on a uniform prior depends on how the possibility space is defined and partitioned. This is very problematic, as we do not have a priori any more reason to suppose that \(p(\text{red}) = p(\text{yellow}) = p(\text{green}) = p(\text{black})\) than to assume that \(p(\text{red}) = p(\text{non} - \text{red})\) or \(p(\text{yellow}) = p(\text{non} - \text{yellow})\) and that we cannot automatically assume that the probability is uniform with respect to the simplest partition (Kelly (2011)).

4. Conclusion and Outlook

To the best of our knowledge, the evaluation of explosion and more specifically thermal runaway probabilities mostly occurs through the use of Bayesian networks based on the opinions of experts and on frequency data while representing uncertainty through a single probability distribution. What is more, while chemical kinetic parameter values are one main source of epistemic uncertainty, no effort has been made to transfer their uncertainties into predictions of explosion, so far as we know. In this pioneering work, we aimed at filling this gap by conducting a study combining aleatory uncertainty (the values of the initial temperature \(T_0\) and epistemic uncertainty (the values of the parameters \(A\) and \(Ea\)) to predict the distribution of the critical time \(tc\) before the thermal runaway gets completely out of hand.

After an introduction in Section 1, we presented our methodology in Section 2 by detailing the physical system and the underlying differential equations, the computation of the PDF of \(tc\) for given values of \(A\) and \(Ea\), the choice of the priors, the experiments used to update the priors, and how to compute \(f(tc)\) given a joint PDF of \((A, Ea)\). In Section 3, we presented our results along with their interpretation. Situation A corresponds to complete ignorance, Situation B to strong ignorance, situation D to very good knowledge, and situation C to insufficient knowledge. Our imprecise Bayesian method relying on 6 crude priors allows us to recognise that we know \(p_{\text{critical}}\) in situation D but are very ignorant in situation A and B. It is much harder to decide how accurate \(p_{\text{critical}}\) is in situation C based solely on the differences between the posteriori.

A Bayesian method considering only one prior but also a degree of accuracy for the various probabilities could potentially also capture the crucial distinction between knowledge and ignorance. However, it would strongly deviate from the precise Bayesian orthodoxy and would be a theory of imprecise probability in its own right. It would also fail in more complex situations such as those involving multinomial distributions.

There are several aspects we intend to explore in future works:

- The thermal runaway model we used is a huge simplification of reality. In addition to considering the thermal self-reinforcement of the reaction, the model should also include the chain reactions involving free radicals and the competition between ramification and chain termination reactions that can lead either to an explosion or to the end of the overall reaction (Warnatz et al., 2017). Ultimately, we shall also consider the complex interactions between chemistry, mass transfer, velocity fields, and heat transfer through complex CFD (Computational Fluid Dynamics) simulations (Seok et al., 2013).
• We considered only an homogeneous gas-phase reaction. Considering liquid-gas or solid-gas explosions would be very relevant for safety engineers as this type of scenario is more likely to be encountered in the industrial world. One example is dust explosion which can have disastrous consequences (Eckhoff, 2005). The simulation of such heterogeneous reactions would be computationally much more demanding (Murillo et al., 2013; Ermoline et al., 2013; Williams, 1979).

• The uniform priors we considered are very simple and as we saw, they are not good at identifying the level of inaccuracy in cases of partial ignorance (situation C). It would be interesting to find out whether our approach would be better at distinguishing different degrees of ignorance by using a class of Gaussian priors also characterised by different coefficients of correlation. However, it does not appear possible to obtain analytical versions of the posteriors, especially not if we use more realistic and complex models. In complex situations involving many parameters, we would then be left with no other choice than to rely on the MCMC (Markov-chain Monte-Carlo) algorithm to approximate the posteriors. Given the very long duration of each CFD simulation, we would also need to develop new surrogate models that are sufficiently trustworthy over a wide range of conditions.

References


Agata Boratyńska. Robust bayesian prediction with asymmetric loss function in poisson model of insurance risk. 2006.


Nikolai Nikolaevich Semenov. Thermal theory of combustion and explosion. 3; theory of normal flame propagation. 1942.
An Imprecise Bayesian Approach to Thermal Runaway Probability


