Supplementary information

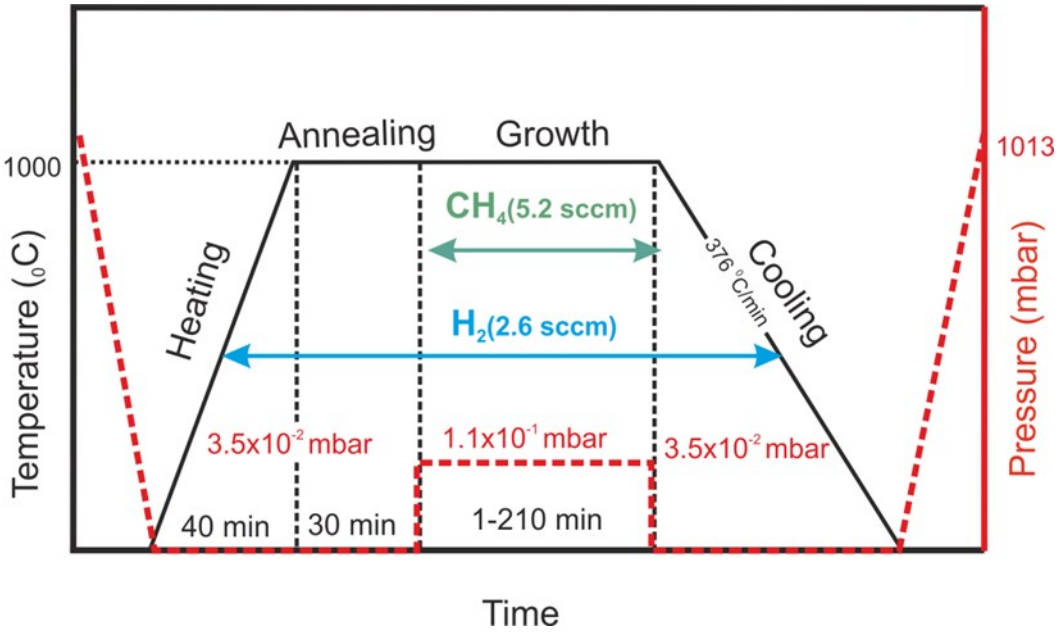

Figure S 1. Schematic illustrates the experimental procedure of CVD growth process.

Table S 1. Substrate specifications used in this work.

Substrate	Purity	Typical Analysis (ppm)	Supplier	Note
Nickel	99.98%	Co 8, Cr 8, Cu 10, Fe 10, Mg 10, Mn 10, Si 8, Ti 10, C 70, S 10	Goodfellow	Annealed
Copper	99.99%	Ag 70, Al 1, Bi 1, Ca 1, Cr <1, Fe 2, Mg 1, Mn <1, Na <1, Ni 2, Pb 2, Si 2, Sn 1	Goodfellow	As rolled
Cu70/Ni30	-	Cu 67.3%, Ni 31.0%, Mn 1.0%, Fe 0.7%.	Goodfellow	As rolled
Cu55/Ni45	-	Fe 2500, Mn 7500, Ni 45%, Cu balance	Goodfellow	As rolled
Cu33/Ni67	-		Alfa Aesar	

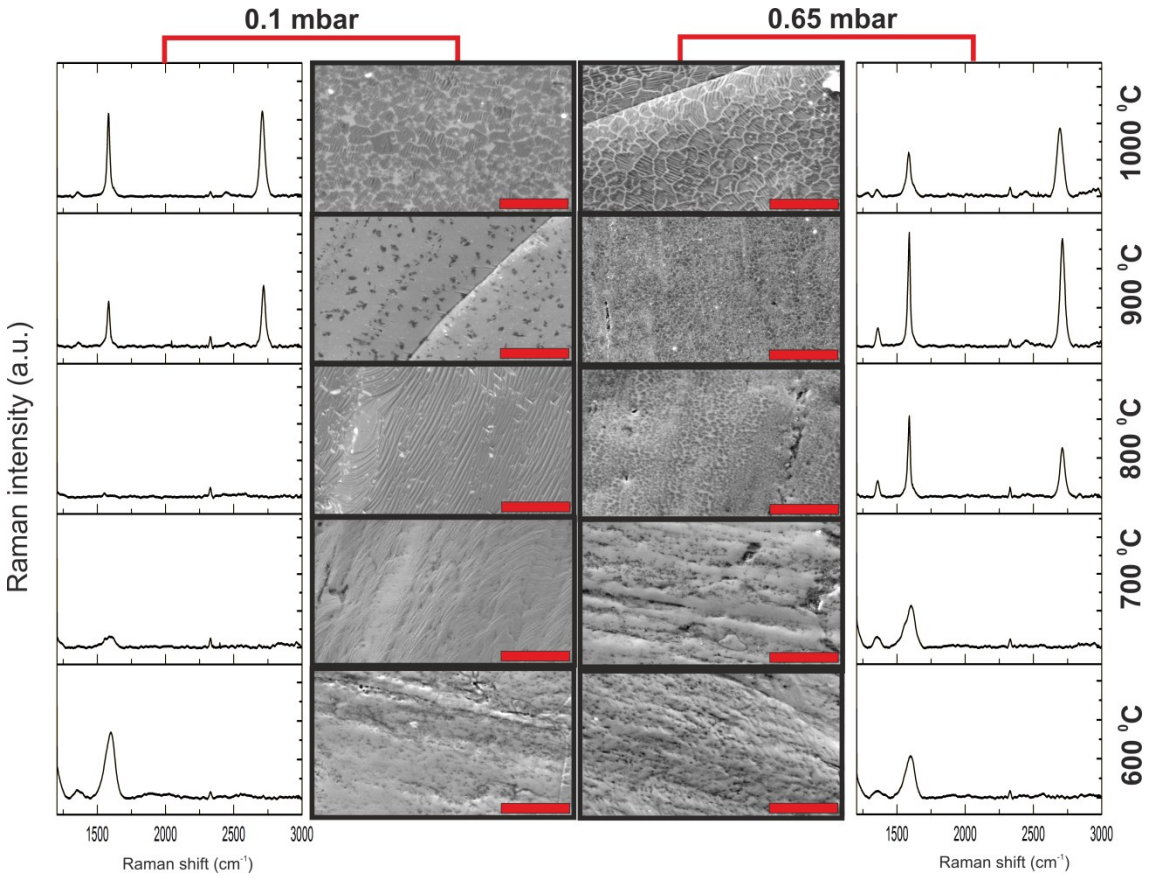

Figure S 2. Panel shows Raman spectrum and correspondence SEM micrograph for CVD graphene grown over a 500µm copper substrate. Scale bar: 5µm. The growth conditions are as follows: temperatures 600 -1000 °C, pressure 0.1 and 0.6 mbar, CH4: H2 ratio 2:1 and exposure time 30 minutes.

The fluid flow interface in COMSOL based on Navier-Stokes equations which in this work take the form [1]:

$$\rho(u.\nabla)u = \nabla.\left[-pI + \mu\left(\nabla u + (\nabla u)^T\right) - \frac{2}{3}\mu(\nabla.u)I\right] + F \qquad S\,1$$

where:

ρ is the density (kg/m^3), u is the velocity vector (m/s), p is the pressure (Pa), F is the volume force vector (N/m^3), T is the absolute temperature (K), µ is the dynamic viscosity (Pa.s) and I is the identity matrix.

The heat -transfer and fluid interface in COMSOL based on the flowing equation:

$$\rho C_p\left(\frac{\partial T}{\partial t} + (u.\nabla)T\right) = -(\nabla.q) + \tau{:}S - \frac{T\partial\rho}{\rho\partial T}|_p\left(\frac{\partial p}{\partial t} + (u.\nabla)p\right) + Q \qquad S2$$

Where:

ρ is the density (kg/m^3), C_p is the specific heat capacity at constant pressure (J/kg.K), T is absolute temperature (K), u is velocity vector (m/s), q is heat flux by conduction (W/m^2), p is pressure (pa), τ is the viscous stress tensor (pa), S is strain-rate tensor (1/s) and Q contains heat sources other than viscous heating (W/m^3). S is given by:

$$S = \frac{1}{2}(\nabla u + (\nabla u)^T) \qquad S3$$

The fluid input parameters are calculated for a CH_4/H_2 gas mixture using equations (6.5)-(6.11) [2] for the same mixing ratio stated in the experimental chapter (CH_4 and H_2 2:1 by volume respectively).

$$C_{P-mix} = \frac{x_1 C_{P1} + x_2 C_{P2}}{x_1 M_1 + x_2 M_2} \qquad S4$$

$$C_{v-mix} = \frac{x_1 C_{v1} + x_2 C_{v2}}{x_1 M_1 + x_2 M_2} \qquad S5$$

$$\rho_{mix} = x_1\rho_1 + x_2\rho_2 \qquad S6$$

$$k_{mix} = \sum_{i=1}^{n} \frac{x_i \mu_i}{\sum_{j=1}^{n} x_j \emptyset_{k,ij}} \qquad S7$$

$$\emptyset_{k,ij} = \frac{1}{\sqrt[2]{2}}\left(1 + \frac{M_i}{M_j}\right)^{-\frac{1}{2}}\left[1 + \left(\frac{k_i}{k_j}\right)^{\frac{1}{2}}\left(\frac{M_j}{M_i}\right)^{\frac{1}{4}}\right]^2 \qquad S8$$

$$\mu_{mix} = \sum_{i=1}^{n} \frac{x_i \mu_i}{\sum_{j=1}^{n} x_j \emptyset_{k,ij}} \qquad S9$$

$$\emptyset_{k,ij} = \frac{1}{\sqrt[2]{2}}\left(1 + \frac{M_i}{M_j}\right)^{-\frac{1}{2}}\left[1 + \left(\frac{\mu_i}{\mu_j}\right)^{\frac{1}{2}}\left(\frac{M_j}{M_i}\right)^{\frac{1}{4}}\right]^2 \qquad S10$$

where:

C_p, C_v is the specific heat at constant pressure and volume respectively, x is the mole fraction, M is the molecular weight, ρ is the density, k is the thermal conductivity and μ is the viscosity.

The required input mixture properties are listed in Table 6.1. The physical properties values of gas mixture were entered into the model as an equation with respect to temperature to enable the model to work with heat transfer consideration over a range of temperature.

Table S 2. Gas mixture properties, all the listed values are calculated for 2:1 CH_4: H_2.

Property	**Symbol**	**Unit**	**Value**
Dynamic viscosity	μ	Pa.s	$(-2 \times 10^{11})T^2 + (6 \times 10^{-8})T + (3 \times 10^{-6})$
Ratio of specific heat	γ	-	1.335
Heat capacity	C_p	J/(Kg.K)	$-0.0008T^2 + 4.2083T + 1783.5$
Density	ρ	Kg/m3	$0.01114P_A/8.1225T$
Thermal conductivity	k	W/(m.K)	$(-3 \times 10^{-8})T^2 + (0.0006)T + 0.0309$

For the metal substrate and quartz tube, the physical properties are assigned from COMSOL materials library itself.

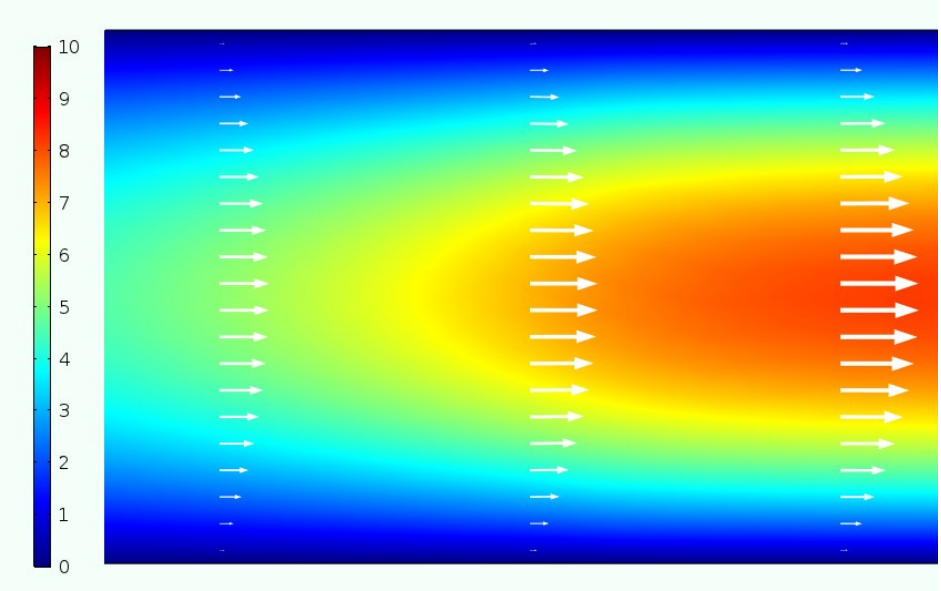

Figure S 3. Gas velocity distribution, at the point between the cold and hot zone. The gas velocity increase rapidly when enters the isothermal zone. However, the gas velocity near the reactor wall is ~ 0 m/s. The velocity scale bar units are m/s.

Table S 3. Elementary chemical reactions used in the model [3–5]

Eqn.	Reaction	A (mol/cm^3.s)	n	E (J/mol)
1	$CH_4 = CH_3 + H$	3.51×10^{15}	0	435344
2	$CH_4 + H = CH_3 + H_2$	2.25×10^{4}	3	36702.85
3	$CH_3 + CH_3 = C_2H_6$	1.01×10^{15}	-0.64	0
4	$C_2H_6 + H = C_2H_5 + H_2$	5.54×10^{2}	3.5	21658.36
5	$C_2H_6 + CH_3 = C_2H_5 + CH_4$	0.55	4	34727.06
6	$C_2H_5 = C_2H_4 + H$	2.00×10^{13}	0	166184.2
7	$CH_3 + CH_3 = C_2H_4 + H_2$	1.00×10^{16}	0	133952
8	$C_2H_4 + CH_3 = C_2H_3 + CH_4$	6.62	3.7	39817.23
9	$C_2H_4 + CH_3 = C_3H_7$	3.31×10^{11}	0	32294.99
10	$C_2H_4 + H = C_2H_3 + H_2$	1.32×10^{6}	2.53	51311.99
11	$C_2H_3 = C_2H_2 + H$	1.93×10^{28}	-4.783	214000.9
12	$CH_3 + C_2H_3 = C_3H_6$	1.00×10^{13}	0	0
13	$C_3H_7 = C_3H_6 + H$	1.58×10^{16}	0	159068

14	$C_3H_6 = C_3H_5 + H$	1.00×10^{15}	0	368368
15	$C_3H_5 = C_2H_2 + CH_3$	3.16×10^{10}	0	151533.2
16	$C_3H_5 = C_3H_4 + H$	5.00×10^{9}	0	146510
17	$C_3H_5 + H = C_3H_4 + H_2$	1.00×10^{13}	0	0
18	$C_3H_6 + H = C_3H_5 + H_2$	3.16×10^{11}	0	18837
19	$C_2H_3 + C_2H_3 = C_4H_6$	1.26×10^{13}	0	0
20	$C_2H_3 + C_2H_4 = C_4H_6 + H$	5.00×10^{11}	0	30620.59
21	$C_2H_2 + H = C_2H + H_2$	6.02×10^{13}	0	93347.8
22	$C_2H_2 + CH_3 = C_2H + CH_4$	1.81×10^{11}	0	72417.8
23	$C_4H_6 + H = C_4H_5 + H_2$	1.00×10^{14}	0	62790
24	$C_4H_5 = C_4H_4 + H$	1.00×10^{14}	0	173300.4
25	$C_2H + H = C_2H_2$	1.81×10^{14}	0	0
26	$C_2H_3 + C_2H_2 = C_4H_5$	1.10×10^{12}	0	16744
27	$CH_3 + CH_3 = C_2H_5 + H$	1.80×10^{12}	0	43534.4
28	$C_4H_5 + C_2H_2 = C_6H_6 + H$	6.02×10^{12}	0	37674
29	$C_2H_4 = C_2H_3 + H$	1.00×10^{16}	0	452088
30	$C_2H_5 + C_2H_2 = C_2H_6 + C_2H$	2.71×10^{11}	0	97952.4
31	$C_2H_5 + H = C_2H_6$	3.07×10^{13}	0	0
32	$C_2H_4 = C_2H_2 + H_2$	7.94×10^{12}	0.44	371549.4
33	$C_2H_3 + H = C_2H_2 + H_2$	9.64×10^{13}	0	0
34	$C_2H_2 + CH_3 = C_3H_4 + H$	6.20×10^{11}	0	83720
35	$C_3H_6 = C_3H_4 + H_2$	8.00×10^{12}	0.44	339693.9
36	$C_3H_6 + CH_3 = C_3H_5 + CH_4$	1.58×10^{12}	0	36836.8

1. **Chemical species*:* CH_4, CH_3, H, H_2, C_2H_6, C_2H_5, C_2H_4, C_2H_3, C_2H_2, C_2H, C_3H_7, C_3H_6, C_3H_5, C_3H_4, C_4H_6, C_4H_5, C_4H_4, C_6H_6.**
2. **Where A is the pre-exponential factor (mol/cm3.s), n the temperature exponent, E the activation energy (J/mol)**

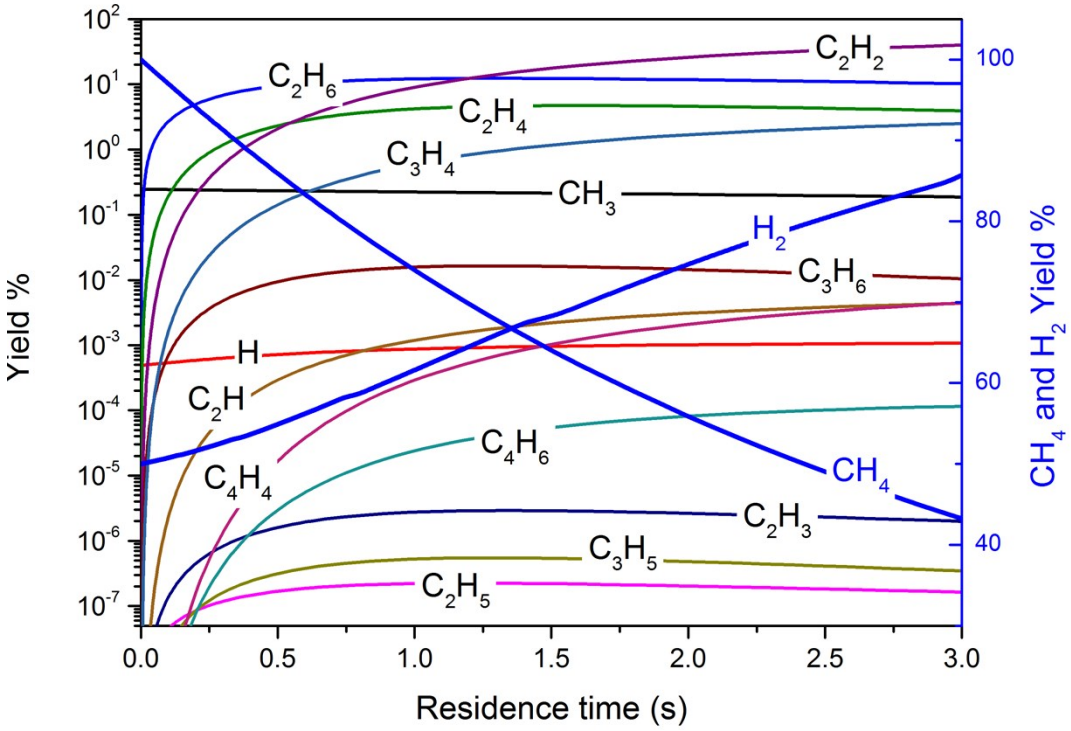

Figure S 4. Chemical species yield versus residence time of CH_4 and H_2 mixture 2:1 by volume mixing ratio at 1000 °C and 0.1 mbar pressure. The conversion of CH_4 increase with increasing residence time and C_2H_2 shows the highest yield after 3 seconds. The blue y-axis is the CH_4 and H_2 yield.

(1) COMSOL Multiphysics, CFD Module User's Guide. **2016**, *V 5.2a*.

(2) Setiawan, I.; Nohtomi, M.; Katsuta, M. Critical Temperature Differences of a Standing Wave Thermoacoustic Prime Mover with Various Helium-Based Binary Mixture Working Gases. *J. Phys. Conf. Ser.* **2015**, *622*, 12010.

(3) Olsvik, O.; Rokstad, O. A.; Holmen, A. Pyrolysis of Methane in the Presence of Hydrogen. *Chem. Eng. Technol.* **1995**, *18*, 349–358.

(4) Holmen, A.; Olsvik, O.; Rokstad, O. A. Pyrolysis of Natural Gas: Chemistry and Process Concepts. *Fuel Process. Technol.* **1995**, *42*, 249–267.

(5) Olsvik, O.; Billaud, F. Modelling of the Decomposition of Methane at 1273 K in a Plug Flow Reactor at Low Conversion. *J. Anal. Appl. Pyrolysis* **1993**, *25*, 395–405.