CHEMICAL KINETIC MECHANISM FOR COMBUSTION IN SUPERCRITICAL CARBON DIOXIDE

James M Harman-Thomas.

Professor Mohamed Pourkashanian

Dr Kevin J. Hughes

Energy 2050, Department of Mechanical Engineering,

The University of Sheffield

Presentation Overview

Presentation Layout:

Presentation Overview

Presentation Layout:

- Introduction the Allam cycle and high-pressure oxyfuel combustion.

Presentation Overview

Presentation Layout:

- Introduction the Allam cycle and high-pressure oxyfuel combustion.
- Description of the modelling procedure used in the present study.

Presentation Overview

Presentation Layout:

- Introduction the Allam cycle and high-pressure oxyfuel combustion.

- Description of the modelling procedure used in the present study.

- Discussion of the modelling work and identification of important reactions.

Presentation Overview

Presentation Layout:

- Introduction the Allam cycle and high-pressure oxyfuel combustion.
- Description of the modelling procedure used in the present study.
- Discussion of the modelling work and identification of important reactions.
- Description of the final mechanism created and its improved performance.

Motivation (1)

Commercial power generation using high-pressure, oxyfuel combustion is a potential way of producing energy from fossil fuels with no greenhouse gas emissions.

T. Lockwood, A comparative review of next-generation carbon capture technologies for coal-fired power plant, 13th International Conference on Greenhouse Gas Control Technologies, GHGT-13 114 (2017) 2658-2670.

Motivation (1)

Commercial power generation using high-pressure, oxyfuel combustion is a potential way of producing energy from fossil fuels with no greenhouse gas emissions.

In a comparative review of emerging carbon capture and storage (CCS) technologies, only the Allam-Fetvedt cycle was identified as only coal combustion technology which could reduce the cost of electricity (Lockwood (2017)).

T. Lockwood, A comparative review of next-generation carbon capture technologies for coal-fired power plant, 13th International Conference on Greenhouse Gas Control Technologies, GHGT-13 114 (2017) 2658-2670.

Motivation (1)

Commercial power generation using high-pressure, oxyfuel combustion is a potential way of producing energy from fossil fuels with no greenhouse gas emissions.

In a comparative review of emerging carbon capture and storage (CCS) technologies, only the Allam-Fetvedt cycle was identified as only coal combustion technology which could reduce the cost of electricity (Lockwood (2017)).

The high-pressures of the Allam-Fetvedt cycle and greater power density of sCO_2 reduces the total footprint compared traditional power plants.

T. Lockwood, A comparative review of next-generation carbon capture technologies for coal-fired power plant, 13th International Conference on Greenhouse Gas Control Technologies, GHGT-13 114 (2017) 2658-2670.

Motivation (2)

One of the limitations for the development of Allam cycle combustors is the lack of experimental data at high-pressures and high-dilutions of CO_2.

Motivation (2)

One of the limitations for the development of Allam cycle combustors is the lack of experimental data at high-pressures and high-dilutions of CO_2.

Existing chemical kinetic mechanisms are validated using experimental data such as laminar flame speed and ignition delay time.

Motivation (2)

One of the limitations for the development of Allam cycle combustors is the lack of experimental data at high-pressures and high-dilutions of CO_2.

Existing chemical kinetic mechanisms are validated using experimental data such as laminar flame speed and ignition delay time.

Over the last 5 years, more ignition delay time data has been published allowing the validation of chemical kinetic mechanisms at these conditions.

Motivation (2)

One of the limitations for the development of Allam cycle combustors is the lack of experimental data at high-pressures and high-dilutions of CO_2.

Existing chemical kinetic mechanisms are validated using experimental data such as laminar flame speed and ignition delay time.

Over the last 5 years, more ignition delay time data has been published allowing the validation of chemical kinetic mechanisms at these conditions.

In this study, we model ignition delay time data using four existing mechanisms and use the results to produce a new mechanism designed for high-pressure, sCO_2 combustion.

Modelling Procedure

The ignition delay time is an important combustion parameter which can be experimentally determined using shock tubes or rapid compression machines.

Y. Liu, J. Cheng, C. Zou, L.X. Lu, H.X. Jing, Ignition delay times of ethane under O_2/CO_2 atmosphere at different pressures by shock tube and simulation methods, Combustion and Flame 204 (2019) 380-390.

Modelling Procedure

The ignition delay time is an important combustion parameter which can be experimentally determined using shock tubes or rapid compression machines.

As many ignition delay time datasets as available were collated for oxyfuel combustion at pressures over 10 atm for methane, hydrogen and syngas.

Y. Liu, J. Cheng, C. Zou, L.X. Lu, H.X. Jing, Ignition delay times of ethane under O_2/CO_2 atmosphere at different pressures by shock tube and simulation methods, Combustion and Flame 204 (2019) 380-390.

Modelling Procedure

The ignition delay time is an important combustion parameter which can be experimentally determined using shock tubes or rapid compression machines.

As many ignition delay time datasets as available were collated for oxyfuel combustion at pressures over 10 atm for methane, hydrogen and syngas.

These ignition delay times were modelled on Chemkin Pro using four chemical kinetic mechanisms which were compared using quantitative analysis using the following equation from Liu et al. (2019).

$$E = \frac{1}{N}\sum_{i=1}^{N}\left|\frac{X_{sim,i} - X_{exp,i}}{X_{exp,i}}\right| \times 100$$

Y. Liu, J. Cheng, C. Zou, L.X. Lu, H.X. Jing, Ignition delay times of ethane under O_2/CO_2 atmosphere at different pressures by shock tube and simulation methods, Combustion and Flame 204 (2019) 380-390.

Modelling Procedure

A series of sensitivity analyses were performed at temperatures where the agreement between different mechanisms was poor, of the agreement between a mechanism and the experimental data.

Modelling Procedure

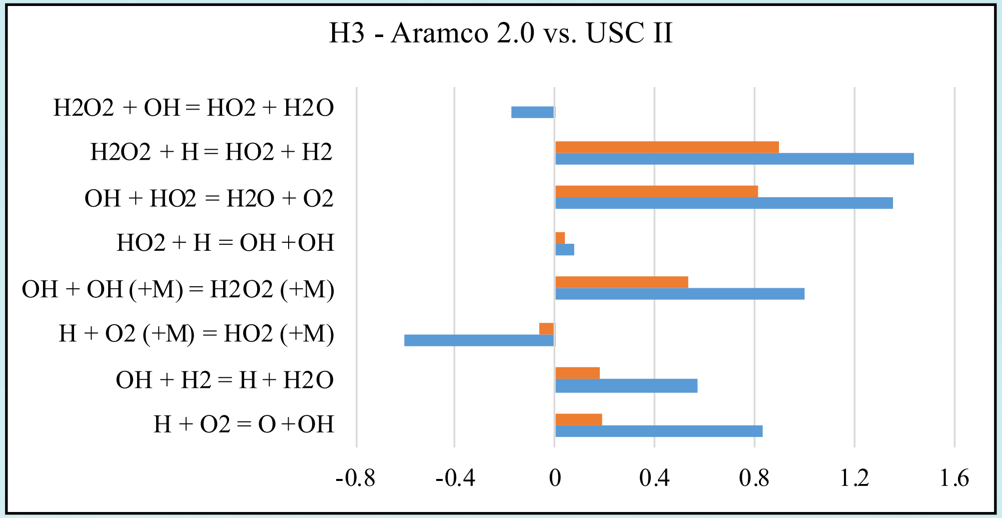
A series of sensitivity analyses were performed at temperatures where the agreement between different mechanisms was poor, of the agreement between a mechanism and the experimental data.

The most important reactions were identified and used to compare the relative importance of different reactions under different conditions.

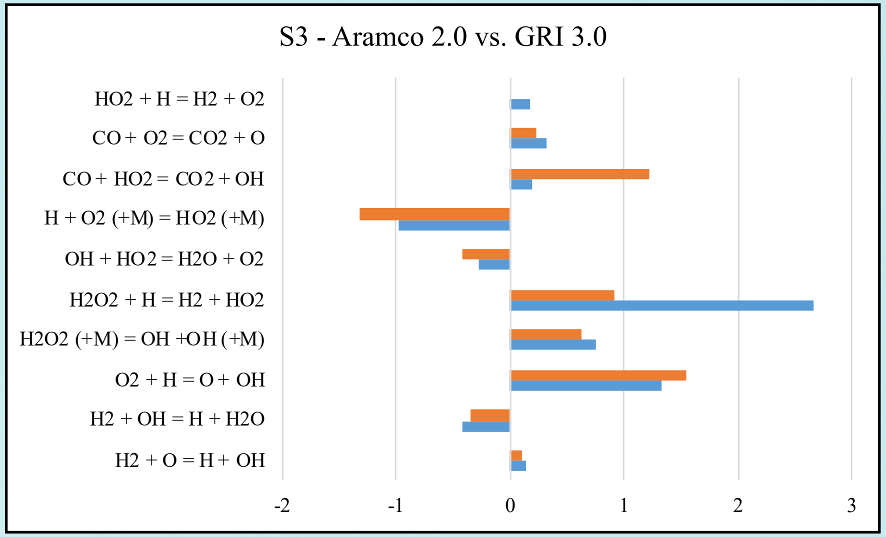
Modelling Procedure

A series of sensitivity analyses were performed at temperatures where the agreement between different mechanisms was poor, of the agreement between a mechanism and the experimental data.

The most important reactions were identified and used to compare the relative importance of different reactions under different conditions.



Comparison of the sensitivity analysis of H3 for Aramco 2.0 and USC II at 1274 K.



S3 dataset sensitivity analysis of GRI 3.0 and Aramco 2.0 at 1280 K.

Methane Datasets

Methane datasets studied.

Dataset	Reference	Average Pressure /atm	Equivalence Ratio (Φ)	CO_2 Dilution (%)
M1	Pryor et al. (2017)	29.6	1.00	85.00
M2	Barak et al. (2020)	79.9	1.00	36.50
M3	Karimi et al. (2019)	99.0	1.00	85.00
M4	Karimi et al. (2019)	97.0	0.50	80.00
M5	Karimi et al. (2019)	201.8	1.00	85.00
M6	Shao et al. (2019)	32.2	1.00	77.50
M7	Shao et al. (2019)	106.3	1.00	77.50
M8	Shao et al. (2019)	260.0	1.00	77.50
M9	Shao et al. (2019)	31.4	1.27	86.17
M10	Shao et al. (2019)	74.7	1.27	86.17
M11	Shao et al. (2019)	266.3	1.27	86.17

O. Pryor, B. Koroglu, S. Barak, J. Lopez, E. Ninnemann, L. Nash, S. Vasu, Ignition delay times of high pressure oxy-methane combustion with high levels of CO_2 dilution, Proceedings of ASME Turbo Expo 2017: Turbo-machinery Technical Conference and Exposition, Charlotte, NC, USA, 2017.

S. Barak, O. Pryor, E. Ninnemann, S. Neupane, S. Vasu, X. Lu, B. Forrest, Ignition delay times of oxy-syngas and oxy-methane in supercritical CO_2 mixtures for direct-fired cycles, Journal of Engineering for Gas Turbines and Power 142 (2020).

M. Karimi, B. Ochs, Z.F. Liu, D. Ranjan, W.T. Sun, Measurement of methane autoignition delays in carbon dioxide and argon diluents at high pressure conditions, Combustion and Flame 204 (2019) 304-319.

J.K. Shao, R. Choudhary, D.E. Davidson, R.K. Hanson, S. Barak, S. Vasu, Ignition delay times of methane and hydrogen highly diluted in carbon dioxide at high pressures up to 300 atm, Proceedings of the Combustion Institute 37 (2019) 4555-4562.

Methane Datasets

Quantitative analysis of methane datasets.

	Aramco 2.0	DTU	GRI 3.0	USC II
M1	41.47	37.98	76.11	17.04
M2	17.10	12.60	54.37	9.55
M3	24.85	27.18	54.76	20.49
M4	14.26	13.44	56.66	8.24
M5	9.53	7.35	32.49	45.53
M6	60.09	47.06	23.63	38.39
M7	15.02	16.64	43.26	20.65
M8	26.87	37.65	204.32	323.56
M9	94.06	99.87	31.97	56.38
M10	18.91	25.79	57.11	13.79
M11	3.99	24.03	32.53	125.37
Average	***29.65***	***31.78***	***60.66***	***61.73***
No. Best Fit	***3***	***1***	***2***	***5***

Methane Datasets

Quantitative analysis of methane datasets.

	Aramco 2.0	DTU	GRI 3.0	USC II
M1	41.47	37.98	76.11	17.04
M2	17.10	12.60	54.37	9.55
M3	24.85	27.18	54.76	20.49
M4	14.26	13.44	56.66	8.24
M5	9.53	7.35	32.49	45.53
M6	60.09	47.06	23.63	38.39
M7	15.02	16.64	43.26	20.65
M8	26.87	37.65	204.32	323.56
M9	94.06	99.87	31.97	56.38
M10	18.91	25.79	57.11	13.79
M11	3.99	24.03	32.53	125.37
Average	***29.65***	***31.78***	***60.66***	***61.73***
No. Best Fit	***3***	***1***	***2***	***5***

Methane Datasets

Quantitative analysis of methane datasets.

	Aramco 2.0	DTU	GRI 3.0	USC II
M1	41.47	37.98	76.11	17.04
M2	17.10	12.60	54.37	9.55
M3	24.85	27.18	54.76	20.49
M4	14.26	13.44	56.66	8.24
M5	9.53	7.35	32.49	45.53
M6	60.09	47.06	23.63	38.39
M7	15.02	16.64	43.26	20.65
M8	26.87	37.65	204.32	323.56
M9	94.06	99.87	31.97	56.38
M10	18.91	25.79	57.11	13.79
M11	3.99	24.03	32.53	125.37
Average	***29.65***	***31.78***	***60.66***	***61.73***
No. Best Fit	***3***	***1***	***2***	***5***

Methane Datasets

Quantitative analysis of methane datasets.

	Aramco 2.0	DTU	GRI 3.0	USC II
M1	41.47	37.98	76.11	17.04
M2	17.10	12.60	54.37	9.55
M3	24.85	27.18	54.76	20.49
M4	14.26	13.44	56.66	8.24
M5	9.53	7.35	32.49	45.53
M6	60.09	47.06	23.63	38.39
M7	15.02	16.64	43.26	20.65
M8	26.87	37.65	204.32	323.56
M9	94.06	99.87	31.97	56.38
M10	18.91	25.79	57.11	13.79
M11	3.99	24.03	32.53	125.37
Average	***29.65***	***31.78***	***60.66***	***61.73***
No. Best Fit	***3***	***1***	***2***	***5***

Methane Datasets

Methane datasets studied.

Dataset	Reference	Average Pressure /atm	Equivalence Ratio (Φ)	CO_2 Dilution (%)
M1	Pryor et al. (2017)	29.6	1.00	85.00
M2	Barak et al. (2020)	79.9	1.00	36.50
M3	Karimi et al. (2019)	99.0	1.00	85.00
M4	Karimi et al. (2019)	97.0	0.50	80.00
M5	Karimi et al. (2019)	201.8	1.00	85.00
M6	Shao et al. (2019)	32.2	1.00	77.50
M7	Shao et al. (2019)	106.3	1.00	77.50
M8	Shao et al. (2019)	260.0	1.00	77.50
M9	Shao et al. (2019)	31.4	1.27	86.17
M10	Shao et al. (2019)	74.7	1.27	86.17
M11	Shao et al. (2019)	266.3	1.27	86.17

O. Pryor, B. Koroglu, S. Barak, J. Lopez, E. Ninnemann, L. Nash, S. Vasu, Ignition delay times of high pressure oxy-methane combustion with high levels of CO_2 dilution, Proceedings of ASME Turbo Expo 2017: Turbo-machinery Technical Conference and Exposition, Charlotte, NC, USA, 2017.

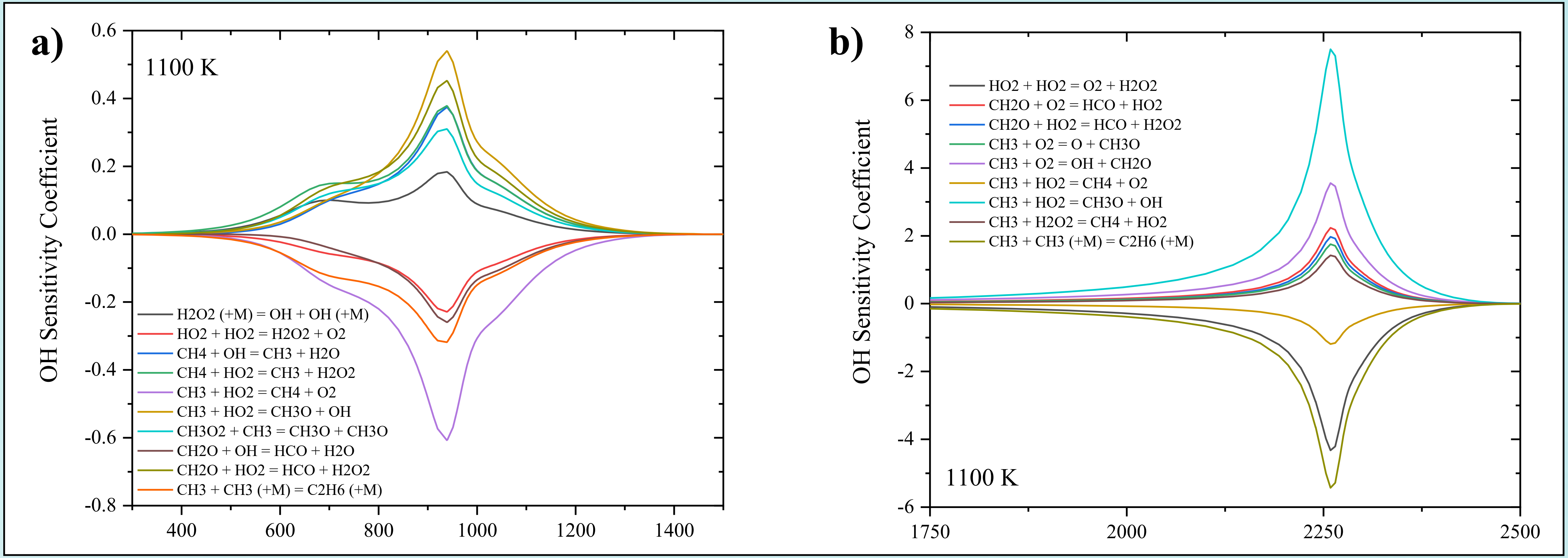
S. Barak, O. Pryor, E. Ninnemann, S. Neupane, S. Vasu, X. Lu, B. Forrest, Ignition delay times of oxy-syngas and oxy-methane in supercritical CO_2 mixtures for direct-fired cycles, Journal of Engineering for Gas Turbines and Power 142 (2020).

M. Karimi, B. Ochs, Z.F. Liu, D. Ranjan, W.T. Sun, Measurement of methane autoignition delays in carbon dioxide and argon diluents at high pressure conditions, Combustion and Flame 204 (2019) 304-319.

J.K. Shao, R. Choudhary, D.E. Davidson, R.K. Hanson, S. Barak, S. Vasu, Ignition delay times of methane and hydrogen highly diluted in carbon dioxide at high pressures up to 300 atm, Proceedings of the Combustion Institute 37 (2019) 4555-4562.

Methane Datasets

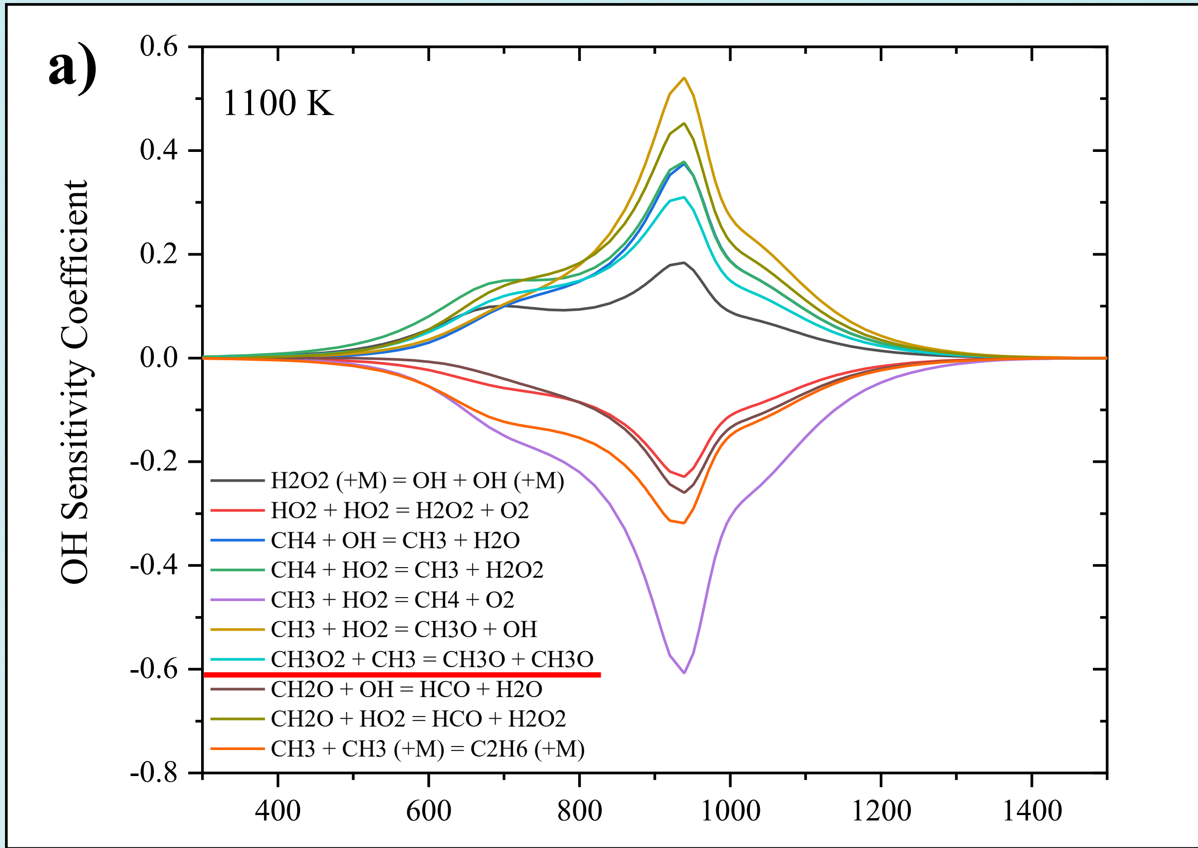
Why is there a large discrepancy between the performance of Aramco 2.0 and DTU, and GRI and USC II at high-pressures?

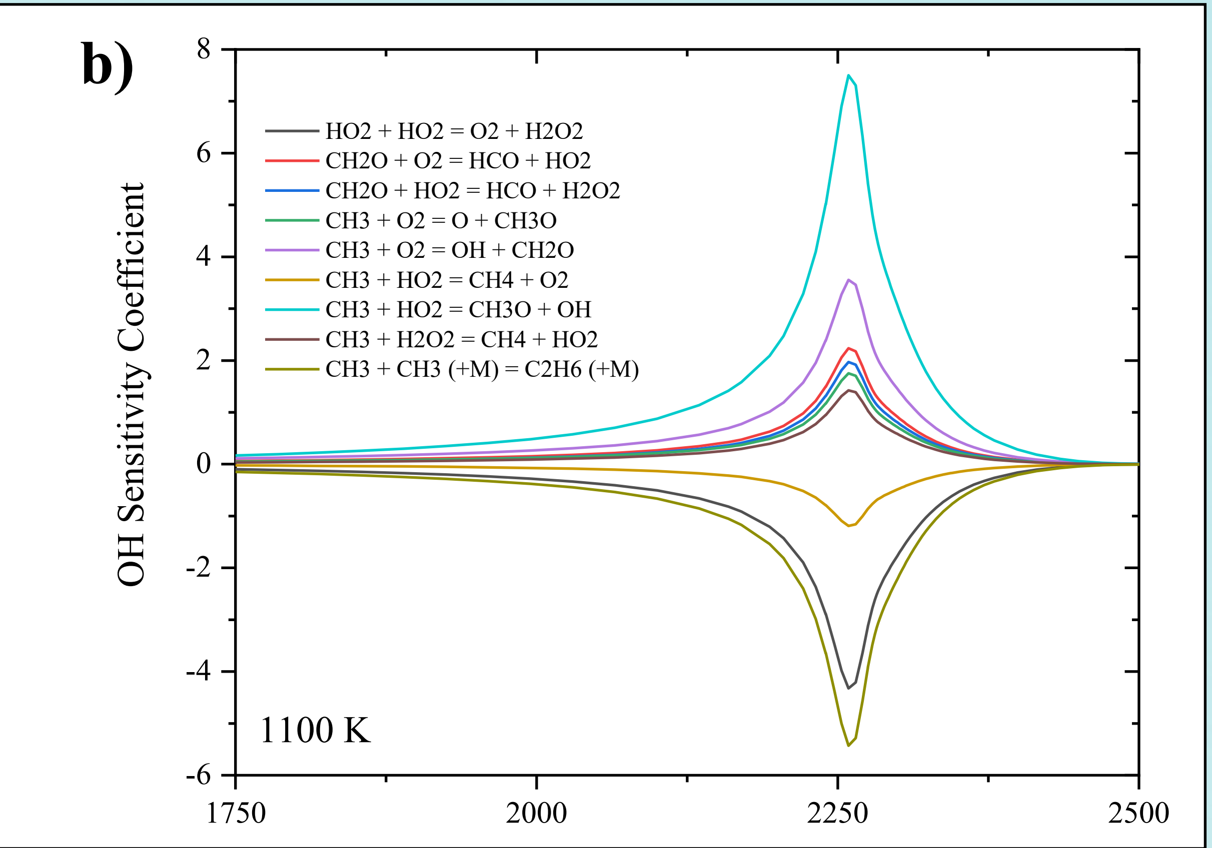


OH sensitivity coefficient against time for condition M11; a) Aramco 2.0 and b) USC II.

Methane Datasets

Why is there a large discrepancy between the performance of Aramco 2.0 and DTU, and GRI and USC II at high-pressures?





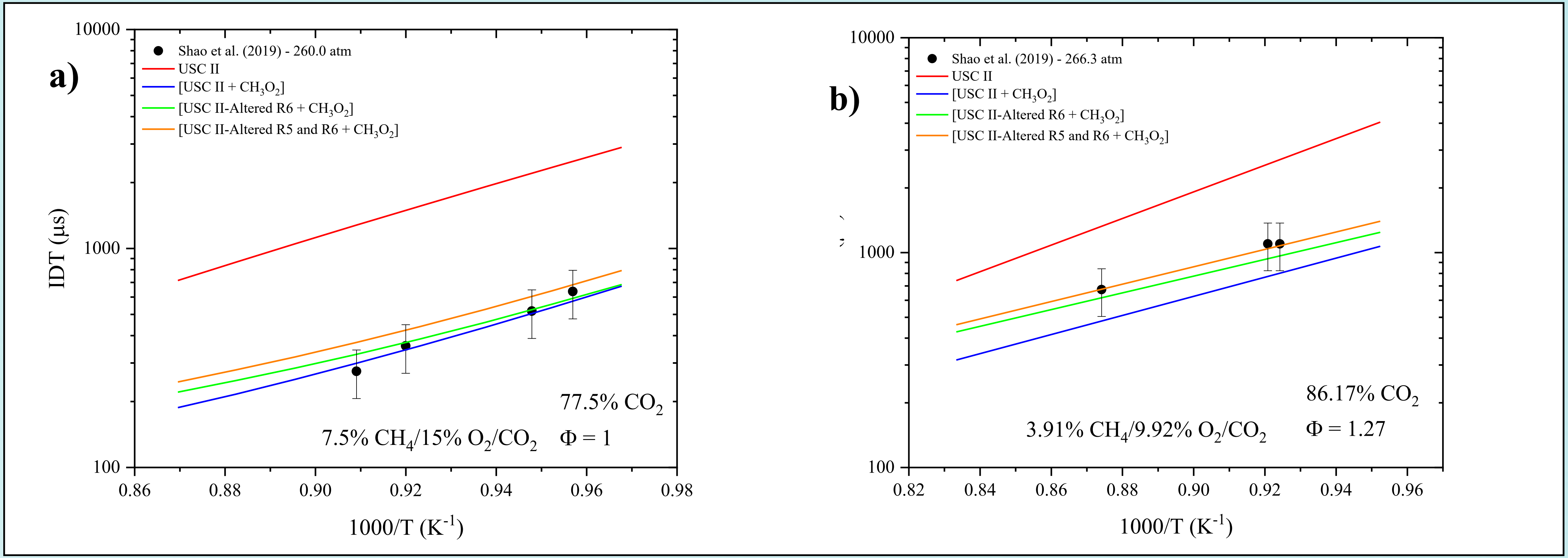
OH sensitivity coefficient against time for condition M11; a) Aramco 2.0 and b) USC II.

Methane Datasets

Adding the CH_3O_2 Chemistry from Aramco 2.0 into the USC II mechanism significantly reduces the model's ignition delay times at these temperatures.

R5 - H_2O_2 (+M) = OH +OH (+M)

R6 - $CH_3 + HO_2 = CH_4 + O_2$



Sequential changes to USC II; a) M8 and b) M11; [USC II + CH_3O_2]: addition of CH_3O_2 chemistry from Aramco 2.0, [USC II-Altered R6 + CH_3O_2] change R6 to Aramco 2.0 rate coefficient, [USC II-Altered R5 and R6 + CH_3O_2]: change R5 to the Aramco 2.0 rate coefficient.

Hydrogen Datasets

Hydrogen datasets studied.

Dataset	Reference	Average Pressure /atm	Equivalence Ratio (Φ)	CO_2 Dilution (%)
H1	Shao et al. (2019)	109.6	1.00	85.00
H2	Shao et al. (2019)	270.6	1.00	85.00
H3	Shao et al. (2019)	38.4	0.25	85.00

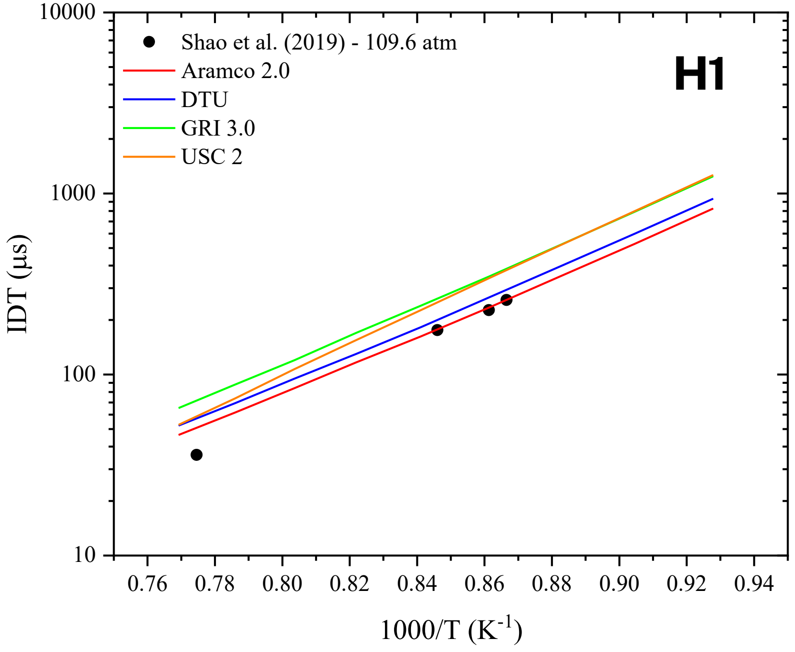
J.K. Shao, R. Choudhary, D.E. Davidson, R.K. Hanson, S. Barak, S. Vasu, Ignition delay times of methane and hydrogen highly diluted in carbon dioxide at high pressures up to 300 atm, Proceedings of the Combustion Institute 37 (2019) 4555-4562.

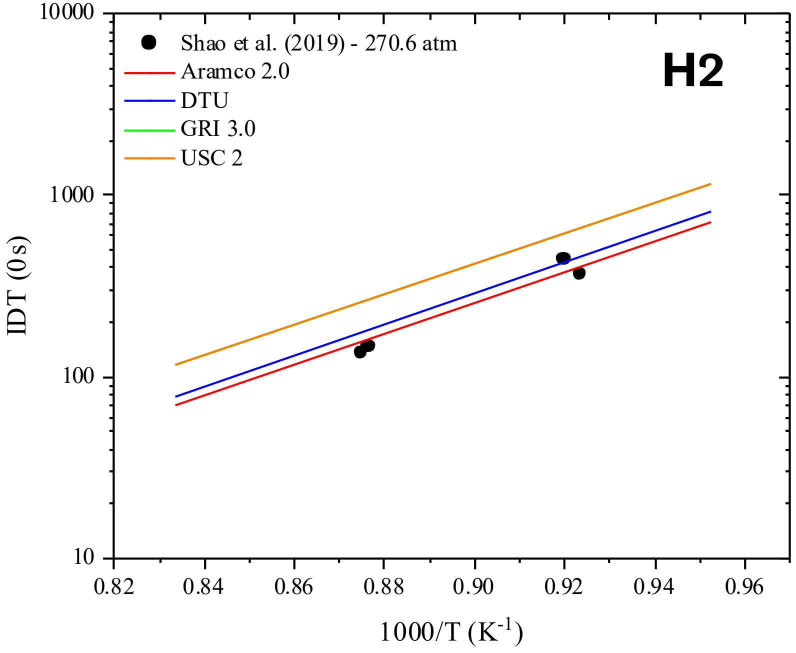
Hydrogen Datasets

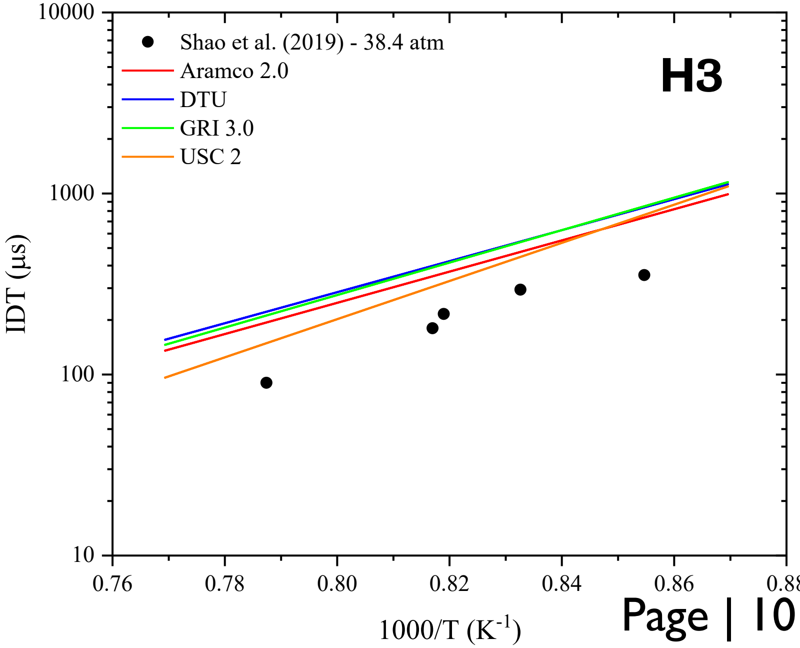
Hydrogen datasets studied.

Dataset	Reference	Average Pressure /atm	Equivalence Ratio (Φ)	CO_2 Dilution (%)
H1	Shao et al. (2019)	109.6	1.00	85.00
H2	Shao et al. (2019)	270.6	1.00	85.00
H3	Shao et al. (2019)	38.4	0.25	85.00

J.K. Shao, R. Choudhary, D.E. Davidson, R.K. Hanson, S. Barak, S. Vasu, Ignition delay times of methane and hydrogen highly diluted in carbon dioxide at high pressures up to 300 atm, Proceedings of the Combustion Institute 37 (2019) 4555-4562.







Hydrogen Datasets

Quantitative analysis of hydrogen datasets

	Aramco 2.0	DTU	GRI 3.0	USC II
H1	11.43	25.12	63.45	50.70
H2	12.20	20.58	79.90	74.33
H3	89.13	116.67	112.95	70.82
Average	***37.59***	***54.12***	***85.43***	***65.28***
No. Best Fit	***2***	***0***	***0***	***1***

Hydrogen Datasets

Quantitative analysis of hydrogen datasets

	Aramco 2.0	DTU	GRI 3.0	USC II
H1	11.43	25.12	63.45	50.70
H2	12.20	20.58	79.90	74.33
H3	89.13	116.67	112.95	70.82
Average	***37.59***	***54.12***	***85.43***	***65.28***
No. Best Fit	***2***	***0***	***0***	***1***

Hydrogen Datasets

Quantitative analysis of hydrogen datasets

	Aramco 2.0	DTU	GRI 3.0	USC II
H1	11.43	25.12	63.45	50.70
H2	12.20	20.58	79.90	74.33
H3	89.13	116.67	112.95	70.82
Average	***37.59***	***54.12***	***85.43***	***65.28***
No. Best Fit	***2***	***0***	***0***	***1***

Hydrogen Datasets

Hydrogen datasets studied.

Dataset	Reference	Average Pressure /atm	Equivalence Ratio (Φ)	CO_2 Dilution (%)
H1	Shao et al. (2019)	109.6	1.00	85.00
H2	Shao et al. (2019)	270.6	1.00	85.00
H3	Shao et al. (2019)	38.4	0.25	85.00

J.K. Shao, R. Choudhary, D.E. Davidson, R.K. Hanson, S. Barak, S. Vasu, Ignition delay times of methane and hydrogen highly diluted in carbon dioxide at high pressures up to 300 atm, Proceedings of the Combustion Institute 37 (2019) 4555-4562.

Hydrogen Datasets

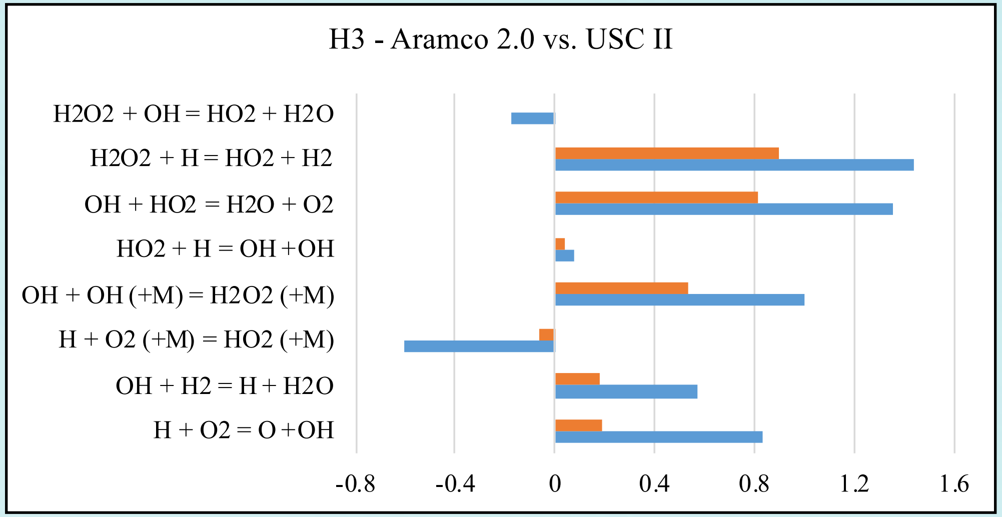
Quantitative analysis of hydrogen datasets

	Aramco 2.0	DTU	GRI 3.0	USC II
H1	11.43	25.12	63.45	50.70
H2	12.20	20.58	79.90	74.33
H3	89.13	116.67	112.95	70.82
Average	***37.59***	***54.12***	***85.43***	***65.28***
No. Best Fit	***2***	***0***	***0***	***1***

Hydrogen Datasets

Quantitative analysis of hydrogen datasets

	Aramco 2.0	DTU	GRI 3.0	USC II
H1	11.43	25.12	63.45	50.70
H2	12.20	20.58	79.90	74.33
H3	89.13	116.67	112.95	70.82
Average	***37.59***	***54.12***	***85.43***	***65.28***
No. Best Fit	***2***	***0***	***0***	***1***

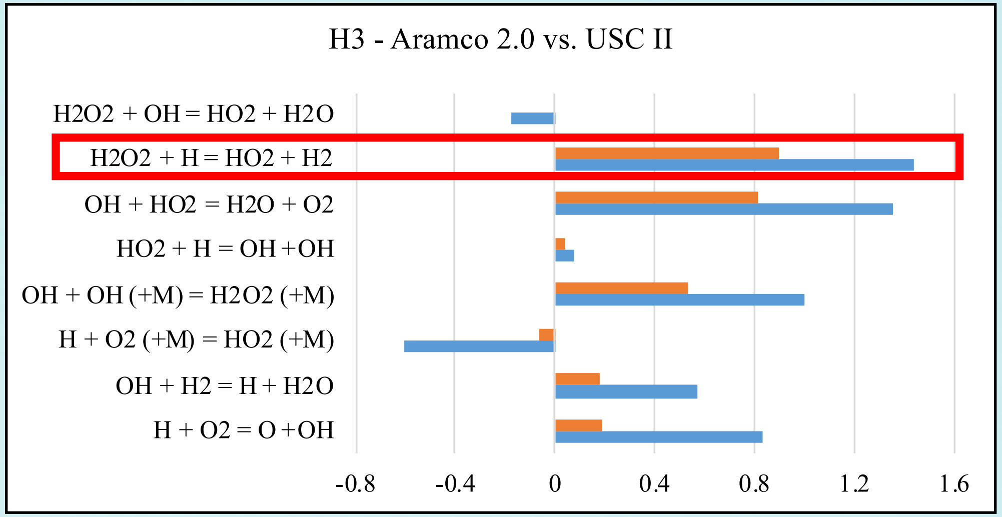


Comparison of the sensitivity analysis of H3 for Aramco 2.0 and USC II at 1274 K.

Hydrogen Datasets

Quantitative analysis of hydrogen datasets

	Aramco 2.0	DTU	GRI 3.0	USC II
H1	11.43	25.12	63.45	50.70
H2	12.20	20.58	79.90	74.33
H3	89.13	116.67	112.95	70.82
Average	***37.59***	***54.12***	***85.43***	***65.28***
No. Best Fit	***2***	***0***	***0***	***1***



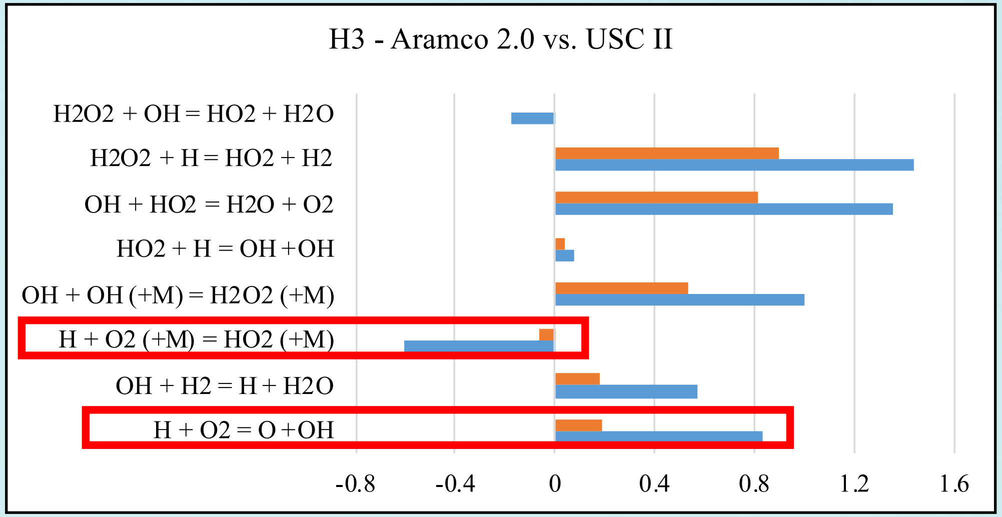
Comparison of the sensitivity analysis of H3 for Aramco 2.0 and USC II at 1274 K.

R7 - $H_2O_2 + H = H_2 + HO_2$

Hydrogen Datasets

Quantitative analysis of hydrogen datasets

	Aramco 2.0	DTU	GRI 3.0	USC II
H1	11.43	25.12	63.45	50.70
H2	12.20	20.58	79.90	74.33
H3	89.13	116.67	112.95	70.82
Average	***37.59***	***54.12***	***85.43***	***65.28***
No. Best Fit	***2***	***0***	***0***	***1***



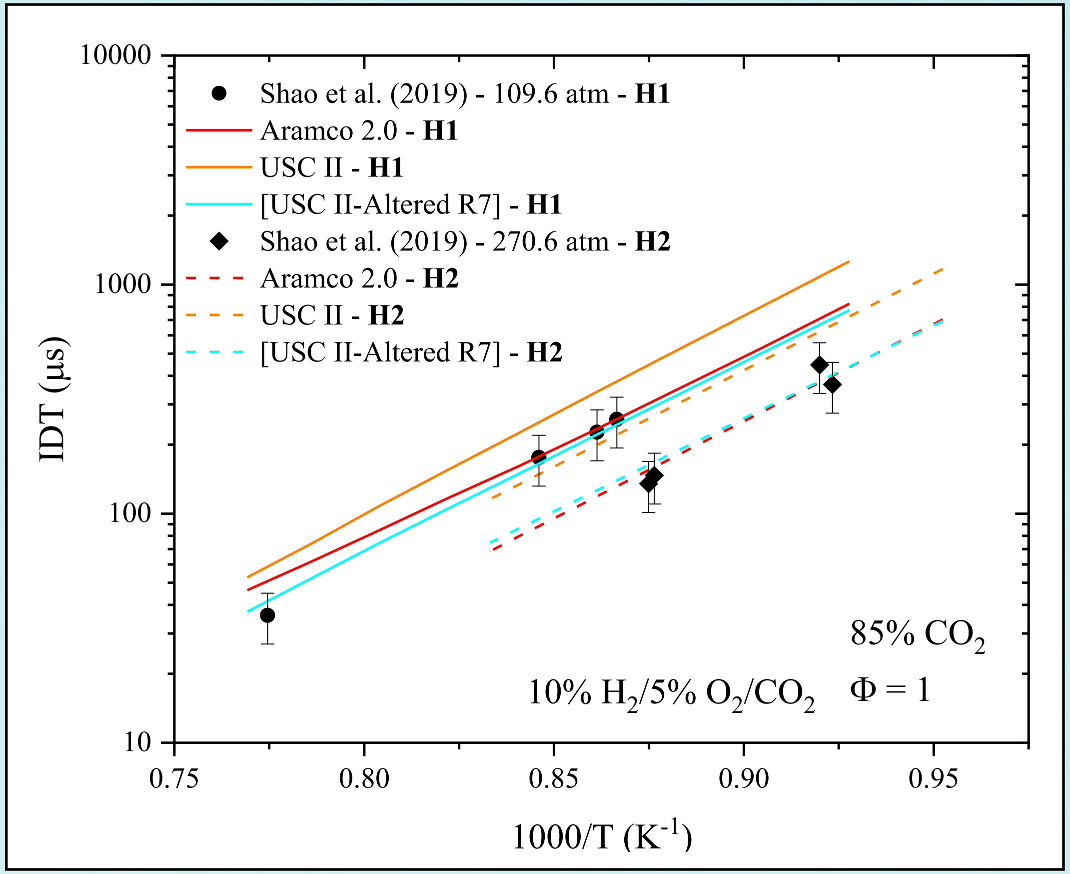
Comparison of the sensitivity analysis of H3 for Aramco 2.0 and USC II at 1274 K.

R7 - $H_2O_2 + H = H_2 + HO_2$

R8 - $O_2 + H = O + OH$

R9 - $H + O_2 (+M) = HO_2 (+M)$

Hydrogen Datasets



H1 and H2 dataset modelled by Aramco 2.0, USC II, and [USC II-Altered R7]: USC II with the updated R7 rate coefficient

J.K. Shao, R. Choudhary, D.E. Davidson, R.K. Hanson, S. Barak, S. Vasu, Ignition delay times of methane and hydrogen highly diluted in carbon dioxide at high pressures up to 300 atm, Proceedings of the Combustion Institute 37 (2019) 4555-4562.

R7 - $H_2O_2 + H = H_2 + HO_2$

Changing the rate coefficient of R7 in USC II from that of Aramco 2.0 reduces the ignition delay time and better matches the experimental data.

Syngas Datasets

Syngas datasets studied.

Dataset	Reference	Average Pressure /atm	Equivalence Ratio (Φ)	CO_2 Dilution (%)
S1	Barak et al. (2020)	78.9	1.02	91.80
S2	Barak et al. (2020)	91.7	0.41	64.50
S3	Barak et al. (2020)	89.6	0.41	92.20
S4	Barak et al. (2020)	89.7	1.09	63.90
S5	Barak et al. (2018)	41.5	1.00	85.00
S6	Barak et al. (2018)	38.6	1.00	85.00
S7	Barak et al. (2018)	38.5	1.00	85.00
S8	Barak et al. (2018)	38.4	1.00	85.00

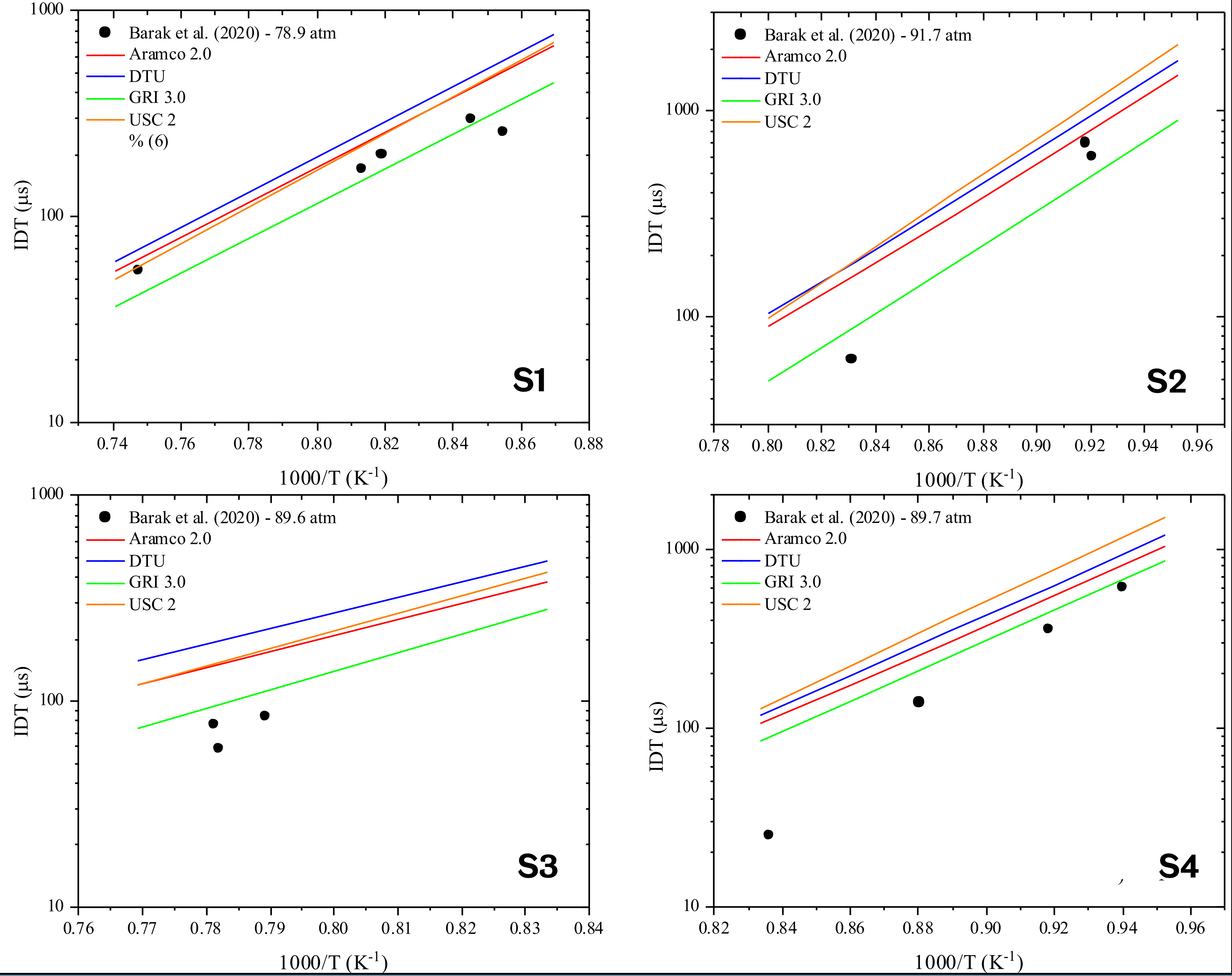
S. Barak, O. Pryor, E. Ninnemann, S. Neupane, S. Vasu, X. Lu, B. Forrest, Ignition delay times of oxy-syngas and oxy-methane in supercritical CO_2 mixtures for direct-fired cycles, Journal of Engineering for Gas Turbines and Power 142 (2020).
S. Barak, E. Ninnemann, S. Neupane, F. Barnes, J. Kapat, S. Vasu, High-pressure oxy-syngas ignition delay times with CO_2 dilution: Shock tube measurements and comparison of the performance of kinetic mechanisms, Journal of Engineering for Gas Turbines and Power 141 (2018).

Syngas Datasets

Quantitative analysis of syngas datasets

	Aramco 2.0	DTU	GRI 3.0	USC II
S1	53.20	70.06	21.84	49.96
S2	66.05	94.50	31.40	109.03
S3	117.50	181.42	32.33	123.44
S4	126.76	158.27	84.69	196.60
S5	240.64	303.37	170.24	242.84
S6	259.59	341.14	169.83	277.12
S7	191.64	277.53	241.41	132.09
S8	174.54	287.50	139.00	89.63
Average E	***153.74***	***214.22***	***111.34***	***152.59***
No. Best Fit	***0***	***0***	***6***	***2***

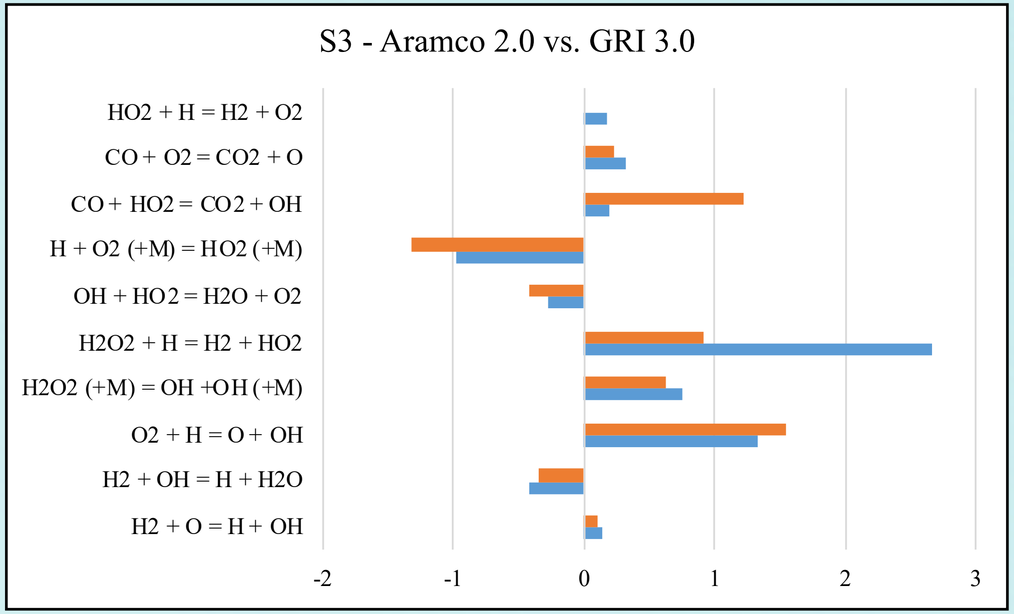
Syngas Datasets



Syngas Datasets

Sensitivity analysis and identification of important syngas reactions.

R10 - $CO + HO_2 = CO_2 + OH$



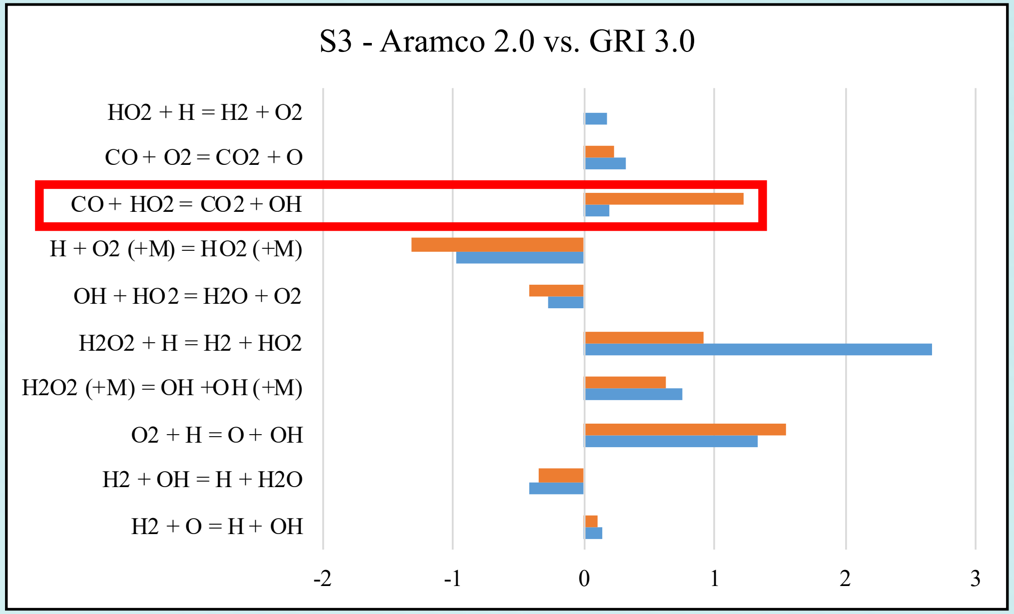
S3 dataset sensitivity analysis of GRI 3.0 and Aramco 2.0 at 1280 K.

D.L. Baulch, D.D. Drysdale, J. Duxbury, S. Grant, Evaluated kinetic data for high temperature reactions, Vol. 3: Homogeneous gas phase reactions of the O_2-O_3 systems, the CO-O_2-H_2 system, and of sulphur-containing species, Butterworths,1976

Syngas Datasets

Sensitivity analysis and identification of important syngas reactions.

R10 - $CO + HO_2 = CO_2 + OH$



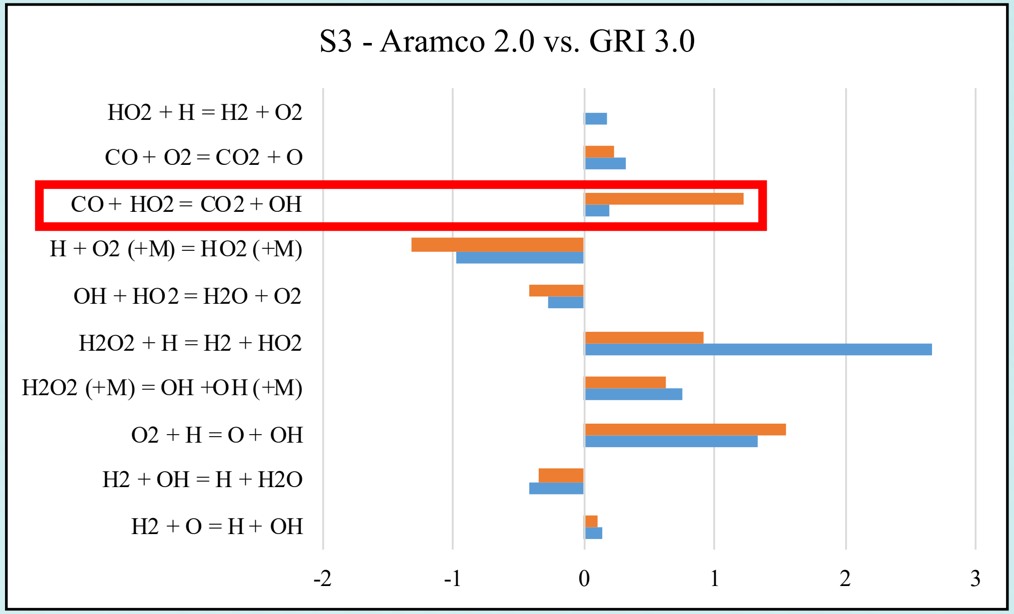
S3 dataset sensitivity analysis of GRI 3.0 and Aramco 2.0 at 1280 K.

D.L. Baulch, D.D. Drysdale, J. Duxbury, S. Grant, Evaluated kinetic data for high temperature reactions, Vol. 3: Homogeneous gas phase reactions of the O_2-O_3 systems, the CO-O_2-H_2 system, and of sulphur-containing species, Butterworths,1976

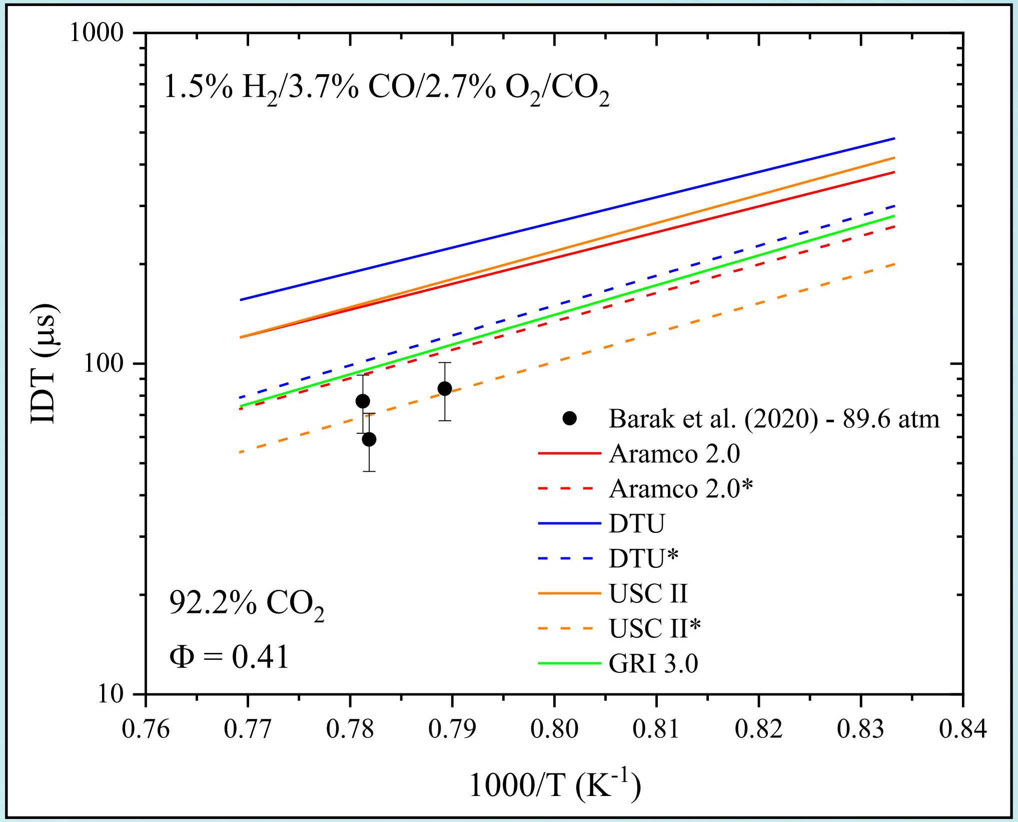
Syngas Datasets

Sensitivity analysis and identification of important syngas reactions.

R10 - $CO + HO_2 = CO_2 + OH$



S3 dataset sensitivity analysis of GRI 3.0 and Aramco 2.0 at 1280 K.



Mechanism comparison for the S3 condition with the R10 rate coefficient from Baulch et al. (1976) denoted by *.

D.L. Baulch, D.D. Drysdale, J. Duxbury, S. Grant, Evaluated kinetic data for high temperature reactions, Vol. 3: Homogeneous gas phase reactions of the O_2-O_3 systems, the CO-O_2-H_2 system, and of sulphur-containing species, Butterworths,1976

Mechanism Creation – UoS sCO_2

The challenge is to culminate the information gained from the sensitivity analysis to create one mechanism which can best model the conditions relevant to sCO_2 combustion.

Mechanism Creation – UoS sCO_2

The challenge is to culminate the information gained from the sensitivity analysis to create one mechanism which can best model the conditions relevant to sCO_2 combustion.

USC II was chosen as a starting mechanism which was updated with rate coefficients which were identified as important through the sensitivity analysis.

Mechanism Creation – UoS sCO_2

The challenge is to culminate the information gained from the sensitivity analysis to create one mechanism which can best model the conditions relevant to sCO_2 combustion.

USC II was chosen as a starting mechanism which was updated with rate coefficients which were identified as important through the sensitivity analysis.

In total 16 reactions were added to incorporate important chemistry of CH_3O_2 which was identified as an important intermediate in high-pressure methane combustion.

Mechanism Creation – UoS sCO_2

The challenge is to culminate the information gained from the sensitivity analysis to create one mechanism which can best model the conditions relevant to sCO_2 combustion.

USC II was chosen as a starting mechanism which was updated with rate coefficients which were identified as important through the sensitivity analysis.

In total 16 reactions were added to incorporate important chemistry of CH_3O_2 which was identified as an important intermediate in high-pressure methane combustion.

9 reactions were altered to rate coefficients from Aramco 2.0 and GRI 3.0 to better model individual datasets.

Final Mechanism – UoS sCO_2

Results from quantitative analysis.

$$E = \frac{1}{N}\sum_{i=1}^{N}\left|\frac{X_{sim,i} - X_{exp,i}}{X_{exp,i}}\right| \times 100$$

Fuel		Aramco 2.0	DTU	GRI 3.0	USC II	UoS sCO_2
Hydrogen	E (%)	37.6	54.1	85.4	65.3	17.5
	No. Best Fit	1	0	0	0	2
Methane	E (%)	29.7	31.8	60.7	61.7	25.1
	No. Best Fit	2	0	2	4	3
Syngas	E (%)	153.7	214.2	111.3	152.6	76.2
	No. Best Fit	0	0	3	0	5
Average E		73.7	100.0	85.8	93.2	39.6
Total No. Best Fit		3	0	5	4	10

Final Mechanism – UoS sCO_2

Results from quantitative analysis.

$$E = \frac{1}{N}\sum_{i=1}^{N}\left|\frac{X_{sim,i} - X_{exp,i}}{X_{exp,i}}\right| \times 100$$

Fuel		Aramco 2.0	DTU	GRI 3.0	USC II	UoS sCO_2
Hydrogen	E (%)	37.6	54.1	85.4	65.3	17.5
	No. Best Fit	1	0	0	0	2
Methane	E (%)	29.7	31.8	60.7	61.7	25.1
	No. Best Fit	2	0	2	4	3
Syngas	E (%)	153.7	214.2	111.3	152.6	76.2
	No. Best Fit	0	0	3	0	5
Average E		73.7	100.0	85.8	93.2	39.6
Total No. Best Fit		3	0	5	4	10

Final Mechanism – UoS sCO_2

Results from quantitative analysis.

$$E = \frac{1}{N}\sum_{i=1}^{N}\left|\frac{X_{sim,i} - X_{exp,i}}{X_{exp,i}}\right| \times 100$$

Fuel		Aramco 2.0	DTU	GRI 3.0	USC II	UoS sCO_2
Hydrogen	E (%)	37.6	54.1	85.4	65.3	17.5
	No. Best Fit	1	0	0	0	2
Methane	E (%)	29.7	31.8	60.7	61.7	25.1
	No. Best Fit	2	0	2	4	3
Syngas	E (%)	153.7	214.2	111.3	152.6	76.2
	No. Best Fit	0	0	3	0	5
Average E		73.7	100.0	85.8	93.2	39.6
Total No. Best Fit		3	0	5	4	10

Final Mechanism – UoS sCO_2

Results from quantitative analysis.

$$E = \frac{1}{N}\sum_{i=1}^{N}\left|\frac{X_{sim,i} - X_{exp,i}}{X_{exp,i}}\right| \times 100$$

Fuel		Aramco 2.0	DTU	GRI 3.0	USC II	UoS sCO_2
Hydrogen	E (%)	37.6	54.1	85.4	65.3	17.5
	No. Best Fit	1	0	0	0	2
Methane	E (%)	29.7	31.8	60.7	61.7	25.1
	No. Best Fit	2	0	2	4	3
Syngas	E (%)	153.7	214.2	111.3	152.6	76.2
	No. Best Fit	0	0	3	0	5
Average E		73.7	100.0	85.8	93.2	39.6
Total No. Best Fit		3	0	5	4	10

Final Mechanism – UoS sCO_2

Results from quantitative analysis.

$$E = \frac{1}{N}\sum_{i=1}^{N}\left|\frac{X_{sim,i} - X_{exp,i}}{X_{exp,i}}\right| \times 100$$

Fuel		Aramco 2.0	DTU	GRI 3.0	USC II	UoS sCO_2
Hydrogen	E (%)	37.6	54.1	85.4	65.3	17.5
	No. Best Fit	1	0	0	0	2
Methane	E (%)	29.7	31.8	60.7	61.7	25.1
	No. Best Fit	2	0	2	4	3
Syngas	E (%)	153.7	214.2	111.3	152.6	76.2
	No. Best Fit	0	0	3	0	5
Average E		73.7	100.0	85.8	93.2	39.6
Total No. Best Fit		3	0	5	4	10

Chemical Kinetic Mechanism for Combustion in Supercritical Carbon Dioxide

Presented by, James M. Harman-Thomas
jmharman-thomas1@sheffield.ac.uk

Thank you for listening, any questions?

Grant number: EP/S022996/1

UKRI Engineering and Physical Sciences Research Council

The University Of Sheffield.

Resilient Decarbonised Fuel Energy Systems