Supplementary material: Chemical kinetics and transport properties [1]

A Chemistry reduction and validation[1]

The Analytically Reduced Chemistry (ARC) used in this study was derived to achieve an accurate description of the combustion process of methane/air/hydrogen flames for several mixing conditions. The reduction was carried out with the ARCANE code co-developed by CERFACS and Cornell University [2]. It is a fully automatic multi-step reduction tool relying on DRGEP [3], chemical lumping [4] and Quasi-Steady State assumption [5].

The well known GRI-Mech 3.0 scheme [6] was chosen as detailed reference mechanism. This scheme comprises 53 species and 325 reactions.

To guarantee the accuracy of the mechanism, several canonical cases were included in the sampled data needed for the reduction. These cases and the thresholds errors associated on interest quantities are detailed in Tab. A.1.

Reactor type	Isochoric 0D reactor	Freely propagating premixed flame
Temperature [K]	1000	300
Pressure [bar]	1	1
Equivalence ratio	1	0.6, 1, 1.5
Error threshold on auto-ignition delay time	5%	/
Error threshold on laminar flame speed	/	5%
Error threshold on maximum heat release rate	/	5%
Error threshold on maximum temperature	1%	1%

Table A.1: Table summarizing the cases used for the reduction and the associated threshold errors.

For each target case, three fuel compositions were considered: pure hydrogen, pure methane and a blend of 50% of hydrogen and 50% of methane in terms of mole fractions. For all cases air was considered as oxidizer. Consequently, the entire reduction database comprises 12 cases, i.e., 4

[1]For addition information on the ARC mechanism and its reduction procedure please contact Q. Cazères. *Email address*: cazeres@cerfacs.fr (personal adress: quentin.cazeres@gmail.com)

operating conditions multiplied by 3 different fuels. To improve precision, different targets were set for each fuel as indicated in Tab. A.2.

Fuels	Hydrogen (H_2)	Methane (CH_4)	Blend (50% H_2 + 50% CH_4)
Targets	H_2, H_2O, Heat release rate	CH_4, CO, CO_2, Heat release rate	H_2, CH_4, CO, CO_2, Heat release rate

Table A.2: Table summarizing the target quantities used for the reduction for each fuel.

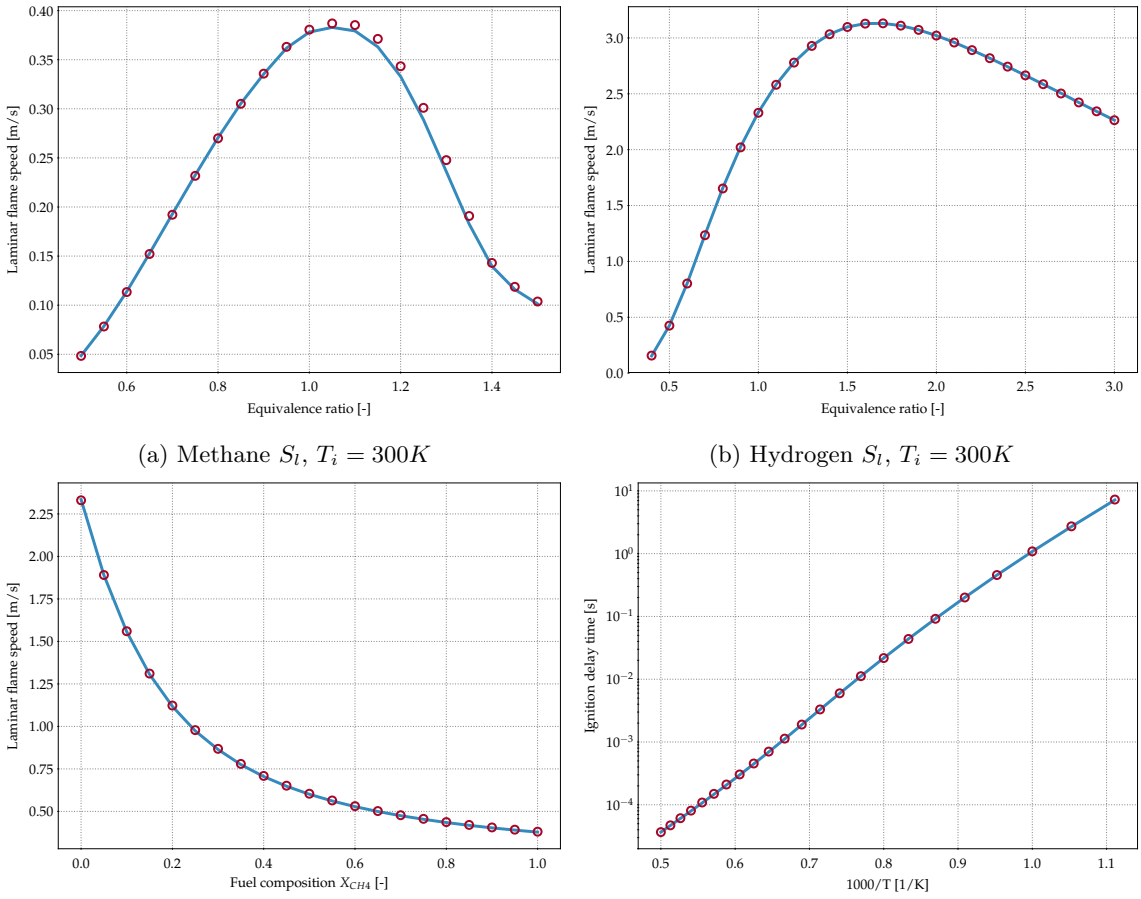
The reduced ARC mechanism generated by ARCANE comprises 20 transported species, 166 reactions and 9 species in Quasi-Steady State:

- The transported species are H_2, H, O, O_2, OH, H_2O, HO_2, H_2O_2, CH_3, CH_4, CO, CO_2, CH_2O, CH_3O, CH_3OH, C_2H_2, C_2H_4, C_2H_6, CH_2CO, N_2.
- The Quasi-Steady State species are CH, CH_2, CH_2(S), HCO, CH_2OH, C_2H_3, C_2H_5, HCCO, CH_2CHO.

A compleate description of the reactions is given in Tab. A.3 and in the attached *ARC.csv* file.

The mechanism was validated on a broader operability spectrum compared to reduction cases. Results of the validation are displayed in Fig. A.1 showing a really good agreement between the ARC and the detailed mechanism with the maximum error on interest quantities being less than 5%. An additional validation specific for the MIRADAS test rig studied in this work was also performed. Indeed, as discussed in the manuscript, the flame developed inside this system is characterized by the presence of a hydrogen/methane-air burnt premix diffusion branch. This is here represented by a counter-flow diffusion flame with pure hydrogen at room temperature coming from one side and equilibrium products of $\phi = 0.8$ mixture of methane and air at adiabatic flame temperature on the other side. The same solution is presented in the manuscript (Fig. 2) considering, however, preheating of the hydrogen. Temperature and heat release rate profiles predicted by the ARC mechanism are found in excellent agreement with the ones obtained with the detailed mechanism as shown in Fig. A.2(a-b).

Concluding, the afore discussed validations give confidence to the authors in the capabilities of the derived ARC mechanism to represent correctly the complex combustion process of the investigated system.



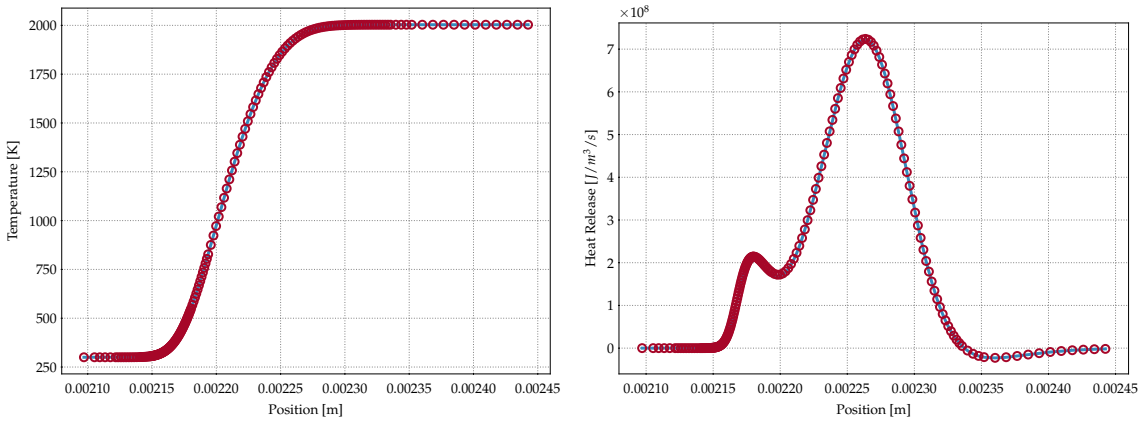
(a) Methane S_l, $T_i = 300K$

(b) Hydrogen S_l, $T_i = 300K$

(c) S_l for varying fuel composition at $\phi = 1$, $T_i = 300K$

(d) Methane τ_{ig} at $\phi = 1$

Figure A.1: Validation of the ARC mechanism on canonical cases. Laminar flame speed S_l as function of the equivalence ratio ϕ for pure methane/air (a) and pure hydrogen/air (b) flames. (c) S_l for varying fuel composition at $\phi = 1$. (d) Methane auto-ignition delay time τ_{ig} at $\phi = 1$. In all plots, blue line refers to results obtained with the GRI-Mech 3.0 mechanism while ARC predictions are indicated by the red circles. For all cases, the pressure is 1 bar.



(a) Temperature profile

(b) Heat Release rate profile

Figure A.2: Validation of the mechanism on canonical cases. Temperature (a) and heat release rate (b) profiles for a counterflow diffusion flame of H_2 ($T^i_{H_2}$ = 300 K) against the equilibrium burnt gases from lean ($\phi = 0.8$) CH_4/Air premixed flame (T^i_g = 2000 K). Blue line refers to results obtained with the GRI-Mech 3.0 mechanism while ARC predictions are indicated by the red circles. Width between the two inlets is 0.04 m but plots are zoomed to show the reactive zone.

Table A.3: List of reactions with A the pre-exponential factor in $m^{3(n-1)})/kmol^{n-1)}/s$ with n the order of the reaction, b the temperature exponent and Ea the activation energy in J/kmol. In the case of fall-off reactions, two sets of Arrhenius coefficients are specified, the first one being the low temperature set and the second one the high temperature set.

No.	Reaction	A	b	E_a
1	H + O + M ⟷ OH + M	5.000000E+11	-1.000000E+00	0.000000E+00
	C2H6:3.00E+00 CH4:2.00E+00 CO:1.50E+00 CO2:2.00E+00 H2:2.00E+00 H2O:6.00E+00			
2	H2 + O ⟷ H + OH	3.870000E+01	2.700000E+00	2.619184E+07
3	HO2 + O ⟷ O2 + OH	2.000000E+10	0.000000E+00	0.000000E+00
4	H2O2 + O ⟷ HO2 + OH	9.630000E+03	2.000000E+00	1.673600E+07
5	CH + O ⟷ CO + H	5.700000E+10	0.000000E+00	0.000000E+00
6	CH2 + O ⟷ H + HCO	8.000000E+10	0.000000E+00	0.000000E+00
7	CH2(S) + O ⟷ CO + H2	1.500000E+10	0.000000E+00	0.000000E+00
8	CH2(S) + O ⟷ H + HCO	1.500000E+10	0.000000E+00	0.000000E+00
9	CH3 + O ⟷ CH2O + H	5.060000E+10	0.000000E+00	0.000000E+00
10	CH4 + O ⟷ CH3 + OH	1.020000E+06	1.500000E+00	3.598240E+07
11	CO + O (+M) ⟷ CO2 (+M)	6.020000E+08	0.000000E+00	1.255200E+07
		1.800000E+07	0.000000E+00	9.978840E+06
	C2H6:3.0E+00 CH4:2.0E+00 CO:1.5E+00 CO2:3.5E+00 H2:2.0E+00 H2O:6.0E+00 O2:6.0E+00			
12	HCO + O ⟷ CO + OH	3.000000E+10	0.000000E+00	0.000000E+00
13	HCO + O ⟷ CO2 + H	3.000000E+10	0.000000E+00	0.000000E+00
14	CH2O + O ⟷ HCO + OH	3.900000E+10	0.000000E+00	1.481136E+07
15	CH2OH + O ⟷ CH2O + OH	1.000000E+10	0.000000E+00	0.000000E+00
16	CH3O + O ⟷ CH2O + OH	1.000000E+10	0.000000E+00	0.000000E+00
17	CH3OH + O ⟷ CH2OH + OH	3.880000E+02	2.500000E+00	1.297040E+07
18	CH3OH + O ⟷ CH3O + OH	1.300000E+02	2.500000E+00	2.092000E+07
19	C2H2 + O ⟷ H + HCCO	1.350000E+04	2.000000E+00	7.949600E+06
20	C2H2 + O ⟷ CH2 + CO	6.940000E+03	2.000000E+00	7.949600E+06
21	C2H3 + O ⟷ CH2CO + H	3.000000E+10	0.000000E+00	0.000000E+00
22	C2H4 + O ⟷ CH3 + HCO	1.250000E+04	1.830000E+00	9.204800E+05
23	C2H5 + O ⟷ CH2O + CH3	2.240000E+10	0.000000E+00	0.000000E+00
24	C2H6 + O ⟷ C2H5 + OH	8.980000E+04	1.920000E+00	2.380696E+07
25	HCCO + O ⟷ 2 CO + H	1.000000E+11	0.000000E+00	0.000000E+00
26	CH2CO + O ⟷ HCCO + OH	1.000000E+10	0.000000E+00	3.347200E+07
27	CH2CO + O ⟷ CH2 + CO2	1.750000E+09	0.000000E+00	5.648400E+06
28	CO + O2 ⟷ CO2 + O	2.500000E+09	0.000000E+00	1.999952E+08
29	CH2O + O2 ⟷ HCO + HO2	1.000000E+11	0.000000E+00	1.673600E+08
30	H + O2 + M ⟷ HO2 + M	2.800000E+12	-8.600000E-01	0.000000E+00
	C2H6:1.50E+00 CO:7.50E-01 CO2:1.50E+00 H2O:0.00E+00 N2:0.00E+00 O2:0.00E+00			
31	H + 2 O2 ⟷ HO2 + O2	2.080000E+13	-1.240000E+00	0.000000E+00
32	H + H2O + O2 ⟷ H2O + HO2	1.126000E+13	-7.600000E-01	0.000000E+00
33	H + N2 + O2 ⟷ HO2 + N2	2.600000E+13	-1.240000E+00	0.000000E+00
34	H + O2 ⟷ O + OH	2.650000E+13	-6.707000E-01	7.129954E+07
35	2 H + M ⟷ H2 + M	1.000000E+12	-1.000000E+00	0.000000E+00
	C2H6:3.00E+00 CH4:2.00E+00 CO2:0.00E+00 H2:0.00E+00 H2O:0.00E+00			

No.	Reaction	A	b	E_a
36	2 H + H2 ⟷ 2 H2	9.000000E+10	-6.000000E-01	0.000000E+00
37	2 H + H2O ⟷ H2 + H2O	6.000000E+13	-1.250000E+00	0.000000E+00
38	H + OH + M ⟷ H2O + M	2.200000E+16	-2.000000E+00	0.000000E+00
	C2H6:3.00E+00 CH4:2.00E+00 H2:7.30E-01 H2O:3.65E+00			
39	H + HO2 ⟷ H2O + O	3.970000E+09	0.000000E+00	2.807464E+06
40	H + HO2 ⟷ H2 + O2	4.480000E+10	0.000000E+00	4.468512E+06
41	H + HO2 ⟷ 2 OH	8.400000E+10	0.000000E+00	2.656840E+06
42	H + H2O2 ⟷ H2 + HO2	1.210000E+04	2.000000E+00	2.175680E+07
43	H + H2O2 ⟷ H2O + OH	1.000000E+10	0.000000E+00	1.506240E+07
44	CH2 + H (+M) ⟷ CH3 (+M)	1.040000E+20	-2.760000E+00	6.694400E+06
	5.62E-01 9.10E+01 5.84E+03 8.55E+03	6.000000E+11	0.000000E+00	0.000000E+00
	C2H6:3.0E+00 CH4:2.0E+00 CO:1.5E+00 CO2:2.0E+00 H2:2.0E+00 H2O:6.0E+00			
45	CH2(S) + H ⟷ CH + H2	3.000000E+10	0.000000E+00	0.000000E+00
46	CH3 + H (+M) ⟷ CH4 (+M)	2.620000E+27	-4.760000E+00	1.020896E+07
	7.83E-01 7.40E+01 2.94E+03 6.96E+03	1.390000E+13	-5.340000E-01	2.242624E+06
	C2H6:3.0E+00 CH4:3.0E+00 CO:1.5E+00 CO2:2.0E+00 H2:2.0E+00 H2O:6.0E+00			
47	CH4 + H ⟷ CH3 + H2	6.600000E+05	1.620000E+00	4.535456E+07
48	H + HCO ⟷ CO + H2	7.340000E+10	0.000000E+00	0.000000E+00
49	CH2O + H (+M) ⟷ CH2OH (+M)	1.270000E+26	-4.820000E+00	2.732152E+07
	7.19E-01 1.03E+02 1.29E+03 4.16E+03	5.400000E+08	4.540000E-01	1.506240E+07
	C2H6:3.0E+00 CH4:2.0E+00 CO:1.5E+00 CO2:2.0E+00 H2:2.0E+00 H2O:6.0E+00			
50	CH2O + H (+M) ⟷ CH3O (+M)	2.200000E+24	-4.800000E+00	2.326304E+07
	7.58E-01 9.40E+01 1.56E+03 4.20E+03	5.400000E+08	4.540000E-01	1.087840E+07
	C2H6:3.0E+00 CH4:2.0E+00 CO:1.5E+00 CO2:2.0E+00 H2:2.0E+00 H2O:6.0E+00			
51	CH2O + H ⟷ H2 + HCO	5.740000E+04	1.900000E+00	1.147253E+07
52	CH2OH + H ⟷ CH2O + H2	2.000000E+10	0.000000E+00	0.000000E+00
53	CH2OH + H ⟷ CH3 + OH	1.650000E+08	6.500000E-01	-1.188256E+06
54	CH2OH + H ⟷ CH2(S) + H2O	3.280000E+10	-9.000000E-02	2.552240E+06
55	CH3O + H (+M) ⟷ CH3OH (+M)	4.660000E+35	-7.440000E+00	5.891072E+07
	7.00E-01 1.00E+02 9.00E+04 1.00E+04	2.430000E+09	5.150000E-01	2.092000E+05
	C2H6:3.0E+00 CH4:2.0E+00 CO:1.5E+00 CO2:2.0E+00 H2:2.0E+00 H2O:6.0E+00			
56	CH3O + H ⟷ CH2OH + H	4.150000E+04	1.630000E+00	8.050016E+06
57	CH3O + H ⟷ CH2O + H2	2.000000E+10	0.000000E+00	0.000000E+00
58	CH3O + H ⟷ CH3 + OH	1.500000E+09	5.000000E-01	-4.602400E+05
59	CH3O + H ⟷ CH2(S) + H2O	2.620000E+11	-2.300000E-01	4.476880E+06
60	CH3OH + H ⟷ CH2OH + H2	1.700000E+04	2.100000E+00	2.037608E+07
61	CH3OH + H ⟷ CH3O + H2	4.200000E+03	2.100000E+00	2.037608E+07
62	C2H2 + H (+M) ⟷ C2H3 (+M)	3.800000E+34	-7.270000E+00	3.020848E+07
	7.51E-01 9.85E+01 1.30E+03 4.17E+03	5.600000E+09	0.000000E+00	1.004160E+07
	C2H6:3.0E+00 CH4:2.0E+00 CO:1.5E+00 CO2:2.0E+00 H2:2.0E+00 H2O:6.0E+00			
63	C2H3 + H ⟷ C2H2 + H2	3.000000E+10	0.000000E+00	0.000000E+00
64	C2H4 + H (+M) ⟷ C2H5 (+M)	6.000000E+35	-7.620000E+00	2.916248E+07
	9.75E-01 2.10E+02 9.84E+02 4.37E+03	5.400000E+08	4.540000E-01	7.614880E+06
	C2H6:3.0E+00 CH4:2.0E+00 CO:1.5E+00 CO2:2.0E+00 H2:2.0E+00 H2O:6.0E+00			

No.	Reaction	A	b	E_a
65	C2H4 + H ⟷ C2H3 + H2	1.325000E+03	2.530000E+00	5.121216E+07
66	C2H5 + H (+M) ⟷ C2H6 (+M)	1.990000E+35	-7.080000E+00	2.797004E+07
	8.42E-01 1.25E+02 2.22E+03 6.88E+03	5.210000E+14	-9.900000E-01	6.610720E+06
	C2H6:3.0E+00 CH4:2.0E+00 CO:1.5E+00 CO2:2.0E+00 H2:2.0E+00 H2O:6.0E+00			
67	C2H5 + H ⟷ C2H4 + H2	2.000000E+09	0.000000E+00	0.000000E+00
68	C2H6 + H ⟷ C2H5 + H2	1.150000E+05	1.900000E+00	3.150552E+07
69	H + HCCO ⟷ CH2(S) + CO	1.000000E+11	0.000000E+00	0.000000E+00
70	CH2CO + H ⟷ H2 + HCCO	5.000000E+10	0.000000E+00	3.347200E+07
71	CH2CO + H ⟷ CH3 + CO	1.130000E+10	0.000000E+00	1.434275E+07
72	CO + H2 (+M) ⟷ CH2O (+M)	5.070000E+21	-3.420000E+00	3.529204E+08
	9.32E-01 1.97E+02 1.54E+03 1.03E+04	4.300000E+04	1.500000E+00	3.330464E+08
	C2H6:3.0E+00 CH4:2.0E+00 CO:1.5E+00 CO2:2.0E+00 H2:2.0E+00 H2O:6.0E+00			
73	H2 + OH ⟷ H + H2O	2.160000E+05	1.510000E+00	1.435112E+07
74	2 OH (+M) ⟷ H2O2 (+M)	2.300000E+12	-9.000000E-01	-7.112800E+06
	7.35E-01 9.40E+01 1.76E+03 5.18E+03	7.400000E+10	-3.700000E-01	0.000000E+00
	C2H6:3.0E+00 CH4:2.0E+00 CO:1.5E+00 CO2:2.0E+00 H2:2.0E+00 H2O:6.0E+00			
75	2 OH ⟷ H2O + O	3.570000E+01	2.400000E+00	-8.828240E+06
76	HO2 + OH ⟷ H2O + O2	1.450000E+10	0.000000E+00	-2.092000E+06
77	H2O2 + OH ⟷ H2O + HO2	2.000000E+09	0.000000E+00	1.786568E+06
78	H2O2 + OH ⟷ H2O + HO2	1.700000E+15	0.000000E+00	1.230514E+08
79	CH + OH ⟷ H + HCO	3.000000E+10	0.000000E+00	0.000000E+00
80	CH2 + OH ⟷ CH2O + H	2.000000E+10	0.000000E+00	0.000000E+00
81	CH2 + OH ⟷ CH + H2O	1.130000E+04	2.000000E+00	1.255200E+07
82	CH2(S) + OH ⟷ CH2O + H	3.000000E+10	0.000000E+00	0.000000E+00
83	CH3 + OH (+M) ⟷ CH3OH (+M)	4.000000E+30	-5.920000E+00	1.313776E+07
	4.12E-01 1.95E+02 5.90E+03 6.39E+03	2.790000E+15	-1.430000E+00	5.564720E+06
	C2H6:3.0E+00 CH4:2.0E+00 CO:1.5E+00 CO2:2.0E+00 H2:2.0E+00 H2O:6.0E+00			
84	CH3 + OH ⟷ CH2 + H2O	5.600000E+04	1.600000E+00	2.267728E+07
85	CH3 + OH ⟷ CH2(S) + H2O	6.440000E+14	-1.340000E+00	5.928728E+06
86	CH4 + OH ⟷ CH3 + H2O	1.000000E+05	1.600000E+00	1.305408E+07
87	CO + OH ⟷ CO2 + H	4.760000E+04	1.228000E+00	2.928800E+05
88	HCO + OH ⟷ CO + H2O	5.000000E+10	0.000000E+00	0.000000E+00
89	CH2O + OH ⟷ H2O + HCO	3.430000E+06	1.180000E+00	-1.870248E+06
90	CH2OH + OH ⟷ CH2O + H2O	5.000000E+09	0.000000E+00	0.000000E+00
91	CH3O + OH ⟷ CH2O + H2O	5.000000E+09	0.000000E+00	0.000000E+00
92	CH3OH + OH ⟷ CH2OH + H2O	1.440000E+03	2.000000E+00	-3.514560E+06
93	CH3OH + OH ⟷ CH3O + H2O	6.300000E+03	2.000000E+00	6.276000E+06
94	C2H2 + OH ⟷ CH2CO + H	2.180000E-07	4.500000E+00	-4.184000E+06
95	C2H2 + OH ⟷ CH3 + CO	4.830000E-07	4.000000E+00	-8.368000E+06
96	C2H3 + OH ⟷ C2H2 + H2O	5.000000E+09	0.000000E+00	0.000000E+00
97	C2H4 + OH ⟷ C2H3 + H2O	3.600000E+03	2.000000E+00	1.046000E+07
98	C2H6 + OH ⟷ C2H5 + H2O	3.540000E+03	2.120000E+00	3.640080E+06
99	CH2CO + OH ⟷ H2O + HCCO	7.500000E+09	0.000000E+00	8.368000E+06
100	2 HO2 ⟷ H2O2 + O2	1.300000E+08	0.000000E+00	-6.819920E+06

No.	Reaction	A	b	E_a
101	2 HO2 ⟷ H2O2 + O2	4.200000E+11	0.000000E+00	5.020800E+07
102	CH2 + HO2 ⟷ CH2O + OH	2.000000E+10	0.000000E+00	0.000000E+00
103	CH3 + HO2 ⟷ CH4 + O2	1.000000E+09	0.000000E+00	0.000000E+00
104	CH3 + HO2 ⟷ CH3O + OH	3.780000E+10	0.000000E+00	0.000000E+00
105	CO + HO2 ⟷ CO2 + OH	1.500000E+11	0.000000E+00	9.874240E+07
106	CH2O + HO2 ⟷ H2O2 + HCO	5.600000E+03	2.000000E+00	5.020800E+07
107	CH + O2 ⟷ HCO + O	6.710000E+10	0.000000E+00	0.000000E+00
108	CH + H2 ⟷ CH2 + H	1.080000E+11	0.000000E+00	1.301224E+07
109	CH + H2O ⟷ CH2O + H	5.710000E+09	0.000000E+00	-3.158920E+06
110	CH + CH3 ⟷ C2H3 + H	3.000000E+10	0.000000E+00	0.000000E+00
111	CH + CH4 ⟷ C2H4 + H	6.000000E+10	0.000000E+00	0.000000E+00
112	CH + CO (+M) ⟷ HCCO (+M)	2.690000E+22	-3.740000E+00	8.100224E+06
	5.76E-01 2.37E+02 1.65E+03 5.07E+03	5.000000E+10	0.000000E+00	0.000000E+00
	C2H6:3.0E+00 CH4:2.0E+00 CO:1.5E+00 CO2:2.0E+00 H2:2.0E+00 H2O:6.0E+00			
113	CH + CO2 ⟷ CO + HCO	1.900000E+11	0.000000E+00	6.607373E+07
114	CH + CH2O ⟷ CH2CO + H	9.460000E+10	0.000000E+00	-2.154760E+06
115	CH2 + O2 ⟶ CO + H + OH	5.000000E+09	0.000000E+00	6.276000E+06
116	CH2 + H2 ⟷ CH3 + H	5.000000E+02	2.000000E+00	3.025032E+07
117	CH2 + CH3 ⟷ C2H4 + H	4.000000E+10	0.000000E+00	0.000000E+00
118	CH2 + CH4 ⟷ 2 CH3	2.460000E+03	2.000000E+00	3.460168E+07
119	CH2 + CO (+M) ⟷ CH2CO (+M)	2.690000E+27	-5.110000E+00	2.968548E+07
	5.91E-01 2.75E+02 1.23E+03 5.18E+03	8.100000E+08	5.000000E-01	1.886984E+07
	C2H6:3.0E+00 CH4:2.0E+00 CO:1.5E+00 CO2:2.0E+00 H2:2.0E+00 H2O:6.0E+00			
120	CH2(S) + N2 ⟷ CH2 + N2	1.500000E+10	0.000000E+00	2.510400E+06
121	CH2(S) + O2 ⟷ CO + H + OH	2.800000E+10	0.000000E+00	0.000000E+00
122	CH2(S) + O2 ⟷ CO + H2O	1.200000E+10	0.000000E+00	0.000000E+00
123	CH2(S) + H2 ⟷ CH3 + H	7.000000E+10	0.000000E+00	0.000000E+00
124	CH2(S) + H2O (+M) ⟷ CH3OH (+M)	1.880000E+32	-6.360000E+00	2.108736E+07
	6.03E-01 2.08E+02 3.92E+03 1.02E+04	4.820000E+14	-1.160000E+00	4.790680E+06
	C2H6:3.0E+00 CH4:2.0E+00 CO:1.5E+00 CO2:2.0E+00 H2:2.0E+00 H2O:6.0E+00			
125	CH2(S) + H2O ⟷ CH2 + H2O	3.000000E+10	0.000000E+00	0.000000E+00
126	CH2(S) + CH3 ⟷ C2H4 + H	1.200000E+10	0.000000E+00	-2.384880E+06
127	CH2(S) + CH4 ⟷ 2 CH3	1.600000E+10	0.000000E+00	-2.384880E+06
128	CH2(S) + CO ⟷ CH2 + CO	9.000000E+09	0.000000E+00	0.000000E+00
129	CH2(S) + CO2 ⟷ CH2 + CO2	7.000000E+09	0.000000E+00	0.000000E+00
130	CH2(S) + CO2 ⟷ CH2O + CO	1.400000E+10	0.000000E+00	0.000000E+00
131	C2H6 + CH2(S) ⟷ C2H5 + CH3	4.000000E+10	0.000000E+00	-2.301200E+06
132	CH3 + O2 ⟷ CH3O + O	3.560000E+10	0.000000E+00	1.275283E+08
133	CH3 + O2 ⟷ CH2O + OH	2.310000E+09	0.000000E+00	8.499796E+07
134	CH3 + H2O2 ⟷ CH4 + HO2	2.450000E+01	2.470000E+00	2.167312E+07
135	2 CH3 (+M) ⟷ C2H6 (+M)	3.400000E+35	-7.030000E+00	1.155621E+07
	6.19E-01 7.32E+01 1.18E+03 1.00E+04	6.770000E+13	-1.180000E+00	2.736336E+06
	C2H6:3.0E+00 CH4:2.0E+00 CO:1.5E+00 CO2:2.0E+00 H2:2.0E+00 H2O:6.0E+00			
136	2 CH3 ⟷ C2H5 + H	6.840000E+09	1.000000E-01	4.435040E+07

No.	Reaction	A	b	E_a
137	CH3 + HCO ⟷ CH4 + CO	2.648000E+10	0.000000E+00	0.000000E+00
138	CH2O + CH3 ⟷ CH4 + HCO	3.320000E+00	2.810000E+00	2.451824E+07
139	CH3 + CH3OH ⟷ CH2OH + CH4	3.000000E+04	1.500000E+00	4.158896E+07
140	CH3 + CH3OH ⟷ CH3O + CH4	1.000000E+04	1.500000E+00	4.158896E+07
141	C2H4 + CH3 ⟷ C2H3 + CH4	2.270000E+02	2.000000E+00	3.849280E+07
142	C2H6 + CH3 ⟷ C2H5 + CH4	6.140000E+03	1.740000E+00	4.372280E+07
143	H2O + HCO ⟷ CO + H + H2O	1.500000E+15	-1.000000E+00	7.112800E+07
144	HCO + M ⟷ CO + H + M	1.870000E+14	-1.000000E+00	7.112800E+07
	C2H6:3.00E+00 CH4:2.00E+00 CO:1.50E+00 CO2:2.00E+00 H2:2.00E+00 H2O:0.00E+00			
145	HCO + O2 ⟷ CO + HO2	1.345000E+10	0.000000E+00	1.673600E+06
146	CH2OH + O2 ⟷ CH2O + HO2	1.800000E+10	0.000000E+00	3.765600E+06
147	CH3O + O2 ⟷ CH2O + HO2	4.280000E-16	7.600000E+00	-1.476952E+07
148	C2H3 + O2 ⟷ CH2O + HCO	4.580000E+13	-1.390000E+00	4.246760E+06
149	C2H4 (+M) ⟷ C2H2 + H2 (+M)	1.580000E+48	-9.300000E+00	4.091952E+08
	7.35E-01 1.80E+02 1.04E+03 5.42E+03	8.000000E+12	4.400000E-01	3.630457E+08
	C2H6:3.0E+00 CH4:2.0E+00 CO:1.5E+00 CO2:2.0E+00 H2:2.0E+00 H2O:6.0E+00			
150	C2H5 + O2 ⟷ C2H4 + HO2	8.400000E+08	0.000000E+00	1.621300E+07
151	HCCO + O2 ⟷ 2 CO + OH	3.200000E+09	0.000000E+00	3.573136E+06
152	CH3 + O ⟶ CO + H + H2	3.370000E+10	0.000000E+00	0.000000E+00
153	C2H4 + O ⟷ CH2CHO + H	6.700000E+03	1.830000E+00	9.204800E+05
154	HO2 + OH ⟷ H2O + O2	5.000000E+12	0.000000E+00	7.250872E+07
155	CH3 + OH ⟶ CH2O + H2	8.000000E+06	5.000000E-01	-7.342920E+06
156	CH2 + O2 ⟶ CO2 + 2 H	5.800000E+09	0.000000E+00	6.276000E+06
157	CH2 + O2 ⟷ CH2O + O	2.400000E+09	0.000000E+00	6.276000E+06
158	CH2(S) + H2O ⟶ CH2O + H2	6.820000E+07	2.500000E-01	-3.912040E+06
159	C2H3 + O2 ⟷ CH2CHO + O	3.030000E+08	2.900000E-01	4.602400E+04
160	C2H3 + O2 ⟷ C2H2 + HO2	1.337000E+03	1.610000E+00	-1.606656E+06
161	CH2CO + H (+M) ⟷ CH2CHO (+M)	1.012000E+36	-7.630000E+00	1.612514E+07
	4.65E-01 2.01E+02 1.77E+03 5.33E+03	4.865000E+08	4.220000E-01	-7.342920E+06
	C2H6:3.0E+00 CH4:2.0E+00 CO:1.5E+00 CO2:2.0E+00 H2:2.0E+00 H2O:6.0E+00			
162	CH2CHO + O ⟶ CH2 + CO2 + H	1.500000E+11	0.000000E+00	0.000000E+00
163	CH2CHO + O2 ⟶ CH2O + CO + OH	1.810000E+07	0.000000E+00	0.000000E+00
164	CH2CHO + O2 ⟶ 2 HCO + OH	2.350000E+07	0.000000E+00	0.000000E+00
165	CH2CHO + H ⟷ CH3 + HCO	2.200000E+10	0.000000E+00	0.000000E+00
166	CH2CHO + H ⟷ CH2CO + H2	1.100000E+10	0.000000E+00	0.000000E+00

B Transport properties modeling[2]

The simplified transport properties adopted in AVBP consist in a constant mixture Prandtl (Pr) number and a constant Schmidt (Sc) number for each species of the chemical scheme. These values are optimized during the mechanism reduction procedure (see Section A). Pr and Sc numbers for the ARC scheme used in the simulations are summarized in Tab. B.1.

Property	Specie	Value
Schmidt	H_2	0.207
	H	0.124
	O	0.484
	O_2	0.751
	OH	0.493
	H_2O	0.558
	HO_2	0.749
	H_2O_2	0.754
	CH_3	0.679
	CH_4	0.682
	CO	0.755
	CO_2	0.944
	CH_2O	0.868
	CH_3O	0.888
	CH_3OH	0.889
	C_2H_2	0.891
	C_2H_4	0.900
	C_2H_6	0.988
	CH_2CO	1.025
	N_2	0.717
Prandlt	Mixture	0.712

Table B.1: Constant Schmidt number for each species and mixture Prandtl number used in the simplified transport model.

To take into account the variability of the transport properties when considering a mixture with light molar weight components (e.g. H_2), a database of Prandtl number and N_2 Schmidt number as a function of the ternary mixture composition (i.e. CH_4, H_2, N_2) has been computed with the complex-transport module of Cantera. The results of this analysis are showed in Fig. 2(d) of the manuscript. It is then straightforward to compute a polynomial fitting of these two variables and express them as a function of the molar fraction X of CH_4 and H_2 (i.e. the two independent variables), as follows:

[2]For additional information on the transport properties modeling please contact P. W. Agostinelli. *E-mail*: agostinelli@cerfacs.fr (personal e-mail: walter-agostinelli@tiscali.it)

$$\text{Prandtl} = f(\mathrm{X_{CH_4}}, \mathrm{X_{H_2}}) = f_{00} + f_{10}\mathrm{X_{CH_4}} + f_{01}\mathrm{X_{H_2}} + ... + f_{ab}{\mathrm{X_{CH_4}}}^{a}{\mathrm{X_{H_2}}}^{b} \quad (1)$$

$$\mathrm{N_2\ Schmidt} = g(\mathrm{X_{CH_4}}, \mathrm{X_{H_2}}) = g_{00} + g_{10}\mathrm{X_{CH_4}} + g_{01}\mathrm{X_{H_2}} + ... + g_{cd}{\mathrm{X_{CH_4}}}^{c}{\mathrm{X_{H_2}}}^{d} \quad (2)$$

In particular, the function f in Eq. (1) and the function g in Eq. (2) have been chosen to be a piecewise-defined polynomials with respect to $\mathrm{X_{CH_4}}$ and $\mathrm{X_{H_2}}$ with the coefficients being determined in order to reduce the root mean square errors with respect to the database from Cantera. The resulting polynomial fitting is able to correctly provide the exact Prandtl number and $\mathrm{N_2}$ Schmidt number, with a coefficient of determination $\mathrm{R^2}$ of 0.9996, while not worsening the code performances. Table B.2 and Tab. B.3 report the coefficients used in the piecewise-defined fitting functions and their intervals of validity for the Pr number and the $\mathrm{N_2}$ Sc number expressions, respectively.

Coefficient (f_{ab})	Value	Interval
$\mathrm{f_{00}}$	0.7076	
$\mathrm{f_{10}}$	0.02157	
$\mathrm{f_{01}}$	-1.608	
$\mathrm{f_{20}}$	-0.013	
$\mathrm{f_{11}}$	0.3687	
$\mathrm{f_{02}}$	3.643	
$\mathrm{f_{30}}$	-0.003734	
$\mathrm{f_{21}}$	0.04177	
$\mathrm{f_{12}}$	-0.5402	
$\mathrm{f_{03}}$	-3.758	
$\mathrm{f_{40}}$	-0.0001393	for $\mathrm{X_{CH_4}}$ in [0,1] and $\mathrm{X_{H_2}}$ in [0,1-$\mathrm{X_{CH_4}}$]
$\mathrm{f_{31}}$	-0.03073	
$\mathrm{f_{22}}$	-0.3168	
$\mathrm{f_{13}}$	-0.2286	
$\mathrm{f_{04}}$	1.302	
$\mathrm{f_{50}}$	-0.001286	
$\mathrm{f_{41}}$	0.001784	
$\mathrm{f_{32}}$	0.09282	
$\mathrm{f_{23}}$	0.4952	
$\mathrm{f_{14}}$	0.7872	
$\mathrm{f_{05}}$	0.3903	

Table B.2: Coefficient of the piecewise-defined polynomial in Eq. (1) expressing the mixture Prandtl number as a function of $\mathrm{X_{CH_4}}$ and $\mathrm{X_{H_2}}$.

Coefficient (g_{cd})	Value	Interval
g_{00}	1.295	
g_{10}	-1.574	
g_{01}	4.886	
g_{20}	7.229	
g_{11}	-35.28	
g_{02}	-7.989	
g_{30}	-18.36	
g_{21}	94.96	
g_{12}	56.5	
g_{03}	5.543	
g_{40}	20.05	for X_{CH_4} in [0.05,1] and X_{H_2} in [0.05,1-X_{CH_4}]
g_{31}	-109.1	
g_{22}	-115.2	
g_{13}	-39.54	
g_{04}	-0.1024	
g_{50}	-7.872	
g_{41}	44.01	
g_{32}	71.57	
g_{23}	39.7	
g_{14}	10.76	
g_{05}	-2.227	
g_{00}	2.833	
g_{10}	-32.13	
g_{01}	-0.04449	
g_{11}	69.78	
g_{02}	0.3827	
g_{12}	-108.3	for X_{CH_4} in [0,0.05] and X_{H_2} in [0.05,1-X_{CH_4}]
g_{03}	-3.604	
g_{13}	90.81	
g_{04}	6.146	
g_{14}	-28.94	
g_{05}	-4.295	
g_{00}	0.717	
g_{10}	10.05	for X_{CH_4} in [0,0.05] and X_{H_2} in [0,0.05-X_{CH_4}]
g_{01}	42.32	
g_{d00}	2.66	
g_{d10}	-28.82	for X_{CH_4} in [0,0.05] and X_{H_2} in [0.05-X_{CH_4},0.05]
g_{d01}	3.457	

Table B.3: Coefficient of the piecewise-defined polynomial in Eq. (2) expressing the N_2 Schmidt number as a function of X_{CH_4} and X_{H_2}.

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