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Combustion and Flame

Analysis and evaluation of steady-state and non-steady-state preserving operator splitting schemes for reaction-diffusion(-advection) problems



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ABSTRACT

Reaction-diffusion(-advection) problems are well-known in chemical engineering and computational fluid dynamics. A common feature of these systems is a linear (or non-stiff) transport sub-system and a nonlinear (or stiff), highly coupled chemistry sub-system. The expected numerical effort often prohibits a fully coupled solution of the system. Therefore, the system is split into a transport and a reaction subsystem, and each sub-system is solved using specialized solvers. Operator splitting schemes are required to reconstruct the solution of the initial system from the sub-system solutions. Steady-state preserving splitting schemes are particularly essential for steady-state calculations since local time stepping (LTS) or other methods based on fictional time rely on large time steps to be efficient. This work formally analyzes common splitting schemes for reaction-diffusion problems by stability analysis and checking the steadystate preservation of a representative linear scalar problem. Balanced and Simpler Balanced splitting are the only steady-state conservative schemes analyzed. Three new steady-state preserving splitting schemes are proposed based on the findings of the formal analysis. To achieve steady-state preservation, the new schemes use splitting constants based on either the mixing derivative or the chemistry derivative. The formal analysis is accompanied by a dimensionless perfectly stirred reactor (PSR) case and a hydrogen combustion case, both known to be challenging for operator splitting schemes. The test case results are in line with the theoretical results but indicate that the scalar linear analysis is nonviable to capture the full effects of the chemical sub-system. The Simpler and the newly proposed Consistent Staggered splitting schemes give significantly better results than the remaining ones while being second-order and firstorder accurate, respectively. If temporal accuracy is irrelevant, e.g., for steady-state solvers, the proposed Consistent Adaptive splitting scheme is promising since it preserves the steady-state solution first-order time accurate with less function evaluations.

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1. Introduction

Novel and modern combustion technologies aim to reduce pollutant emissions and increase process efficiency. MILD combustion is a promising technology that aims to reduce pollutants by distributing the flame over large regions compared to classical combustion [1]. Spreading the reaction layer reduces combustion intensity and final combustion temperatures and, thus, pollution formation. Oxidizer dilution and thorough mixing of fuel, oxidizer, and recirculated exhaust gas enable the reaction zone transformation compared to classical combustion. These process conditions

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increases the occurrence of near-extinction and near-ignition states in flames. Another technology is (ultra-)lean combustion [2,3], also referred to as cool flames, which aims to keep the combustion temperature low by employing fuel-to-air ratios $\phi \ll 1$, which reduces NO_X emissions and increases combustion efficiency. Operating near or below the flammability limit facilitates near-extinction and near-ignition states in (ultra-)lean flames. Capturing these effects is crucial for the reliable modeling of MILD and (ultra-)lean flames and the advancement of novel combustion technologies through Computational Fluid Dynamics (CFD) or other modeling approaches.

CFD codes use operator splitting to reduce the numerical effort of combustion problems. The governing equations consist of a transport (convection, diffusion) and a chemistry part. The trans-

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port problem can be solved segregated, while chemistry is a nonlinear, coupled problem. Solving the reaction-diffusion problem fully coupled is challenging because of the numerical effort. Splitting the original problem into a transport and a chemistry problem and solving them independently enables the use of specialized solvers for both equation types. The splitting, however, requires special algorithms to combine the independent solutions to reconstruct the original problem. These algorithms are called operator splitting schemes. Splitting schemes should reproduce the original problem accurately in time and preserve its steady-state characteristics.

In mathematics, operator splitting is widely used to solve differential equations. The fractional-step method is one example used to solve multi-dimensional problems [4,5], e.g., the threedimensional Navier-Stokes equations. Operator splitting is used in reactive flow problems to reduce the numerical effort since it enables the following advantages or speed-up possibilities:

- i) in-situ adaptive tabulation (ISAT) for chemistry [6,7]: ISAT is a method to generate a chemistry lookup table on the fly during simulations, which automatically adapts to the chemistry sub-problem [6,7].
- ii) reduction of memory consumption: Segregated solution of the transport equations significantly reduces the peak memory consumption compared to solving them fully coupled [8].
- iii) application of specialized solvers: The loosely-coupled transport part and the highly-coupled chemistry part can be solved using specialized solvers for the different subproblem types [9,10].

The main disadvantage of operator splitting is the temporal and steady-state errors introduced by dividing the problem into subproblems.

Near-ignition and near-extinction states have been shown to be challenging for operator splitting schemes [11,12]. These conditions emerge as the Damköhler number (Da) decreases towards unity [13]. Da is defined as the ratio of the mixing time scale (τ_{mix}) and the chemistry time scale (τ_{chem}):

$$Da = \frac{\tau_{\rm mix}}{\tau_{\rm chem}} \tag{1}$$

The mixing time scale is easily defined by turbulence parameters for reacting flows or by the residence time for Perfectly Stirred Reactors (PSR). On the contrary, defining the chemistry time scale is more complex [14]. Modern combustion technologies like MILD or (ultra-)lean combustion feature significant regions near limit states [1–3]. Therefore, it is crucial to employ operator splitting schemes that can to correctly predict such conditions.

Besides using operator splitting for combustion CFD, it can also be used to speed-up perfectly stirred reactor (PSR) calculations, e.g., for the fine structure closure of the Eddy Dissipation Concept (EDC) [15,16] or 0D/1D flame codes like Cantera [17] or CHEMKIN [18].

A generic reaction-diffusion problem is introduced, and the relevant relations for the subsequent analysis of operator splitting schemes are derived (Section 2). Steady-state behavior and temporal accuracy of commonly used operator splitting schemes are evaluated using a linear problem (Section 3). New steady-state preserving operator splitting schemes are proposed based on the evaluation of the existing ones (Section 4). Furthermore, scalar linear stability analysis is employed to evaluate the splitting schemes' stability properties (Section 5). Subsequently, the theoretical findings are verified using a scalar (Section 6) and a combustion (Section 7) test case, which are known to cause troubles for the Strang splitting scheme [11,12].

2. Theory

Operator splitting schemes are widely used in CFD of reactive flows. The aim of splitting schemes is an accurate solution to the original problem with the least possible effort. Reaction-diffusion(advection) reaction equations are split into: i) the diffusionadvection and ii) the reaction part to avoid the solution of the fully coupled problem. Linear matrix solvers can handle the convectivediffusion part, while special non-linear solvers are used to solve the strongly coupled stiff chemical sub-system. The operator splitting scheme combines the sub-systems' solutions to retain the original system's solution. The reaction-diffusion equation of a perfectly stirred reactor (PSR) is given by Wu et al. [19]:

$$\frac{d\Phi}{dt} = \frac{1}{\tau_{\rm res}} (\Phi_{\infty} - \Phi) + \dot{\omega}(\Phi)$$
(2)

where $\mathbf{\Phi} = \{\mathbf{Y}, h\}$ is the state vector consisting of the species mass fractions and the enthalpy, τ_{res} is the residence time, $\mathbf{\Phi}_{\infty}$ is the inflow state vector, and $\dot{\omega}(\mathbf{\Phi})$ is the non-linear chemistry term.

The problem can be split into a mixing (\mathcal{T}) and chemical (\mathcal{R}) sub-problem:

$$\frac{d\Phi}{dt} = \mathcal{T} + \mathcal{R} \tag{3}$$

The mixing operator is typically non-stiff, while the chemistry operator can become stiff.

Speth et al. [20] introduced balancing constants (C_N) to modify the stability and steady-state properties of the Strang spitting schemes without changing the original problem. The balancing constants change the initial problem to:

$$\frac{d\Phi}{dt} = \mathcal{T}' + \mathcal{R}' = (\mathcal{T} + \mathcal{C}_N) + (\mathcal{R} - \mathcal{C}_N)$$
$$= (\mathbf{A}\Phi + \mathbf{a} + \mathcal{C}_N) + (\mathbf{B}\Phi + \mathbf{b} - \mathcal{C}_N)$$
(4)

Adding balancing constants modifies the solution of the subproblems while not affecting the solution of the initial problem.

The relation for the time integration can be generalized based on a recurrence relation or growth factor (\mathbf{R}) and the constant part or matrix factor (\mathbf{Q}), which is given by Wu et al. [19], Speth et al. [20]:

$$\Phi_{n+1} = \boldsymbol{R} \Phi_n + \boldsymbol{Q} \tag{5}$$

The analytical ($\Phi(t)$) solution of Eq. (4) for $C_N = 0$ can be easily found [20]:

$$\boldsymbol{\Phi}(t) = \boldsymbol{\Phi}_{\infty} + \exp\left((\boldsymbol{A} + \boldsymbol{B}) \cdot t\right) \cdot (\boldsymbol{\Phi}_{0} - \boldsymbol{\Phi}_{\infty})$$
(6)

The steady-state solution (Φ_{∞}) can be found by setting $\Phi_{n+1} = \Phi_n = \Phi_{\infty}$ and rearranging Eq. (5) and solving it for $h \to \infty$:

$$\boldsymbol{\Phi}_{\infty} = \boldsymbol{Q}(\boldsymbol{I} - \boldsymbol{R})^{-1} \tag{7}$$

The analytic solution for the steady-state is then given by:

$$\boldsymbol{\Phi}_{\infty} = -(\boldsymbol{a} + \boldsymbol{b})(\boldsymbol{A} + \boldsymbol{B})^{-1}$$
(8)

The Maclaurin series (Taylor expansion around zero) of Eq. (7) can be used to identify the steady-state error of operator splitting schemes [20]. Taking the limit $h \rightarrow \infty$ of Eq. (7) identifies the tendency of the steady-state solution towards one of the sub-problems or a combination of them. Combining both approaches reveals if an operator splitting scheme is steady-state preserving and, in addition, gives the dominant sub-problem if it is not steady-state preserving.

The general analytical solutions of the sub-problems are given by:

$$\int_0^{\tau} \mathcal{T}'(t) dt = e^{\mathbf{A}\tau} + \left(e^{\mathbf{A}\tau} - 1\right) \mathbf{A}^{-1} \cdot (\mathbf{a} + \mathcal{C}_N)$$
(9a)

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$$\int_{0}^{\tau} \mathcal{R}'(t) dt = e^{\mathbf{B}\tau} + (e^{\mathbf{B}\tau} - 1)\mathbf{B}^{-1} \cdot (\mathbf{b} - \mathcal{C}_{N})$$
(9b)

Speth et al. [20] introduced the following entities:

$$e^{\mathbf{A}\tau} := \boldsymbol{\alpha}_{\#}, \quad e^{\mathbf{B}\tau} := \boldsymbol{\beta} \tag{10a}$$

$$\boldsymbol{A}_{\#}^{*} := \left(\boldsymbol{e}^{\boldsymbol{A}\boldsymbol{\tau}} - \boldsymbol{I}\right)\boldsymbol{A}^{-1}, \quad \boldsymbol{B}^{*} := \left(\boldsymbol{e}^{\boldsymbol{B}\boldsymbol{\tau}} - \boldsymbol{I}\right)\boldsymbol{B}^{-1}$$
(10b)

where τ is replaced with the corresponding time step sizes of the splitting scheme and # is a wild card, which is blank for $\tau = h$ and h/2 for $\tau = h/2$. Using these identities yields simpler expressions for the analytic solutions of Eq. (9):

$$\int_0^t \mathcal{T}'(t) dt = \boldsymbol{\alpha} \boldsymbol{\Phi}_n + \boldsymbol{A}^*(\boldsymbol{a} + \mathcal{C}_N)$$
(11a)

$$\int_0^\tau \mathcal{R}'(t) \, dt = \boldsymbol{\beta} \boldsymbol{\Phi}_n + \boldsymbol{B}^*(\boldsymbol{b} - \mathcal{C}_N) \tag{11b}$$

These solutions are also valid if no balancing constant is used $(\mathcal{C}_N=0).$

Operating spitting schemes must conserve the steady-state solution and be as accurate in time as possible. Various first and second-order accurate splitting schemes have been presented in the literature. The analyzed operator splitting schemes' temporal accuracy is evaluated based on the Maclaurin series of the growth factor. The series expansion of the growth factor of the reactiondiffusion problem is given by:

$$\mathbf{R}(h) = e^{(\mathbf{A}+\mathbf{B})h} = \sum_{n=0}^{\infty} \frac{(\mathbf{A}+\mathbf{B})^n h^n}{n!} \approx 1 + (\mathbf{A}+\mathbf{B})h + \frac{(\mathbf{A}^2 + \mathbf{A}\mathbf{B} + \mathbf{B}\mathbf{A} + \mathbf{B}^2)h^2}{2} + \frac{(\mathbf{A}^3 + \mathbf{A}^2\mathbf{B} + \mathbf{A}\mathbf{B}\mathbf{A} + \mathbf{A}\mathbf{B}^2 + \mathbf{B}\mathbf{A}^2 + \mathbf{B}\mathbf{A}\mathbf{B} + \mathbf{B}^3)h^3}{6} + \mathcal{O}(h^4)$$
(12)

The temporal accuracy of operator splitting schemes can be divided into two different cases: i) the local accuracy, which is equal to the leading order error of the Maclaurin series expansion, and ii) the global accuracy, which is one order lower than the error of the Maclaurin series because of the O(1/h) time steps required to solve the problem [20].

The stability of operator splitting schemes is typically evaluated by scalar linear stability analysis for simplicity [19,20]. In the scalar case, Eq. (4) simplifies to:

$$\frac{d\Phi}{dt} = \mathcal{T} + \mathcal{R} = (A\phi + a + \mathcal{C}_N) + (B\phi + b - \mathcal{C}_N)$$
(13)

Using the scalar variants of the identities from Eq. (10) gives the following solutions to the linearized scalar sub-problems:

$$\int_{0}^{\tau} \mathcal{T}'(t) dt = \alpha \Phi_n + A^*(a + \mathcal{C}_N)$$

$$\int_{0}^{\tau} \mathcal{R}'(t) dt = \beta \Phi_n + B^*(b - \mathcal{C}_N)$$
(14a)
(14b)

The accuracy and steady-state properties of common and newly proposed splitting schemes are discussed in Sections 3 and 4, while their stability properties are discussed in Section 5.

3. Review of selected operator splitting schemes

The presented operator splitting schemes have been used for reaction-diffusion problems [19-24]. The general calculation procedure, the steady-state solution, and the accuracy of the splitting schemes are subsequently discussed similar to the work of Speth et al. [20] and Wu et al. [19].

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3.1. Strang splitting

The Strang splitting scheme has been proposed in the 1960s [21] and is a second-order accurate splitting scheme. Strang splitting embeds a full time step of the chemical sub-problem between two half time steps of the mixing sub-problem:

$$d_t \mathbf{\Phi}^{(1)} = \mathcal{T}(\mathbf{\Phi}^{(1)}), \quad \mathbf{\Phi}^{(1)}(t_n) = \mathbf{\Phi}_n \tag{15a}$$

$$d_t \Phi^{(2)} = \mathcal{R}(\Phi^{(2)}), \quad \Phi^{(2)}(t_n) = \Phi^{(1)}(t_n + h/2)$$
 (15b)

$$d_t \Phi^{(3)} = \mathcal{T}(\Phi^{(3)}), \quad \Phi^{(3)}(t_n + h/2) = \Phi^{(2)}(t_n + h)$$
 (15c)

$$\Phi_{n+1} = \Phi^{(3)}(t_n + h/2)$$
(15d)

Integrating Eq. (15) over a single time step gives the following expressions for the growth and matrix factors:

$$\boldsymbol{R} = \boldsymbol{\alpha}_{h/2} \boldsymbol{\beta} \boldsymbol{\alpha}_{h/2} \tag{16a}$$

$$\mathbf{Q} = (\boldsymbol{\alpha}_{h/2}\boldsymbol{\beta} + \boldsymbol{I})\boldsymbol{A}^*\boldsymbol{a} + \boldsymbol{\alpha}_{h/2}\boldsymbol{B}^*\boldsymbol{b}$$
(16b)

The Maclaurin series of the growth factor, which defines the scheme's accuracy, is given by Speth et al. [20]:

$$\mathbf{R}(h) = \boldsymbol{\alpha}_{h/2} \boldsymbol{\beta} \boldsymbol{\alpha}_{h/2} = \mathbf{I} + (\mathbf{A} + \mathbf{B})h + \left(\mathbf{A}^2 + \mathbf{A}\mathbf{B} + \mathbf{B}\mathbf{A} + \mathbf{B}^2\right)\frac{h^2}{2} + \mathcal{O}(h^3)$$
(17)

which confirms second-order accuracy for O(1/h) time steps (globally) or third order accuracy locally.

The series expansion of Eq. (7) reveals time step size dependence of the steady-state solution, which is of second order for Strang splitting:

$$\Phi_{\infty} = -(\boldsymbol{a} + \boldsymbol{b})(\boldsymbol{A} + \boldsymbol{B})^{-1} + (\boldsymbol{A} + 2\boldsymbol{B})(\boldsymbol{A}\boldsymbol{a} - \boldsymbol{B}\boldsymbol{b})(\boldsymbol{A} + \boldsymbol{B})^{-1}\frac{h^2}{24} + \mathcal{O}(h^4)$$
(18)

Using the limit $h \to \infty$ of Eq. (7) shows that the steady-state solution of Strang splitting tends towards the solution of the mixing sub-problem:

$$\boldsymbol{\Phi}_{\infty} = -\boldsymbol{a}\boldsymbol{A}^{-1} \tag{19}$$

3.2. Balanced splitting

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Balanced splitting [20] is identical to Strang splitting except for the balancing constant, which is defined as $C_N = \frac{1}{2} \cdot (\mathcal{R} - \mathcal{T})$:

$$d_t \mathbf{\Phi}^{(1)} = \mathcal{T}(\mathbf{\Phi}^{(1)}) + \mathcal{C}_{\mathbf{N}}, \quad \mathbf{\Phi}^{(1)}(t_n) = \mathbf{\Phi}_n$$
(20a)

$$d_t \Phi^{(2)} = \mathcal{R}(\Phi^{(2)}) - \mathcal{C}_N, \quad \Phi^{(2)}(t_n) = \Phi^{(1)}(t_n + h/2)$$
(20b)

$$d_t \Phi^{(3)} = \mathcal{T}(\Phi^{(3)}) + \mathcal{C}_N, \quad \Phi^{(3)}(t_n + h/2) = \Phi^{(2)}(t_n + h)$$
 (20c)

$$\Phi_{n+1} = \Phi^{(3)}(t_n + h/2)$$
(20d)

Integrating Eq. (20) over a single time step gives the following growth and matrix factor:

$$\boldsymbol{R} = \boldsymbol{\alpha}_{h/2} \boldsymbol{\beta} \boldsymbol{\alpha}_{h/2} + \frac{1}{2} (\boldsymbol{\alpha}_{h/2} \boldsymbol{B}^* - (\boldsymbol{\alpha}_{h/2} \boldsymbol{\beta} + \boldsymbol{I}) \boldsymbol{A}^*) (\boldsymbol{A} - \boldsymbol{B})$$
(21a)

$$\mathbf{Q} = \frac{1}{2} (\boldsymbol{\alpha}_{h/2} \boldsymbol{B}^* + (\boldsymbol{\alpha}_{h/2} \boldsymbol{\beta} + \boldsymbol{I}) \boldsymbol{A}^*) (\boldsymbol{a} + \boldsymbol{b})$$
(21b)

The global and local accuracy remain second and third order, respectively:

$$\mathbf{R}(h) = \boldsymbol{\alpha}_{h/2} \boldsymbol{\beta} \boldsymbol{\alpha}_{h/2} + \frac{1}{2} (\boldsymbol{\alpha}_{h/2} \boldsymbol{B}^* - (\boldsymbol{\alpha}_{h/2} \boldsymbol{\beta} + \boldsymbol{I}) \boldsymbol{A}^*) (\boldsymbol{A} - \boldsymbol{B})$$

= $\boldsymbol{I} + (\boldsymbol{A} + \boldsymbol{B})h + (\boldsymbol{A}^2 + \boldsymbol{A}\boldsymbol{B} + \boldsymbol{B}\boldsymbol{A} + \boldsymbol{B}^2) \frac{h^2}{2}$
+ $\mathcal{O}(h^3)$ (22)

However, the balancing constant removes the time step dependence of the steady-state solution (see Eq. (8)).

3.3. Simpler splitting

Wu et al. [19] proposed the Simpler splitting scheme based on the balanced splitting scheme of Speth et al. [20]. The balancing constant for Simpler splitting is only based on the mixing step. As a result, the first mixing sub-step is in equilibrium, and can be skipped to reduce numerical effort. Eq. (23) summarizes the Simpler splitting scheme.

$$\mathcal{C}_{N} = -\mathcal{T}(\Phi) \tag{23a}$$

$$d_t \boldsymbol{\Phi}^{(1)} = \boldsymbol{\mathcal{R}}(\boldsymbol{\Phi}^{(1)}) - \boldsymbol{\mathcal{C}}_{\boldsymbol{N}}, \quad \boldsymbol{\Phi}^{(1)}(t_n) = \boldsymbol{\Phi}_n$$
(23b)

$$d_t \Phi^{(2)} = \mathcal{T}(\Phi^{(2)}) + \mathcal{C}_{\mathcal{N}}, \quad \Phi^{(2)}(t_n + h/2) = \Phi^{(1)}(t_n + h)$$
 (23c)

$$\boldsymbol{\Phi}_{n+1} = \boldsymbol{\Phi}^{(2)}(t_n + h) \tag{23d}$$

The growth and matrix factors for the Simpler splitting scheme are given by:

$$\boldsymbol{R} = \boldsymbol{\alpha}_{h/2} \boldsymbol{\beta} \boldsymbol{\alpha}_{h/2} + \left(\boldsymbol{\alpha}_{h/2} \boldsymbol{B}^* - \left(\boldsymbol{\alpha}_{h/2} \boldsymbol{\beta} + \boldsymbol{I} \right) \boldsymbol{A}^* \right) \boldsymbol{A}$$
(24a)

$$\mathbf{Q} = \boldsymbol{\alpha}_{h/2} \boldsymbol{B}^* (\boldsymbol{a} + \boldsymbol{b}) \tag{24b}$$

The accuracy of the Simpler splitting is similar to the Balanced splitting; second-order globally and third order locally. The series expansion of the growth factor is given by:

$$\mathbf{R}(h) = \boldsymbol{\alpha}_{h/2}\boldsymbol{\beta}\boldsymbol{\alpha}_{h/2} + (\boldsymbol{\alpha}_{h/2}\boldsymbol{B}^* - (\boldsymbol{\alpha}_{h/2}\boldsymbol{\beta} + \boldsymbol{I})\boldsymbol{A}^*)\boldsymbol{A}$$

= $\boldsymbol{I} + (\boldsymbol{A} + \boldsymbol{B})h + (\boldsymbol{A}^2 + \boldsymbol{A}\boldsymbol{B} + \boldsymbol{B}\boldsymbol{A} + \boldsymbol{B}^2)\frac{h^2}{2}$
+ $\mathcal{O}(h^3)$ (25)

Furthermore, the steady-state solution is independent of the time step size and resembles Eq. (8).

3.4. Staggered splitting

The Staggered splitting scheme was proposed by Ren and Pope [22] and, similar to Simpler splitting, aims to reduce the number of function evaluations. The scheme consists of two sub-steps, which are averaged to obtain the solution of the time integration. Figure 1 illustrates the sub-step arrangement of the Staggered splitting scheme. The solution of the last mixing time step is used as starting point for the initial chemistry step, which is the basis for the mixing step. Time shifts are done to create an overlap between the chemistry and mixing steps. Eq. (26) gives the formula for the Staggered splitting scheme.

$$d_t \boldsymbol{\Phi}^{(1)} = \mathcal{R}(\boldsymbol{\Phi}^{(1)}), \quad \boldsymbol{\Phi}^{(1)}(t_n) = \boldsymbol{\Phi}_{t_n + h/2}$$
(26a)

$$d_t \Phi^{(2)} = \mathcal{T}(\Phi^{(2)}), \quad \Phi^{(2)}(t_n + h/2) = \Phi^{(1)}(t_n + h)$$
 (26b)

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Fig. 1. Schematics of the Staggered operator splitting scheme [22]. T and R denote a mixing and chemistry step, respectively.

$$\Phi_{n+1} = \frac{\Phi^{(1)}(t_n+h) + \Phi^{(2)}(t_n+3/2h)}{2}$$
(26c)

A half mixing sub-step is required at the beginning of the integration since Staggered splitting relies on h/2 shifted points for the integration [22]:

$$d_t \Phi^{(1)} = \mathcal{T}(\Phi^{(1)}), \quad \Phi^{(1)}(t_n) = \Phi_n$$
(27a)

$$\mathbf{\Phi}_{n+1/2} = \mathbf{\Phi}^{(1)}(t_n + h/2) \tag{27b}$$

In the concept of Staggered splitting, any time step starts with half of a mixing step. The subsequent chemistry and mixing steps are then averaged to obtain the time step solution. Therefore, the initial state ($\Phi_{n+1/2}$) is replaced by a half mixing step: $\alpha_{h/2} + A_{h/2}^*a$. The growth and matrix factor for the representative integration step are given by:

$$\mathbf{R} = \frac{1}{2} \left[\boldsymbol{\alpha} \boldsymbol{\beta} \boldsymbol{\alpha}_{h/2} + \boldsymbol{\beta} \boldsymbol{\alpha}_{h/2} \right]$$
(28a)

$$\mathbf{Q} = \frac{1}{2} \Big[\boldsymbol{\alpha} \big(\boldsymbol{\beta} \boldsymbol{A}_{h/2}^* \boldsymbol{a} + \boldsymbol{B}^* \boldsymbol{b} \big) + \boldsymbol{\beta} \boldsymbol{A}^* \boldsymbol{a} + \boldsymbol{A}^* \boldsymbol{a} + \boldsymbol{B}^* \boldsymbol{b} \Big]$$
(28b)

Numerical tests suggested that Staggered splitting is secondorder accurate [22]. The series expansion of the growth factor given in Eq. (29) contradicts this observation. Staggered splitting is only second-order accurate locally since there is an error of $(1/8A^2 - AB/4)h^2$ in the second-order expansion:

$$\mathbf{R}(h) = \boldsymbol{\alpha}\boldsymbol{\beta}\boldsymbol{\alpha}_{h/2} + \boldsymbol{\beta}\boldsymbol{\alpha}_{h/2}$$
$$= \mathbf{I} + (\mathbf{A} + \mathbf{B})h + \left(\frac{5}{4}\mathbf{A}^2 + \frac{\mathbf{A}\mathbf{B}}{2} + \mathbf{B}\mathbf{A} + \mathbf{B}^2\right)\frac{h^2}{2}$$
$$+ \mathcal{O}(h^3)$$
(29)

The minor error in the growth factor can resemble second-order behavior, although Staggered splitting is not second-order accurate.

Moreover, the splitting scheme is not steady-state preserving and has a zeroth-order dependence of the steady-state solution on the time step size:

$$\boldsymbol{\Phi}_{\infty} = -\left(\frac{5}{4}\boldsymbol{a} + \boldsymbol{b}\right)(\boldsymbol{A} + \boldsymbol{B})^{-1} + \mathcal{O}(h)$$
(30)

The limit $h \rightarrow \infty$ of Eq. (7) for Staggered splitting resembles a linear combination of the sub-problem solutions:

$$\boldsymbol{\Phi}_{\infty} = -\frac{1}{2}\boldsymbol{a}\boldsymbol{A}^{-1} - \frac{1}{2}\boldsymbol{b}\boldsymbol{B}^{-1} \tag{31}$$

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3.5. Lie-Trotter

The Lie–Trotter scheme solves the chemical sub-step based on the previous solution of the mixing sub-step [23]:

$$d_t \boldsymbol{\Phi}^{(1)} = \boldsymbol{\mathcal{T}}(\boldsymbol{\Phi}^{(1)}), \quad \boldsymbol{\Phi}^{(1)}(t_n) = \boldsymbol{\Phi}_n \tag{32a}$$

$$d_t \Phi^{(2)} = \mathcal{R}(\Phi^{(2)}), \quad \Phi^{(2)}(t_n) = \Phi^{(1)}(t_n + h)$$
 (32b)

$$\mathbf{\Phi}_{n+1} = \mathbf{\Phi}^{(2)}(t_n + h) \tag{32c}$$

The growth and matrix factor of the Lie–Trotter splitting are given by:

$$R = \beta \alpha \tag{33a}$$

 $Q = \boldsymbol{\beta} \boldsymbol{A}^* \boldsymbol{a} + \boldsymbol{B}^* \boldsymbol{b} \tag{33b}$

Eq. (34) shows that Lie–Trotter splitting is first-order accurate globally and second-order accurate locally:

$$\mathbf{R}(h) = \boldsymbol{\beta}\boldsymbol{\alpha}$$

= $\mathbf{I} + (\mathbf{A} + \mathbf{B})h + (\mathbf{A}^2 + \mathbf{B}^2)\frac{h^2}{2} + \mathcal{O}(h^3)$ (34)

The series expansion of Eq. (7) for Lie–Trotter splitting indicates a first-order dependence of the steady-state solution on the time step size:

$$\boldsymbol{\Phi}_{\infty} = -(\boldsymbol{a} + \boldsymbol{b})(\boldsymbol{A} + \boldsymbol{B})^{-1} + (\boldsymbol{A}\boldsymbol{b} - \boldsymbol{B}\boldsymbol{a})(\boldsymbol{a} + \boldsymbol{b})^{-1}\frac{h}{2} + \mathcal{O}(h^2) \quad (35)$$

The limit $h \rightarrow \infty$ of Eq. (7) for the Lie–Trotter splitting tends towards the solution of the chemical sub-problem:

$$\boldsymbol{\Phi}_{\infty} = -\boldsymbol{b}\boldsymbol{B}^{-1} \tag{36}$$

3.6. Adaptive splitting

The Adaptive splitting [24,25] uses a different approach than the previous schemes. The splitting scheme integrates both subproblems independently and subsequently sums both solutions. The solution for the new time step is obtained by subtracting the initial value from this sum:

$$d_t \boldsymbol{\Phi}^{(1)} = \boldsymbol{\mathcal{T}}(\boldsymbol{\Phi}^{(1)}), \quad \boldsymbol{\Phi}^{(1)}(t_n) = \boldsymbol{\Phi}_n \tag{37a}$$

$$d_t \mathbf{\Phi}^{(2)} = \mathcal{R}(\mathbf{\Phi}^{(2)}), \quad \mathbf{\Phi}^{(2)}(t_n) = \mathbf{\Phi}_n \tag{37b}$$

$$\mathbf{\Phi}_{n+1} = \mathbf{\Phi}^{(1)}(t_n + h) + \mathbf{\Phi}^{(2)}(t_n + h) - \mathbf{\Phi}_n$$
(37c)

The growth and matrix factor of the Adaptive splitting are given by:

$$\boldsymbol{R} = \boldsymbol{\alpha} + \boldsymbol{\beta} - \boldsymbol{I} \tag{38a}$$

$$\mathbf{Q} = \mathbf{A}^* \mathbf{a} + \mathbf{B}^* \mathbf{b} \tag{38b}$$

Similar to Lie–Trotter, the local and global accuracies of the Adaptive splitting are second and first-order, respectively:

$$\mathbf{R}(h) = \boldsymbol{\alpha} + \boldsymbol{\beta} - \mathbf{I}$$

= $\mathbf{I} + (\mathbf{A} + \mathbf{B})h + (\mathbf{A}^2 + \mathbf{B}^2)\frac{h^2}{2} + \mathcal{O}(h^3)$ (39)

The series expansion of Eq. (7) for Adaptive splitting indicates first-order dependence of the steady-state solution on the time step size:

$$\Phi_{\infty} = -(\boldsymbol{a} + \boldsymbol{b})(\boldsymbol{A} + \boldsymbol{B})^{-1} - (\boldsymbol{A} - \boldsymbol{B})(\boldsymbol{A}\boldsymbol{b} - \boldsymbol{B}\boldsymbol{a})(\boldsymbol{A} + \boldsymbol{B})^{-2}\frac{h}{2} + \mathcal{O}(h^2)$$
(40)

The limit $h \rightarrow \infty$ of Eq. (7) for Adaptive splitting is, similar to Staggered splitting, a linear combination of the sub-system solutions:

$$\boldsymbol{\Phi}_{\infty} = -\frac{1}{2}\boldsymbol{a}\boldsymbol{A}^{-1} - \frac{1}{2}\boldsymbol{b}\boldsymbol{B}^{-1} \tag{41}$$

4. New steady-state preserving operator splitting schemes

Steady-state preserving versions of the Staggered (Section 3.4), Adaptive (Section 3.6), and Lie–Trotter (Section 3.5) splitting scheme are derived in this section. The procedure to obtain steadystate preservation is based on balancing constants similar to Speth et al. [20] who proposed Balanced splitting (Section 3.2), a steadystate preserving version of Strang splitting (Section 3.1).

The balancing constants were chosen based on i) the initial results of preliminary investigations and ii) the tendency of the steady-state solution (Eq. (30), Eq. (41), and Eq. (36)) of the corresponding non-conservative splitting schemes. The mixing derivative was chosen as a balancing constant when the preliminary tests indicated a dominance of the chemical sub-problem, e.g., early ignition or failed extinction. In case of inconclusive results of the preliminary tests (e.g., Strang and Lie–Trotter scheme), the tendency of the steady-state solution was also considered in the choice of the balancing constant. Preliminary investigations revealed that balancing constants based on the derivative of the steady-state solution's tendency give steady-state preserving splitting schemes.

4.1. Consistent staggered splitting

Introducing a balancing constant to Staggered splitting (Eq. (26)) alters the steady-state solution and ensures consistency. The initial mixing derivative proved to be a suitable choice for the balancing constant:

$$\mathcal{C}_{N} = -\mathcal{T} \tag{42a}$$

$$d_t \boldsymbol{\Phi}^{(1)} = \boldsymbol{\mathcal{R}}(\boldsymbol{\Phi}^{(1)}) - \boldsymbol{\mathcal{C}}_{\boldsymbol{N}}, \quad \boldsymbol{\Phi}^{(1)}(t_n) = \boldsymbol{\Phi}_{n+1/2}$$
(42b)

$$d_t \Phi^{(2)} = \mathcal{T}(\Phi^{(2)}) + \mathcal{C}_N, \quad \Phi^{(2)}(t_n + h/2) = \Phi^{(1)}(t_n + h)$$
 (42c)

$$\Phi_{n+1} = \frac{\Phi^{(1)}(t_n+h) + \Phi^{(2)}(t_n+3/2h)}{2}$$
(42d)

Introducing the balancing constant makes the first step's special treatment redundant since it is in equilibrium anyway. Similar to the original Staggered splitting scheme (Section 3.4), the representative time step starting with a half mixing step is used for the subsequent analysis. The growth and matrix factors for the steadystate preserving splitting scheme are given by:

$$\boldsymbol{R} = \frac{1}{2} \Big[\boldsymbol{\alpha} \boldsymbol{\beta} - \boldsymbol{\alpha} + \boldsymbol{\beta} + \boldsymbol{I} + (\boldsymbol{\alpha} + \boldsymbol{I}) \boldsymbol{B}^* \boldsymbol{A} \Big]$$
(43a)

$$\mathbf{Q} = \frac{1}{2} [(\boldsymbol{\alpha} \boldsymbol{A} + \boldsymbol{I}) \boldsymbol{B}^* (\boldsymbol{a} + \boldsymbol{b})]$$
(43b)

The global and local accuracy orders remain almost unaffected despite modifying the steady-state solution and the numerical algorithm. Similar to the original Staggered splitting, Consistent Staggered splitting misses a term for second-order accuracy (A^2) :

$$\mathbf{R}(h) = \frac{1}{2} \Big[\boldsymbol{\alpha} \boldsymbol{\beta} - \boldsymbol{\alpha} + \boldsymbol{\beta} + \mathbf{I} + (\boldsymbol{\alpha} + \mathbf{I}) \boldsymbol{B}^* \boldsymbol{A} \Big]$$

= $\mathbf{I} + (\boldsymbol{A} + \boldsymbol{B})h + (\boldsymbol{A} \boldsymbol{B} + \boldsymbol{B} \boldsymbol{A} + \boldsymbol{B}^2) \frac{h^2}{2}$
+ $\mathcal{O}(h^3)$ (44)

4.2. Consistent Lie-Trotter splitting

Consistent Lie-Trotter splitting (Eq. (32)) can be achieved by introducing a balancing constant based on the chemical sub-system: The splitting scheme, thus, changes to:

$$\mathcal{C}_{N} = \mathcal{R} \tag{45a}$$

$$d_t \mathbf{\Phi}^{(1)} = \mathcal{T}(\mathbf{\Phi}^{(1)}) + \mathcal{C}_N, \quad \mathbf{\Phi}^{(1)}(t_n) = \mathbf{\Phi}_n$$
(45b)

$$d_t \Phi^{(2)} = \mathcal{R}(\Phi^{(2)}) - \mathcal{C}_N, \quad \Phi^{(2)}(t_n) = \Phi^{(1)}(t_n + h)$$
(45c)

$$\boldsymbol{\Phi}_{n+1} = \boldsymbol{\Phi}^{(2)}(t_n + h) \tag{45d}$$

The growth and matrix factors of Consistent Lie-Trotter splitting are given by:

$$\mathbf{R} = \boldsymbol{\beta}\boldsymbol{\alpha} - \boldsymbol{\beta} + \mathbf{I} + \boldsymbol{\beta}\mathbf{A}^*\mathbf{B}$$
(46a)

$$\mathbf{Q} = \boldsymbol{\beta} \mathbf{A}^* (\boldsymbol{a} + \boldsymbol{b}) \tag{46b}$$

As a result of adding the splitting constant, the steady-state is preserved, while the global and local accuracy orders remain virtually unchanged:

$$\mathbf{R}(h) = \boldsymbol{\beta}\boldsymbol{\alpha} - \boldsymbol{\beta} + \mathbf{I} + \boldsymbol{\beta}\mathbf{A}^*\mathbf{B}$$

= $\mathbf{I} + (\mathbf{A} + \mathbf{B})h + (\mathbf{A}^2 + \mathbf{A}\mathbf{B} + 2\mathbf{B}\mathbf{A} + \mathbf{B}^2)\frac{h^2}{2}$
+ $\mathcal{O}(h^3)$ (47)

4.3. Consistent adaptive splitting

Adaptive splitting requires a balancing constant based on the mixing derivative to ensure steady-state consistency. The modified scheme is given by:

$$\mathcal{C}_{\mathbf{N}} = -\mathcal{T} \tag{48a}$$

$$d_t \boldsymbol{\Phi}^{(1)} = \mathcal{T}(\boldsymbol{\Phi}^{(1)}) + \mathcal{C}_{\boldsymbol{N}}, \quad \boldsymbol{\Phi}^{(1)}(t_n) = \boldsymbol{\Phi}_n$$
(48b)

$$d_t \mathbf{\Phi}^{(2)} = \mathcal{R}(\mathbf{\Phi}^{(2)}) - \mathcal{C}_{\mathbf{N}}, \quad \mathbf{\Phi}^{(2)}(t_n) = \mathbf{\Phi}_n \tag{48c}$$

$$\mathbf{\Phi}_{n+1} = \mathbf{\Phi}^{(1)}(t_n + h) + \mathbf{\Phi}^{(2)}(t_n + h) - \mathbf{\Phi}_n$$
(48d)

The growth and matrix factors of Consistent Adaptive splitting are given by:

$$\boldsymbol{R} = \boldsymbol{\alpha} + \boldsymbol{\beta} - \boldsymbol{I} + (\boldsymbol{B}^* - \boldsymbol{A}^*)\boldsymbol{A}$$
(49a)

$$\boldsymbol{Q} = \boldsymbol{B}^*(\boldsymbol{a} + \boldsymbol{b}) \tag{49b}$$

The global and local accuracy orders are slightly affected by the splitting constants but are still first-order globally and secondorder locally accurate:

$$\mathbf{R}(h) = \mathbf{\alpha} + \mathbf{\beta} - \mathbf{I} + (\mathbf{B}^* - \mathbf{A}^*)\mathbf{A}$$

= $\mathbf{I} + (\mathbf{A} + \mathbf{B})h + (\mathbf{B}\mathbf{A} + \mathbf{B}^2)\frac{h^2}{2}$
+ $\mathcal{O}(h^3)$ (50)

5. Stability analysis

Stability is a critical issue of splitting schemes for reactiondiffusion(-advection) processes. The nature of chemistry leads to unbalanced characteristics between the sub-processes and can require minuscule time step sizes. This partial decoupling of mixing and chemistry poses a challenging problem for operator splitting schemes. Speth et al. [20] and Wu et al. [19] defined three different limiting cases for reaction-diffusion(-advection) systems for the linear scalar stability analysis:

i) large time step limit:

 $\lim_{\infty} R \Big|_{A,B}$

ii) chemistry becomes dominant over mixing at constant time step size:

$$\lim_{|B|\to\infty} R \bigg|_{A,h}$$

iii) chemical rates increase while the time step decreases (keep |B|h = const.):

$$\lim_{|B|\to\infty} R \bigg|_{A,|B|h}$$

These limiting cases can be evaluated based on the recurrence expressions of the splitting schemes. The stability properties of the different splitting schemes are given and discussed below. The term well-posed is subsequently used for stable matrices, e.g., matrices having eigenvalues with negative real parts and negative values for the scalar case.

5.1. Strang splitting

Considering the three cases, Strang splitting's stability is unconditional for large time steps and stiff chemistry operators. Stability for reducing time steps at increasing stiffness is a function of β :

$$\lim_{h \to \infty} R \Big|_{A,B} = 0 \tag{51a}$$

$$\lim_{|B| \to \infty} R \bigg|_{A,h} = 0 \tag{51b}$$

$$\lim_{|B| \to \infty} R \bigg|_{A,|B|h} = \beta \tag{51c}$$

The limit implies that Strang splitting is stable for any problem as long as the chemistry sub-problem is well-posed.

5.2. Balanced splitting

Wu et al. [19] investigated the stability of Balanced splitting and arrived at the following relations:

$$\lim_{h \to \infty} R \Big|_{A,B} = \frac{1}{2} (1 - A^{-1}B)$$
(52a)

$$\lim_{|B|\to\infty} R \bigg|_{A,h} = \frac{1}{2} \left(1 - (\alpha - 1)A^{-1}B \right) = \infty$$
(52b)

$$\lim_{|B| \to \infty} R \bigg|_{A,|B|h} = \frac{1}{4} (1+\beta)(2+Bh)$$
(52c)

These relations indicate that Balanced splitting is unstable for large step sizes if |B| > 3|A| and if the chemistry operator becomes stiff while keeping the step size constant. In case step size is reduced, Balanced splitting is stable for any B < 0 only if |B|h < 5.99.

5.3. Simpler splitting

Similar to Balanced splitting, the stability of Simpler splitting was already assessed by Wu et al. [19]:

$$\lim_{h \to \infty} R \Big|_{A,B} = 1 \tag{53a}$$

$$\lim_{|B| \to \infty} R \bigg|_{A,h} = 1 - \alpha^2$$
(53b)

$$\lim_{|B| \to \infty} R \bigg|_{A \mid B \mid h} = \beta$$
(53c)

Changing the balancing constant compared to Balanced splitting significantly improved the splitting scheme. Simpler splitting is stable for all h > 0 as long as both sub-problems are well-posed.

5.4. Staggered splitting

Staggered splitting features the same stability properties as Strang splitting:

$$\lim_{h \to \infty} R \Big|_{A,B} = 0 \tag{54a}$$

$$\left. \lim_{|B| \to \infty} R \right|_{A,b} = 0 \tag{54b}$$

$$\lim_{|B| \to \infty} R \bigg|_{A,|B|h} = \beta$$
(54c)

The splitting scheme is stable for all h > 0 provided the chemistry operator is well-posed.

5.5. Lie–Trotter splitting

Lie–Trotter stability analysis also revealed similar properties as Strang and Staggered splitting:

$$\lim_{h \to \infty} R \Big|_{A,B} = 0 \tag{55a}$$

$$\lim_{|B| \to \infty} R \bigg|_{A,h} = 0$$
(55b)

$$\lim_{|B| \to \infty} R \bigg|_{A,|B|h} = \beta$$
(55c)

5.6. Adaptive splitting

Adaptive splitting stability is different. It depends on the stiffness of the mixing and chemistry operator:

$$\lim_{h \to \infty} R \Big|_{A,B} = -1 \tag{56a}$$

$$\lim_{|B| \to \infty} R \bigg|_{A,h} = \alpha - 1$$
(56b)

$$\lim_{|B| \to \infty} R \bigg|_{A,|B|h} = \beta$$
(56c)

Negligible damping occurs for large time steps, while the splitting scheme is stable for $Ah < \ln 2$ if the chemistry operator becomes stiff at constant time step size. If the time step size is reduced, a well-posed chemistry operator ensures stability.

5.7. Consistent staggered splitting

Contrary to Staggered splitting, Consistent Staggered Splitting has more rigorous stability requirements:

$$\lim_{h \to \infty} R \Big|_{A,B} = -\frac{1}{2} \left(1 - AB^{-1} \right)$$
(57a)

$$\lim_{|B| \to \infty} R \bigg|_{A,h} = \frac{1}{2} (1 - \alpha)$$
(57b)

$$\lim_{|B| \to \infty} R \Big|_{A,|B|h} = \beta \tag{57c}$$

Consistent Staggered splitting requires 3A < B and A < 0 for large time steps to be stable. If the chemistry operator becomes stiff, the mixing operator must be well-posed (A < 0). In contrast, the chemistry operator needs to be well-posed for shrinking time steps and increasing stiffness of the operator itself.

5.8. Consistent Lie-Trotter splitting

The Consistent Lie–Trotter scheme has also worse stability compared to the original Lie–Trotter scheme:

$$\lim_{h \to \infty} R \Big|_{A,B} = 1 \tag{58a}$$

$$\lim_{|B| \to \infty} R \bigg|_{A,h} = 1$$
(58b)

$$\lim_{|B| \to \infty} R \bigg|_{A, |B|h} = \infty$$
(58c)

Negligible damping occurs for large time steps and high stiffness of the chemistry operator. However, Consistent Lie–Trotter is unstable for small time steps and high stiffness of the chemistry operator.

5.9. Consistent adaptive splitting

Consistent Adaptive splitting requires $|A| \le |B|$ for large time steps, while it is unconditionally stable if the chemistry operator becomes stiff compared to the mixing one:

$$\lim_{h \to \infty} R \Big|_{A,B} = -AB^{-1} \tag{59a}$$

Table 1					
Summarv	scalar	test	case	settings	[11.19]

j			0.1	· 1·	
	T _{ad}	Ta	T _{in}	T ₀	Da
ignition extinction	1.15 1.15	1.80 1.80	0.15 1.15	0.15 1.00	833.00 15.89

$$\lim_{|B| \to \infty} R \bigg|_{A,h} = 0$$
(59b)

$$\lim_{|B| \to \infty} R \bigg|_{A,|B|h} = \beta$$
(59c)

In case of decreasing time step size and stiff chemistry operator, Consistent Adaptive splitting is stable if the chemistry operator is well-posed.

Strang, Simpler, Staggered, Lie–Trotter, and Adaptive splitting are stable if the transport and chemistry sub-problems are wellposed. Additional requirements regarding the ratio of |A| and |B|have to be satisfied for Consistent Staggered and Consistent Adaptive splitting for large time steps. Consistent Lie–Trotter is unstable if the chemistry operator becomes stiff and the time step size is reduced. Balanced splitting is unstable if the chemistry operator becomes stiff while the time step size stays constant and impose a limitation for the |B|h term in case the time step size is reduced. These findings indicate that balancing constants impair the stability of splitting schemes.

6. Scalar test case

The operator splitting schemes are tested with a scalar case equivalent of a dimensionless PSR. The case has also been used by Lu et al. [11], Wu et al. [19] to test the accuracy and steady-state convergence of operating splitting schemes and is known to cause problems for Strang splitting [11]. The scalar test case's governing equation is given by Law [26]:

$$\frac{dT}{dt} = \frac{1}{Da} \left(T_{in} - T \right) + \left(T_{ad} - T \right) \exp\left(-\frac{T_a}{T} \right)$$
(60)

where *t*, *T*, *T*_{in}, *T*_{ad}, and *T*_a are time, temperature, inlet temperature, adiabatic reaction temperature, and the activation temperature, which is equal to the activation energy over the ideal gas constant (E_a/\Re).

An ignition and extinction problem is used to test the stability and accuracy of the splitting schemes discussed in Sections 3 and 4. The subsequent computations were carried out in python [27] using NumPy [28] and SciPy [29]. The settings for the different cases are given in Table 1. Lu et al. [11] determined that the critical Da numbers for the ignition and extinction cases were 832.84 and 15.90, respectively.

The Courant number [30,31] for the test cases is defined as the ratio between the residence time (τ_{res}) and the time step size (*h*). For the presented scalar test case, this relation can also be expressed using *Da*:

$$Co = \frac{h}{\tau_{res}} = \frac{h}{Da}$$
(61)

The Co number limit is some numerical efficiency measure and values above unity are usually prohibitive due to loss of information [30,31]. In general, schemes requiring low Co for stability need more sub-steps to solve the problem than schemes allowing high Co. However, the accuracy of the splitting schemes will increase compared to the direct integration (DI) for lower Co numbers due to their temporal accuracy.



Fig. 2. Comparison of operator splitting schemes and direct integration for the scalar ignition case for a Co number of 0.1.

Balanced splitting was skipped in the subsequent investigations because it proved to be unstable for Co > 0.05 in the ignition case. This is might be caused by the required limitation of the mixture to chemical rate ratio [19] (Eq. (52)).

Figures 2 and 3 show the solution of the ignition problem for the different splitting schemes and Co numbers of 0.1 and 0.5. In case of Co = 0.1, DI predicts the ignition after around 350 times Da time and a final normalized temperature of 1.144. Strang, Staggered, Adaptive, and Consistent Lie-Trotter under-predict the steady-state temperature by up to 22%, while Lie-Trotter slightly over-predicts the steady-state temperature (0.5%). Simpler, Consistent Staggered, and Consistent Adaptive splitting predict the same steady-state temperature as DI. The temporal evolution of the temperature is correctly captured by Simpler and Consistent Staggered splitting. All other splitting schemes predict an early ignition compared to DI. In case of Co = 0.5, under-prediction of the steady-state temperature worsens for Strang, Staggered, and Adaptive splitting, while it improves for Consistent Lie-Trotter splitting. Simpler, Consistent Staggered, and Consistent Adaptive predict the correct steady-state. All splitting schemes predict earlier ignition for the Co = 0.5 case compared to the Co = 0.1 case.

Figures 4 and 5 compare the results of the extinction test case for the different splitting schemes with DI for Co numbers of 0.1 and 0.5. Staggered and Adaptive splitting fail to predict extinction. On the contrary, Strang, Lie–Trotter, Consistent Adaptive, and Consistent Lie–Trotter predict earlier extinction than the DI case for the Co = 0.1 case. Similar to the ignition case, Simpler and Consistent Staggered splitting are the only schemes to correctly predict the temperature profile. The results for the Co = 0.5 case are in line with the results of the ignition case and the Co = 0.1 extinction case. Extinction is predicted earlier for all splitting schemes





Fig. 3. Comparison of operator splitting schemes and direct integration for the scalar ignition case for a Co number of 0.5.



Fig. 4. Comparison of operator splitting schemes and direct integration for the scalar extinction case for a Co number of 0.1.



Fig. 5. Comparison of operator splitting schemes and direct integration for the scalar extinction case for a Co number of 0.5.

compared to the Co = 0.1 case, but Simpler and Consistent Staggered are closest to the DI solution.

Figure 6 compares the relative steady-state error versus the Co number for the scalar ignition and extinction case. The steady-state error for the ignition case (Fig. 6(a)) is negligibly small for Simpler, Consistent Staggered, and Consistent Adaptive. At the same time, the time step size dependence is confirmed for Strang, Staggered, and Adaptive splitting. Lie–Trotter splitting over-predicts the steady-state temperature by 0.5% for all but the smallest investigated Co numbers. Consistent Lie–Trotter splitting also overpredicts the correct steady-state for large Co numbers. However, after under-predicting the steady-state around Co = 0.1, it predicts the correct steady-state for smaller Co numbers.

Figure 6(b) compares the steady-state results of the scalar extinction case. In general, the correct steady-state is predicted by all steady-state preserving schemes. Consistent Staggered and Consistent Adaptive splitting become unstable for Co > 1, while Consistent Lie–Trotter becomes unstable for Co > 2. All three schemes give oscillating steady-states when becoming unstable due to not fulfilling stability criterion i) from Section 5. Strang splitting under and Lie–Trotter over-predicts the steady-state for high Co numbers, while both approach the correct steady-state for small Co numbers. Adaptive splitting fails to predict extinction for Co > 0.0001 and starts to become unstable for Co > 0.5 due to the negligible damping for large time step sizes. Staggered splitting gives the correct steady-state for Co < 0.001 and Co > 1 but fails to predict extinction between them.

Figure 7 shows the absolute ignition (a) and extinction (b) time errors versus the Co number for the investigated splitting schemes. The Co > 2 cases are neglected due to stability issues of the schemes. The temporal accuracy shows similar characteristics for the different splitting schemes in both cases. Consistent Adaptive and Consistent Lie–Trotter overlap in the diagrams and show firstorder accuracy over the investigated range. Staggered and Adap-



Fig. 6. Comparison of relative steady-state splitting error versus Co number for the scalar ignition (a) and extinction (b) cases. Unstable cases are not shown.



Fig. 7. Comparison of relative ignition (a) and extinction (b) time error versus Co for the scalar test cases. Unstable cases are not shown. The lines indicate first (dotted) and second (dashed) order. Strang and Lie–Trotter as well as Consistent Adaptive and Consistent Lie–Trotter overlap.

tive splitting approach first-order accuracy for Co < 0.001 but show lower-order accuracy for higher Co numbers. The accuracy of the Adaptive splitting scheme remains unknown for the extinction case because it predicted extinction only for one case. Strang and Lie–Trotter splitting also overlap and show first-order accuracy for Co > 0.1, while being second-order accurate for smaller Co numbers. Simpler and Consistent Staggered show approximately second-order accuracy.

In general, the scalar test case results are in line with the theoretic observations from Sections 3 to 5. Larger time steps decrease the accuracy of the predicted temporal evolution and affect the steady-state results of the non-steady-state preserving splitting schemes.

7. Combustion test case

The combustion test cases are H radical doped ignition and near-limit extinction [11]. The ignition case is known to cause problems for operating splitting schemes [12]. The governing equations are given by:

$$\frac{dY_i}{dt} = \frac{1}{\tau_{res}} \left(Y_{i,in} - Y_i \right) + \frac{\dot{\omega}_i}{\rho}$$
(62a)

$$\frac{dh}{dt} = \frac{1}{\tau_{res}} \sum_{i=1}^{N_s} Y_{i,in}(h_{i,in} - h_i) + \frac{1}{\rho} \sum_{i=1}^{N_s} \dot{\omega}_i h_i$$
(62b)

where Y, τ_{res} , $\dot{\omega}$, ρ , *h*, and *N*_s are the species mass fraction, residence time, species consumption/production rate, density, enthalpy, and number of species. The hydrogen combustion mechanism of Li et al. [32] was used for the thermodynamic properties and chemistry rates. The computations were done in python [27] using NumPy [28], SciPy [29], and the open-source tool Cantera [17].

The ignition case features a hydrogen/air mixture with an equivalence ratio (ϕ) of 0.5 with a $0.1\%_{\frac{\text{mol}}{\text{mol}}}$ H radical enrichment. The initial reactor state is equal to the equilibrium concentrations at constant temperature and pressure of the inlet stream. In contrast, the initial reactor state is equal to the equilibrium at constant enthalpy and pressure of the inlet stream for the extinction case. The inlet states of both cases are summarized in Table 2. The critical residence time for the extinction case is 1.4211×10^{-7} s, while the ignition case is far from the critical residence time of 1.0710×10^{-3} s and features H enrichment [11].

The Balanced splitting scheme has been disregarded for the investigations of the combustion test cases for the same reasons it was omitted in the scalar test cases: Balanced splitting proved to be unstable for Co > 0.05 for the investigated test cases.

Figures 8 and 9 compare the temperature profiles of the ignition case of the different splitting schemes for Co numbers of 0.1 and 0.5. The results compare with the scalar ignition case of Section 6. Simpler and Consistent Staggered splitting give similar results as DI for a Co number of 0.1, while Consistent Adaptive splitting predicts the correct steady-state but early ignition. Strang, Staggered, Lie-Trotter, and Adaptive splitting predict late ignition and fail to predict the steady-state temperature. Strang, Staggered, and Adaptive splitting under-predict the steady-state temperature, while Lie-Trotter splitting over-predicts it. Contrary to the scalar ignition case, Consistent Lie-Trotter splitting fails to predict ignition. This might be related to the balancing constant based on the chemistry derivative. For a Co number of 0.5, all splitting schemes except Simpler, Consistent Staggered, Consistent Adaptive, and Adaptive fail to predict the ignition process. Adaptive splitting significantly under-predicts the steady-state temperature compared to DI, while the other splitting schemes reproduce the correct steady-state. Adaptive splitting also predicts delayed ignition,

Table 2Summary combustion test case settings [11,12].

	T _{in} (K)	p (atm)	H ₂ (kg/kg)	H (kg/kg)	$N_2 \ (kg/kg)$	O ₂ (kg/kg)	τ_{res} (s)
ignition	875	80	0.170	0.023	0.637	0.170	$\begin{array}{l} 2.00 \times 10^{\text{-6}} \\ 1.42 \times 10^{\text{-7}} \end{array}$
extinction	1000	20	0.296	-	0.556	0.148	



Fig. 8. Comparison of operator splitting schemes and direct integration for the ignition case for a Co number of 0.1.

while the other schemes predict premature ignition. It is interesting to note that the solution of both Adaptive splitting schemes overshoot prior to reaching the steady-state. The same behavior was observed by Lu et al. [11] for the Balanced splitting.

Figures 10 and 11 compare the temperature profiles of the extinction case of the different splitting schemes for Co numbers of 0.1 and 0.5. In general, the results are comparable to the scalar extinction case. For a Co number of 0.1, Staggered and Adaptive splitting fail to predict extinction. Furthermore, Strang, Lie–Trotter, Consistent Lie–Trotter, and Consistent Adaptive under-predict the extinction time, while Simple and Consistent Staggered match the DI results. For a Co number of 0.5, besides Staggered and Adaptive splitting, Consistent Lie–Trotter splitting fails to predict extinction. The remaining splitting schemes, Strang, Lie–Trotter, Consistent Adaptive, Consistent Staggered, and Simpler, predict earlier extinction onset than the Co = 0.1 case.

Figure 12 compares the relative steady-state error versus the Co number for the ignition and extinction case. In principle, the results resemble the scalar test case for the ignition and extinction case. Figure 12(a) indicates that the steady-state preserving schemes predict the correct steady-states for the ignition case, while the time step dependency is indicated for the other schemes. The stability limit of the splitting schemes is shifted towards lower Co numbers compared to the scalar test cases. The investigated splitting schemes become unstable for Co > 2, except for Consis-



Fig. 9. Comparison of operator splitting schemes and direct integration for the ignition case for a Co number of 0.5.

tent Adaptive splitting, which becomes unstable for Co > 0.5. The ignition cases reveal a significant accuracy increase for the nonpreserving splitting schemes around Co = 0.4. All splitting schemes predict the correct steady-state for Co < 0.01. The Consistent Lie– Trotter results deviate from the scalar test cases because it significantly under-predicts the correct steady-state for Co > 0.01. Figure 12(b) compares the relative steady-state errors for the extinction case. Adaptive and Strang splitting fail to predict extinction for 0.001 > Co < 2. In addition, Consistent Lie–Trotter fails to predict the correct steady-state for Co > 0.25, while Consistent Adaptive is unstable for Co > 1.5.

Figure 13 investigates the temporal accuracy for the combustion ignition and extinction cases. The extinction case results are in line with the scalar accuracy results. In contrast, the ignition case results differ from their scalar counterpart, and all investigated splitting schemes show similar orders of accuracy. The different accuracies compared to the scalar case and the analytical results are caused by the nature of the ignition test case since it is particularly challenging for splitting schemes [11]. The deviation of the ignition case from the scalar and theoretical results indicates that the problem properties also affect the accuracy of splitting schemes.

In general, the combustion case results are in line with the theoretical and scalar test case results. Consistent Lie–Trotter splitting is an exception because the linear scalar stability analysis suggests that the scheme is steady-state conservative. One reason for the



Fig. 10. Comparison of operator splitting schemes and direct integration for the extinction case for a Co number of 0.1.



Fig. 11. Comparison of operator splitting schemes and direct integration for the extinction case for a Co number of 0.5.



Fig. 12. Comparison of relative steady-state splitting error versus Co number for the ignition (a) and extinction (b) cases. Unstable cases are not shown.



Fig. 13. Comparison of relative ignition (a) and extinction (b) time error versus Co for the combustion test cases. Unstable cases are not shown. The lines indicate first (dotted) and second (dashed) order. Strang and Lie–Trotter as well as Consistent Adaptive and Consistent Lie–Trotter overlap.

different characteristics could be caused by shortcomings of the linear analysis for the non-linear chemical sub-system. The differences between the scalar test case and the combustion test case also indicate that the non-linear matrix has additional effects on the operator splitting schemes compared to the non-linear scalar operator. Moreover, the ignition case reveals an accuracy dependence of operator splitting schemes on the problem properties.

8. Summary and conclusion

Stability and steady-state preservation of operator splitting schemes for reaction-diffusion problems were examined. These investigations showed that Balanced and Simpler splitting preserve the correct steady-state solution independently of the time step size.

Three new steady-state preserving splitting schemes were proposed based on the initial evaluation of the existing splitting. The new schemes are based on the Staggered, Lie–Trotter, and Adaptive splitting schemes and feature suitable balancing constants to preserve the steady-state.

All discussed schemes were also tested using test cases known to cause problems for operator splitting. The cases consist of ignition and extinction problems based on a dimensionless PSR case and a hydrogen combustion case. The results confirmed steadystate preservation of Simpler, Consistent Staggered, and Consistent Adaptive splitting, while the Consistent Lie-Trotter splitting failed to conserve the steady-state. The the balancing constant is the difference between the Consistent Lie-Trotter and the other schemes. The former uses the chemical derivative as balancing constant, while the others use the mixing derivative. Furthermore, the temporal and steady-state accuracies of the schemes were investigated for Co numbers between 0.0001 and 10. In principle, the numerical result confirmed the theoretical ones, except for the hydrogen combustion ignition case, where all schemes have similar accuracies. This result indicates an influence of the problem properties on the actual splitting scheme order of accuracy.

The scalar and combustion test cases showed that Simpler and Consistent Staggered splitting should be used for transient combustion simulations. Simpler splitting is second-order accurate, while Consistent Staggered splitting is first-order accurate but gives significantly better results than Consistent Adaptive splitting. Consistent Adaptive splitting can be used for steady-state solution algorithms since it is only first-order accurate in time but also steady-state preserving.

The differences between the linear scalar analysis and the numerical test cases indicate that the linear approximation incorrectly captures the non-linear chemistry operator. In addition, the differences between the scalar and combustion test cases also indicate that scalar analysis is insufficient to capture the non-linear chemistry operator. Future work should focus on non-linear stability analysis of operator splitting schemes to capture non-linear and matrix effects.

Declaration of Competing Interest

The author declares that he has no known competing financial interests or personal relationships that could have influenced the work reported in this paper.

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