Uncertainty quantification with dual-based mesh adaption in a chemical system

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Simulating hydrogen oxidation in supercritical conditions

Many challenges since chemical systems typically involve:
- poorly known parameters (activation energy, reaction rates, delays, ...),
- several statistically independent sources of uncertainty,
- non-linear dependence on the parameters,
- a wide spectrum of time scales (potentially very stiff problem).

Objectives:
- account for and propagate the uncertainty of the parameters to the solution of the problem (say PDFs),
- solve the resulting stochastic problem in a numerically efficient way,
- control the error in the solution.

⇒ requires an efficient strategy!
Multi-elements Generalized Polynomial Chaos (ME-GPC) is appealing thanks to:

- its high convergence rate in many cases (smooth solution),
- a variational framework allowing a rigorous mathematical analysis.

Potentially large dimensionality of the stochastic problem → how to enrich the discretization in order to optimize the CPU / memory load given a required accuracy ?

⇒ Use of an adaptive strategy in the stochastic space.
A rigorous approach: a posteriori analysis

Stochastic problem in variational formulation for $U \in \mathcal{V}_t \otimes \mathcal{V}_\Theta$:

\[
\begin{cases}
A(U; \Phi|D) = F(\Phi|D) & \text{a.s. } \forall \Phi \in \mathcal{V}_t \otimes \mathcal{V}_\Theta, \\
U = U_0(D) & \text{on } \partial_0 \Omega_t \times \Omega_\xi.
\end{cases}
\]

$\mathcal{V}_\Theta = L^2(\Theta, dP)$.

The solution $U$ is dependent on some data $D$ which may not be exactly known (uncertain). $D$ is parameterized by a finite number of iid random variables defined on $(\Omega_\xi, \mathcal{B}, dP_\xi)$ with value in $\Xi_\xi$.

Solution evaluated in terms of $\mathcal{J}(U)$ (= $U$ here).
A posteriori analysis

In the spirit of the error analysis for finite elements:

\[ \mathcal{J}(U) - \mathcal{J}(U^h) = \rho(U^h, Z - Z^h) + R, \]
\[ \equiv \sum_l \sum_r \eta_{l,r}. \]

where \( \rho(U^h, \cdot) \equiv F(\cdot) - A(U^h; \cdot), \)

\[ R = \int_0^1 \left[ A''(U^h + s E_U; E_U^2, Z) - \mathcal{J}''(U^h + s E_U; E_U^2) \right] s \, ds, \]

\( E_U \equiv U - U^h. \)

The residual \( R \) is quadratic in \( E_U \Rightarrow \) neglected (\(| E_U | \ll 1\)).

\( Z \) is the “exact” adjoint solution \( \Rightarrow \) computed on a fine grid.

\( Z^h \) is the adjoint variable computed on the regular grid.
Anisotropic refinement

Potentially large dimensionality $\Rightarrow$ isotropic treatment of the refinement procedure is not an option!
Anisotropic refinement

Potentially large dimensionality ⇒ isotropic treatment of the refinement procedure is not an option!

Anisotropic error indicator based on the directional error estimate:

- for a block \( \{l, r\} \), compute the solution error estimate \( \eta_{l,r} \),
- consider a discretization omitting terms depending on direction \( j \) and compute the error estimate “contribution” from \( j \) solely:

\[
\eta^j_{l,r} = \left| \int_{\Omega_l^{(r)}} \int_{\Omega_x^{(l)}} \left[ A \left( U_j^h; Z - Z^h | D^h \right) - F \left( Z - Z^h | D^h \right) \right] dP_\xi(\xi) dt \\
+ \int_{\Omega_x^{(l)}} \left[ U^h \right]^{r-1} (Z - Z^h)^{(r-1)+} dP_\xi(\xi) \right|
\]

⇒ tends to balance error among all the elements and directions.
Adaption algorithm

1. solve the discrete primal equation on the regular grid \((U^h)\),
2. solve the adjoint equation both on regular and fine grids \((Z^h\) and \(\tilde{Z}\)),
3. compute approximation error \(\eta_{l,m}\) on each block \(\{l, m\}\),
4. if \(\eta > \text{Tol}_\eta\), \(\eta^+\)-mark the block
   - if maximum (over species) directional indicator \(\nu^j > \text{Tol}_\nu\) ⇒ \(h\)-refine along dimension \(j\)
     - if no \(h\)-refined direction → \(h\)-refine along direction with highest directional error,
5. if \(\eta < \text{Tol}_\eta/\gamma\) (\(\gamma = 4\) typically), \(\eta^-\)-mark the block,
   - determine all directions with a directional error above tolerance,
   - check if some surrounding blocks also require coarsening along one of those directions,
   - if so, check if the merging is possible (blocks conformity),
   - if so, merge the two blocks,
6. go back to (1) until no marked blocks remain.
Chemical system: supercritical hydrogen oxidation

Simplified mechanism but uncertain reaction rates.

<table>
<thead>
<tr>
<th>j</th>
<th>reaction</th>
<th>$\hat{\Gamma}_{f,j}$</th>
<th>$\gamma_j$</th>
<th>$\lambda_j$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>OH + H $\rightleftharpoons$ H$_2$O</td>
<td>$1.479 \times 10^{14}$</td>
<td>$+\infty$</td>
<td>3.16</td>
</tr>
<tr>
<td>2</td>
<td>H$_2$ + OH $\rightleftharpoons$ H$_2$O + H</td>
<td>$6.295 \times 10^{11}$</td>
<td>$4.380 \times 10^{-4}$</td>
<td>1.26</td>
</tr>
<tr>
<td>3</td>
<td>H + O$_2$ $\rightleftharpoons$ HO$_2$</td>
<td>$8.314 \times 10^{13}$</td>
<td>$9.879 \times 10^{-14}$</td>
<td>1.58</td>
</tr>
<tr>
<td>4</td>
<td>HO$_2$ + HO$_2$ $\rightleftharpoons$ H$_2$O$_2$ + O$_2$</td>
<td>$7.281 \times 10^{11}$</td>
<td>$1.045 \times 10^{-9}$</td>
<td>1.41</td>
</tr>
<tr>
<td>5</td>
<td>H$_2$O$_2$ + OH $\rightleftharpoons$ H$_2$O + HO$_2$</td>
<td>$3.469 \times 10^{12}$</td>
<td>$3.382 \times 10^{-9}$</td>
<td>1.58</td>
</tr>
<tr>
<td>6</td>
<td>H$_2$O$_2$ + H $\rightleftharpoons$ HO$_2$ + H$_2$</td>
<td>$1.696 \times 10^{11}$</td>
<td>$7.723 \times 10^{-6}$</td>
<td>2.00</td>
</tr>
<tr>
<td>7</td>
<td>H$_2$O$_2$ $\rightleftharpoons$ OH + OH</td>
<td>$3.993 \times 10^{1}$</td>
<td>$1.589 \times 10^{11}$</td>
<td>3.16</td>
</tr>
<tr>
<td>8</td>
<td>OH + H$_2$O $\rightleftharpoons$ H$_2$O + O$_2$</td>
<td>$3.917 \times 10^{13}$</td>
<td>$3.534 \times 10^{-18}$</td>
<td>3.16</td>
</tr>
</tbody>
</table>

$\hat{\Gamma}_{f,j}$: median forward rate value of reaction $#j$ determined from experimental data. $\gamma_j = \hat{\Gamma}_{f,j}/\hat{\Gamma}_{r,j}$.

$\lambda_j$ such as $P(\Gamma_{f,j} \in [\hat{\Gamma}_{f,j}/\lambda_j; \hat{\Gamma}_{f,j} \lambda_j]) = 0.95$.

Reaction rates modeled as independent random variables following a log-normal distribution.
Solution inspection - $p = 1$

Resulting stochastic partition ($t = 10\text{ s}$).

$\Rightarrow$ Fine grid in the steep regions only.
Coarse grid in smooth regions.

$[\text{H}]$ response surface at $t = 10\text{ s}$. 
All the reactions are now considered to be uncertain \( \Rightarrow \) the problem becomes 8-D. Very computationally demanding without a specific strategy.

First order polynomial chaos: \( p = 1 \)
Initial grid: \( 1^8 = 1 \) block.
Impact of the coarsening capability

$p = 1$, $Tol_\eta = 0.25$, $Tol_\nu = 0.08$.

Same accuracy!
Stochastic space partition statistics

Number of blocks marked for fusion (thick) and fission (thin) along each of the 8 directions ([OH], [H], [H₂O], [H₂], [O₂], [HO₂], [H₂O₂]).

⇒ The strong discrepancy between the reactions steepness is well captured.
Concluding remarks

- Rigorous treatment of an adaptive $hp$-GPC scheme for problems with parametric uncertainty,
- highly accurate results with reasonable CPU cost,
- stochastic problems with a larger dimensionality remain affordable using anisotropic adaption,
- high flexibility ($h/p$ modularity),
- suitable for unsteady problem thanks to the coarsening capability,
- efficient parallel treatment.
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- Rigorous treatment of an adaptive $hp$-GPC scheme for problems with parametric uncertainty,
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Further work

- improve the anisotropic treatment (better directional error indicator).
A posteriori analysis (cont’d)

Approximation error writes

\[
\eta = \left| \left\langle A \left( U^h; Z - Z^h|D^h \right) - F \left( Z - Z^h|D^h \right) \right\rangle_{\Omega_\xi} \right|
= \left| \int_{\Omega_t} \int_{\Omega_\xi} \left[ A \left( U^h; Z - Z^h|D^h \right) - F \left( Z - Z^h|D^h \right) \right] dP_\xi(\xi) dt \right|
\]

Minimum of \( \eta \Rightarrow \) optimization problem.

We have \( \eta \leq \sum_{l=1}^{N_b} \sum_{r=1}^{N_r} \eta_{l,r} \) \( l, r \) refers to the stochastic-time element \( \{l, r\} \).

\[
\eta_{l,r} = \left| \int_{\Omega_t^{(r)}} \int_{\Omega_\xi^{(l)}} \left[ A \left( U^h; Z - Z^h|D^h \right) - F \left( Z - Z^h|D^h \right) \right] dP_\xi(\xi) dt \right|
\]

We prescribe \( \eta_{l,r} \leq Tol_\eta \).
Anisotropic refinement (cont’d)

The directional error indicator is

\[ \nu_{l,r}^j = \frac{\eta_{l,r}^j}{\eta_{l,r}} \]

The refinement procedure aims at

\[ \nu_{l,r}^j \leq \text{Tol}_\nu \quad \forall j \in [1; n] \]

⇒ tends to equilibrate error among all the elements and directions.

Another anisotropic estimator is also used based on solution jump across contiguous elements in the stochastic space (not described).
Solution method (cont’d)

Chemical system governed by a set of coupled ODEs of the form, say for [OH]:

\[
\frac{d[OH](\zeta, t)}{dt} = \tilde{\Gamma}_{r,1}(\zeta, t) [H_2O](\zeta, t) + \tilde{\Gamma}_{r,2}(\zeta, t) [H_2O](\zeta, t) [H](\zeta, t) \\
+ \tilde{\Gamma}_{r,5}(\zeta, t) [H_2O](\zeta, t) [HO_2](\zeta, t) \\
+ \tilde{\Gamma}_{r,8}(\zeta, t) [H_2O](\zeta, t) [O_2](\zeta, t) - 2 \tilde{\Gamma}_{f,7}(\zeta, t) [H_2O_2](\zeta, t)
\]

Deterministic initial conditions:

\[
[OH] = 0 \\
[H] = 0 \\
[HO_2] = 0 \\
[H_2O_2] = 0
\]
Solution method

Reaction rates modeled as independent random variables following a log-normal distribution. It is chosen to parameterize the reaction rates with 8 iid normalized Gaussian variables $\xi_j$:

$$\Gamma_{f,j} = \hat{\Gamma}_{f,j} \exp^{\log \lambda_j} \xi_j \quad \forall j \in \{1, \ldots, 8\}$$

Mapping of the problem onto a bounded stochastic domain partioned in blocks:

$$\tilde{\Gamma}_{f,j}(t, \zeta) = \sum_{i=0}^{P(l)} \left(\tilde{\Gamma}_{f,j}\right)_i(t) \Psi_i(\zeta)$$

with $\zeta = (\zeta_1, \ldots, \zeta_8)$, $\zeta_j$ being uniformly distributed on $[-1; 1]$. 
**First statistical moments**

Reaction rates of reactions 7 and 8 solely are considered uncertain.

- **Time evolution of the mean hydrogen concentration**
- **Time evolution of the standard deviation of [H]**

⇒ Variance grows faster than the mean value.