

Relationship between the radial dynamics and the chemical production of a harmonically driven spherical bubble

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Abstract

The sonochemical activity and the radial dynamics of a harmonically excited spherical bubble are investigated numerically. A detailed model is employed capable to calculate the chemical production inside the bubble placed in water that is saturated with oxygen. Parameter studies are performed with the control parameters of the pressure amplitude, the forcing frequency and the bubble size. Three different definitions of collapse strengths (extracted from the radius vs. time curves) are examined and compared with the chemical output of various species. A mathematical formula is established to estimate the chemical output as a function of the collapse strength; thus, the chemical activity can be predicted without taking into account the chemical kinetics into the bubble model. The calculations are carried out by an in-house code exploiting the high processing power of professional graphics cards (GPUs). **Results verify the widely accepted rule in the literature that the incidence of the chemical activity happens when the relative expansion is $(R_{max} - R_E)/R_E > 2$.** Here R_E is the equilibrium bubble radius (bubble size) and R_{max} is the maximum bubble expansion before a collapse phase. After the incidence of cavitation, the chemical output increases rapidly with the relative expansion according to a power function of the form $y = \alpha x^\beta$. The large number of investigated parameter combinations (approximately two millions) allowed us to provide good estimates for the parameters $\alpha(R_E)$ and $\beta(R_E)$ as a function of the bubble size R_E .

Keywords: bubble dynamics, sonochemistry, collapse strength, chemical production, GPU programming

1. Introduction

If a liquid domain is irradiated by ultrasound, the originally dissolved gas combines into numerous micro-sized bubbles and form structures called bubble clusters [1, 2]. Due to the effect of the ultrasonic forcing (periodic pressure waves), these bubbles start to oscillate around their equilibrium state. The magnitude of this oscillation is influenced

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by the intensity of the ultrasound: the higher the pressure amplitude the stronger the oscillation. At sufficiently large excitation pressure amplitude, above Blake’s critical threshold [3], the velocity of the bubble wall can reach extremely high values. This phenomenon is often called acoustic cavitation [4].

Both experimental [5–8] and numerical [9–13] studies have shown that the bubble wall velocity can approach the sound speed of the liquid domain generating shock waves. Moreover, due to the high excitation frequency, the internal gas obeys a rather fast state of change during the compression phase compared to the speed of heat exchange. Therefore, this nearly adiabatic compression results in extreme conditions inside the bubble like thousands of degrees of Kelvin temperature or hundreds of bars of pressure around the minimum radii of the bubbles [14]. Under these extreme conditions, even chemical reactions start to take place. For instance, the dissociation of water and O_2 molecules produce H_2 molecules, OH radicals or even oxygen and hydrogen atoms. Chemical reactions generated by ultrasonic irradiation is called sonochemistry in the literature [15, 16]. It is worth noting that around the collapse phase, visible light emission was also observed experimentally [17, 18]; this phenomenon is often called sonoluminescence and can be explained by the ionization of several gas components [19–21].

These kinds of intense conditions can be exploited in several important sonochemical applications by taking advantage of the production of the aforementioned substances. In this sense, a bubble is often considered as a micron-sized chemical reactor where several types of elements are generated. These can react with each other inside the bubble or they can even leave the bubble and enter the liquid via diffusion. Inside the liquid, they usually react with other dissolved components modifying the composition of the liquid domain. In general, sonochemical behaviour is widely used in polymerization [22, 23], nanosynthesis [24], disposing pollutants [25, 26], wastewater technologies [27] or in sonodynamic therapy as a promising technology for cancer treatment [28–30].

Describing the dynamics of bubbles is certainly the most important aspect in acoustic cavitation. It has been studied by numerous researchers both numerically and experimentally in the last couple of decades [9, 31–33]. The exact behaviour of a certain bubble is influenced by many different parameters such as liquid features, gas content, excitation properties and many others [34, 35]. Depending on the current parameter values, the oscillation of a bubble generally consists of three different phases: a rather slow expansion stage when the bubble grows and the internal pressure decreases; a much more rapid collapse phase when the bubble suddenly shrinks and the pressure inside increases significantly; and finally, an occasionally appearing afterbounce phase which is a consequence of the highly non-linear nature of bubble dynamics. A typical bubble radius vs. time curve as a function of time is presented in Fig. 1 under single frequency ultrasound excitation where the aforementioned phases are highlighted. An intense collapse phase is expected in every sonochemical application [36]. Nevertheless, in some special cases, other features may also become important, for instance chaotic oscillation [37] in micromixing [38, 39].

There are plenty of studies that focus on describing the sonochemical behaviour of bubbles; essentially they can be divided into two main groups. A smaller number of studies carry out simulations by taking into account the entire chemical modelling inside the bubble [40, 41]. These models have a detailed and sophisticated implementation of the bubble interior; thus, their complexity is significantly higher compared to the classical Rayleigh approach [42]. Consequently, the numerical implementation can be

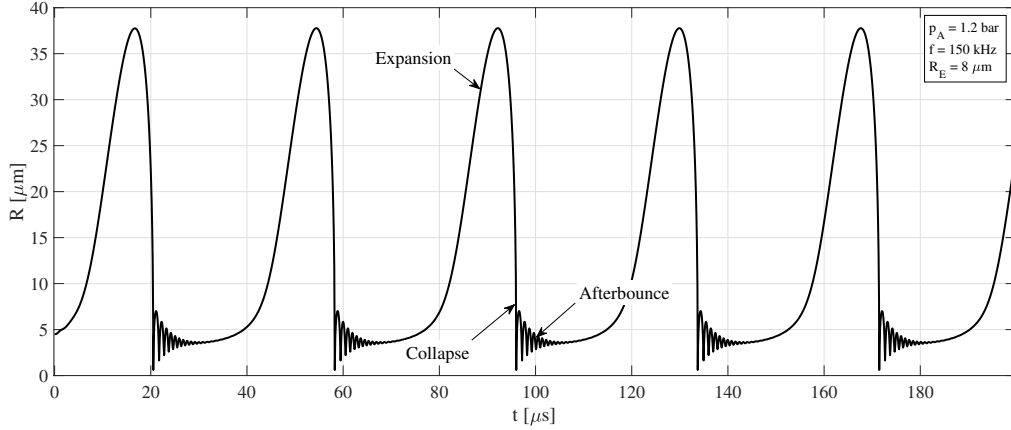


Figure 1: A typical bubble radius vs. time curve as a function of time under single frequency ultrasonic forcing. A relatively slow expansion phase is followed by a rapid collapse and some afterbounces.

a cumbersome task, especially when runtime is an important factor. *The first aim of the present paper is to implement a mathematical model, which is capable to describe the chemical kinetics inside a bubble and to exploit the high processing power of professional graphics cards (GPUs).* Therefore, complex modelling and large parameter studies can be achieved.

The other group of studies make conclusions only from the dynamical properties of the bubbles without modelling the exact internal chemical processes. They define various kinds of collapse strengths as an indicator for the chemical activity. Their obvious advantage is that the model is quite simple and easy to implement; however, they suffer from several general approximations. They state that the defined collapse strength is strongly related to chemical activity [43]; moreover, it is declared that above a given threshold of the collapse strength, the bubble is considered as chemically active. It is a simple “binary” statement about the existence of chemical reactions; however, it can not quantify the chemical output. By the best knowledge of the authors, the relationship between the bubble dynamics and the chemical activity is poorly investigated in the literature. *The second purpose of the present study is to prove the existence of a clear correlation between the chemical output and the dynamic features (collapse strength) of a bubble by solving the detailed internal chemical processes numerically. Furthermore, we aim to create mathematical formulae to characterise this relationship; thereby, propose a “tool” that can estimate the chemical output only from the dynamical attributes of the system (bubble radius vs. time curves).*

The employed liquid is water saturated with pure oxygen. In our work, the control parameters are the pressure amplitude and the frequency of the ultrasound, and the bubble size. Changing the excitation parameters is the easiest way to influence bubble behaviour in an ultrasonic technology. Such a three-dimensional parameter space gives a good overview of the behaviour of the system that can be used to find trends and dependencies between different quantities.

80 2. The physical and the mathematical model

81 The physical model applied in the present study takes into account the following
 82 effects. First of all, a single spherical bubble in water is examined in the presence of
 83 harmonic pressure excitation. The bubble contains different types of non-condensable
 84 gases (oxygen and various chemical products) and water vapour. Heat transfer, diffusion
 85 into the liquid, evaporation and condensation of water, and reaction kinetics are included
 86 in the model. The internal pressure and temperature is considered spatially uniform
 87 except in a thin thermal boundary layer described later.

The radial dynamics of the bubble is described by the Keller–Miksis equation [44] in the form of

$$\left(1 - \frac{\dot{R}}{c_L}\right) R\ddot{R} + \left(1 - \frac{\dot{R}}{3c_L}\right) \frac{3}{2}\dot{R}^2 = \left(1 + \frac{\dot{R}}{c_L} + \frac{R}{c_L} \frac{d}{dt}\right) \frac{(p_L - p_\infty(t))}{\rho_L}, \quad (1)$$

where $R(t)$ is the radius of the bubble, t is the time, c_L is the sound speed in the liquid and ρ_L is the density of the liquid. The dots stand for derivatives with respect to time. Note that the equation is of second order. The far field pressure $p_\infty(t)$ consists of static and dynamic parts written as

$$p_\infty(t) = P_\infty + p_A \sin(2\pi ft), \quad (2)$$

where P_∞ is the static ambient pressure, p_A and f are the pressure amplitude and frequency of the excitation, respectively. The liquid pressure p_L at the bubble wall is related to the internal pressure as

$$p(t) = p_L + \frac{2\sigma}{R} + 4\mu_L \frac{\dot{R}}{R}, \quad (3)$$

88 where $p(t)$ is the internal pressure of the bubble, σ is the surface tension and μ_L is the
 89 dynamic viscosity of the liquid. **It must be stressed that several researches [45–47] have**
 90 **shown that at intense collapse regimes (when the bubble can grow up to ten times of**
 91 **its equilibrium size) the bubble wall velocity can exceed the sound speed of the liquid.**
 92 **This limits the validity range of our model. In this paper, however, we are focusing on**
 93 **establishing model that describes the chemical model properly but still simple enough**
 94 **to perform large parameter studies (approximately two million parameter combinations,**
 95 **see Sec. 4). Thus this effect is neglected in the present study.**

The internal pressure of the gas mixture is calculated via the van der Waals equation of state

$$\left(p + \frac{an^2}{V^2}\right) (V - nb) = nR_g T, \quad (4)$$

96 where a and b are the van der Waals constants of the mixture, n is the amount of the gas
 97 mixture, $V = 4R^3\pi/3$ is the volume of the bubble, R_g is the universal gas constant and
 98 T is the internal temperature. The van der Waals constants of the mixture are calculated
 99 as

$$a = \frac{\sum_{i=1}^C N_i a_i}{4N_t}, \quad (5)$$

$$b = \frac{\sum_{i=1}^C N_i b_i}{N_t}, \quad (6)$$

where C is the number of the different chemical components in the bubble, N_i is the molecule number of component i , N_t is the total molecule number, a_i and b_i are the van der Waals constants of component i . In order to calculate the internal pressure from Eq.(4), one has to determine the temperature inside the bubble. It is estimated from the internal energy of the mixture, which can be written as

$$E = \left(\sum_{i=1}^C N_i c_{v,i} \right) \frac{T}{N_A} - \left(\frac{N_t}{N_A} \right)^2 \frac{a}{V}, \quad (7)$$

where N_A is the Avogadro-constant and $c_{v,i}$ is the molar heat capacity of component i calculated by

$$c_{v,i} = \frac{f_i}{2} R_g, \quad (8)$$

100 where f_i is the degree of freedom of molecule i . For monoatomic gases, $f_i = 3$; for
 101 diatomic gases, $f_i = 5$; and for gases of 3 or more atoms, $f_i = 6$. The rate of change of
 102 the internal energy will be described later in this section.

103 For the estimation of the heat transfer between the bubble and the liquid, the bubble
 104 temperature (T) is assumed to be spatially uniform except in a thermal boundary layer
 105 near the bubble wall in the internal side. In this layer, the temperature changes linearly
 106 from the internal temperature of the bubble to the wall temperature (T_R). Throughout
 107 the manuscript, T represents the uniform part of the internal temperature that governs
 108 the chemical processes. This approach of temperature distribution is a fairly simple model
 109 for the internal temperature and thermal processes. Zhang et al. [48] made a detailed
 110 research of the internal temperature and the internal pressure distributions. They have
 111 shown that the temperature is the highest mostly at the bubble center and it decreases
 112 drastically with the radial co-ordinate. Due to the reasons mentioned previously (large
 113 number of parameter combination), a simplified approach is employed here to be able to
 114 use a “non-distributed” computations of the chemical reactions.

At the bubble wall, a temperature jump occurs [49]:

$$\Delta T = -\frac{1}{2kn'} \sqrt{\frac{\pi m}{2kT}} \frac{2 - a'\alpha_e}{\alpha_e} \kappa \left. \frac{\partial T}{\partial r} \right|_{r=R}, \quad (9)$$

where k is the Boltzmann-constant, n' is the number density of the gas mixture, m is the average mass of a molecule inside the bubble, a' and α_e are constants [49], κ is the average thermal conductivity of the mixture and r is the radial coordinate. The liquid temperature T_0 is assumed to be constant, thus, the internal temperature at the wall is $T_R = T_0 + \Delta T$. The thickness of the thermal boundary layer is $n\lambda$, where n is constant and λ is the mean free path of a molecule calculated as [50]

$$\lambda = \frac{V}{\sqrt{2}\sigma' N_t}, \quad (10)$$

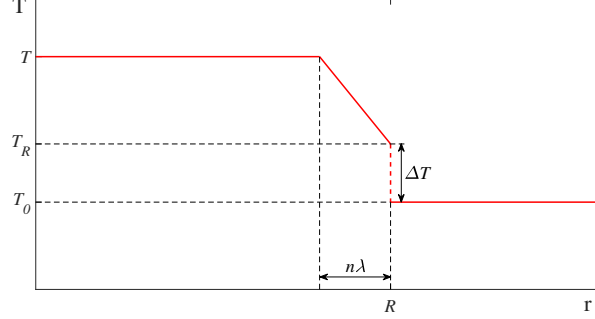


Figure 2: Sketch of the temperature distribution inside and outside the bubble. The internal temperature T remains spatially uniform except for a thermal boundary layer, where the temperature changes linearly from T to T_R . A temperature jump (ΔT) exists at the bubble wall.

where σ' is the average cross-section of the molecules. With this, the derivative of the temperature at the bubble wall became

$$\left. \frac{\partial T}{\partial r} \right|_{r=R} = \frac{T_R - T}{n\lambda}. \quad (11)$$

115 A schematic drawing about the temperature distribution is illustrated in Fig. 2.

During the oscillation of a bubble, evaporation and condensation takes place due to the change of internal pressure and temperature. The rate of evaporation for unit area and time is [51, 52]

$$\dot{m}_{eva} = \frac{\alpha_M}{\sqrt{2\pi R_v}} \frac{p_v^*}{\sqrt{T_0}}, \quad (12)$$

where $\dot{m}_{eva}$ is the rate of evaporation (in $\text{kg}/\text{m}^2\text{s}$), α_M is the accommodation coefficient for evaporation, R_v is the specific gas constant of water vapour and p_v^* is the saturated pressure of vapour (at T_0). The rate of condensation for unit area and time is calculated by the expression

$$\dot{m}_{con} = \frac{\alpha_M}{\sqrt{2\pi R_v}} \frac{\Gamma p_v}{\sqrt{T}}, \quad (13)$$

where $\dot{m}_{con}$ is the rate of condensation (in $\text{kg}/\text{m}^2\text{s}$), Γ is the correction factor and p_v is the actual partial pressure of the vapour inside the bubble, which is determined as

$$p_v = \frac{N_{\text{H}_2\text{O}}}{N_t} p, \quad (14)$$

where $N_{\text{H}_2\text{O}}$ is the number of vapor molecules inside the bubble. Now, the net rate of evaporation $\dot{m}$ for unit area and time can be expressed as

$$\dot{m} = \dot{m}_{eva} - \dot{m}_{con}. \quad (15)$$

The energy carried by an evaporating/condensing molecule is

$$e_{eva} = \frac{c_{v,\text{H}_2\text{O}}}{N_A} T_0, \quad (16)$$

$$e_{con} = \frac{c_{v,H_2O}}{N_A} T_R, \quad (17)$$

where e_{eva} and e_{con} are the energy carried by an evaporating and condensing molecule, respectively. It should be noted here that based on the measurements of Hickman [53] and Maa [54], correction factor Γ was always chosen to be unity.

The diffusion into the liquid is modelled similarly to that of heat transport. The rate of diffusion for each component is determined by [40, 55]

$$\left(\frac{dm}{dt}\right)_i = \left(D \frac{\partial c}{\partial r} \Big|_{r=R}\right)_i \approx D \frac{c_{0,i} - c_i}{l_{diff}}, \quad (18)$$

where $(dm/dt)_i$ is the rate of diffusion for component i (for unit area and time), D is the diffusion coefficient, $c_{0,i}$ is the concentration of component i at $r = \infty$, c_i is the saturation concentration of component i at the bubble wall in the liquid and l_{diff} is the diffusion length approximated by

$$l_{diff} = \min \left(\sqrt{\frac{RD}{|\dot{R}|}}, \frac{R}{\pi} \right). \quad (19)$$

The saturation concentration c_i is expressed by Henry's law as

$$c_i = \frac{10^3 \rho_L N_A}{M_{H_2O} K_B} p_i, \quad (20)$$

where K_B is the Henry-constant in water and p_i is the partial pressure of component i inside the bubble calculated by Dalton's law as

$$p_i = \frac{N_i}{N_t} p. \quad (21)$$

The energy carried by a diffusing molecule is

$$e_i = \begin{cases} \frac{c_{v,i}}{N_A} T_R, & \text{if } \left(\frac{dm}{dt}\right)_i < 0, \\ \frac{c_{v,i}}{N_A} T_0, & \text{if } \left(\frac{dm}{dt}\right)_i > 0, \end{cases} \quad (22)$$

where e_i is the energy carried by one diffusing molecule of component i .

The chemical reactions taking place inside the bubble are estimated as follows. Let us consider the reaction



Here, A and B are called reactants, C and D are called products. In each reaction, one molecule of each reactant is consumed and one molecule of each product is produced. The rate of reaction γ is calculated by the modified Arrhenius-equation as

$$r_\gamma = k_\gamma [A][B] = A_\gamma T^{b_\gamma} e^{-\frac{c_\gamma}{T}} [A][B], \quad (24)$$

where r_γ is the rate of reaction γ , k_γ is the Arrhenius-coefficient of reaction γ , the brackets mean the concentration of the component. The quantities A_γ , b_γ and c_γ are constants specific to reaction γ . The reaction rates relate to unit volume and unit time. Naturally, one component can take part in more than one chemical reactions. Consequently, the number of the molecules of a specific component changes due to every reactions it takes part in. As a result, the rate of change of the number of the molecules of a component is

$$\dot{N}_i = V \sum (r_{i,\text{prod}} - r_{i,\text{destr}}) + A \left(\frac{dm}{dt} \right)_i, \quad (25)$$

where $r_{i,\text{prod}}$ and $r_{i,\text{destr}}$ are the sum of every reaction rates where component i takes place in as product and reactant, respectively. Here, A stands for the surface of the bubble, since diffusion into the liquid also affects the molecule number. Only the molecule number of vapour is treated differently as evaporation and condensation have to be taken into account:

$$\dot{N}_{\text{H}_2\text{O}} = V \sum (r_{\text{H}_2\text{O},\text{prod}} - r_{\text{H}_2\text{O},\text{destr}}) + A \dot{m}. \quad (26)$$

As it is mentioned before, the rate of change of the internal energy has to be determined. It is calculated by the first law of thermodynamics:

$$\dot{E} = -p\dot{V} + \dot{Q}, \quad (27)$$

where $\dot{E}$ is the rate of change of the internal energy and $\dot{Q}$ is the sum of heats transferred into the bubble by each physical effect. Here, it is calculated by

$$\begin{aligned} \dot{Q} = A \left(\sum_{i=1}^C \left(\frac{dm}{dt} \right)_i e_i + \kappa \frac{\partial T}{\partial r} \Big|_{r=R} + \frac{10^3 N_A}{M_{\text{H}_2\text{O}}} \dot{m}_{\text{eva}} e_{\text{eva}} - \frac{10^3 N_A}{M_{\text{H}_2\text{O}}} \dot{m}_{\text{con}} e_{\text{con}} \right) \\ + V \left(\sum_{\gamma} (r_{\gamma,b} - r_{\gamma,f}) \Delta H_{\gamma,f} \right), \quad (28) \end{aligned}$$

where γ stands for every reactions taken into account. The indices f and b denote the forward and backward reactions, respectively. $\Delta H_{\gamma,f}$ means the reaction heat of the forward reaction. The first term in the first parentheses is the energy carried by diffusing molecules, the second term is the thermal heat flux into the bubble. The third and fourth terms are the energy carried by evaporating and condensing water molecules, respectively. The second parenthesis stands for the heat change due to chemical reactions.

Finally, the overall ordinary differential equation (ODE) system that has to be solved consists of the following equations: the Keller–Miksis-equation (1), the energy equation (27) and the molecule number equations for all different chemical substances taken into account by Eqs. (25) and (26). This results in an ODE system of $C + 3$ equations, where C is the number of different chemical species (the second order Keller–Miksis-equation is solved as a first order system). During each ODE function evaluation, one has to calculate the internal temperature and pressure from Eqs. (4) and (7), then the different quantities in Eq. (28). This includes thermal conduction, evaporation, diffusion and reaction kinetics.

The numerical method, that is applied here to solve the system is the Runge–Kutta–Casp–Karp-method, which is a 4th-5th order explicit scheme with embedded error estimation. It must be stressed that from Eq. (4), expressing the derivative of p , needed in

the Keller–Miksis equation, would become extremely complicated, therefore it is approximated linearly as

$$\left(\frac{dp}{dt}\right)_j \approx \frac{p_j - p_{j-1}}{t_j - t_{j-1}}, \quad (29)$$

where j relates to the current time step. Surely, an initial value of dp/dt has to be specified for $j = 0$. In our calculations, the Keller–Miksis equation and the energy equation is solved in dimensionless form, the process is described in Appendix A in detail.

In the present work, we assumed initially pure O₂ and water vapour bubbles similarly to the investigations by [56–58]. We considered altogether 9 different molecules and 44 different chemical reactions (22 reactions with their backward reactions as well). The equations, the A, b, c and ΔH_f constants are given in Tab. 2 [59–61]. This results in an ODE system of $9 + 3 = 12$ equations.

The main purpose of the present study is to investigate the chemical production of a driven bubble in the $(f - p_A)$ plane for various bubble sizes. In our survey, the resolution of this parameter plane is $512 \times 512 = 262144$ with ranges of $p_A \in [0, 2]$ bar (linear scale) and $f \in [50, 1000]$ kHz (logarithmic scale). The bubble size is described by the equilibrium bubble radius R_E , that is the radius of the unexcited bubble. In our simulations we examined 7 different sizes with the values of $R_E = 2, 4, 6, 8, 10, 12$ and $14 \mu\text{m}$. Thus, the total number of the investigated parameter combinations are 1835008. Every other parameters are fixed, their values are shown in Tab. 1.

Table 1: The parameters kept constant during the simulations. The references for some non-trivial values are indicated.

Name	Abbrev.	Value	Unit	Ref.
Liquid sound speed	c_L	1483	m/s	
Liquid density	ρ_L	998.2	kg/m ³	
Ambient pressure	P_∞	1	bar	
Ambient temperature	T_0	293.15	K	
Surface tension	σ	0.07257	N/m	
Liquid viscosity	μ_L	0.001	Pa·s	
Thermal constant	a'	0.827	-	[49]
Boltzmann-constant	k	$1.38 \cdot 10^{-23}$	J/K	
Accommodation coeff.	α_e	1	-	[51]
Avg. cross section	σ'	$0.4 \cdot 10^{-18}$	m ²	
Boundary layer constant	n	7	-	[62]
Accommodation coeff. for evap.	α_M	0.35	m/s	[63]
Gas constant of vapour	R_v	461.5	Pa	
Correction factor	Γ	1	-	[53, 54]
Saturated vapour pressure	p_v^*	2338.1	Pa	
Diffusion coeff.	D	$1.76 \cdot 10^{-9}$	m ² /s	
Henry-constant	K_B	$6.737 \cdot 10^9$	Pa	
Universal gas constant	R_g	8.3146	J/(mol·K)	
Avogadro-constant	N_A	$6.022 \cdot 10^{23}$	1/mol	

Table 2: The applied chemical reactions in the model [59–61]. The indices f and b refer to the forward and backward reactions, respectively. The unit of c is K, b is dimensionless and ΔH_f is in kJ/mol. The units of A is $\text{m}^3/(\text{mol}\cdot\text{s})$ for two-body reactions and $\text{m}^6/(\text{mol}^2\cdot\text{s})$ for three-body reactions (see Eq. (24) for details). M denotes third body that does not take part in the reaction itself.

Reaction	Forward			Backward			ΔH_f
	A_f	b_f	c_f	A_b	b_b	c_b	
$\text{H}_2\text{O}+\text{M}\rightarrow\text{OH}+\text{H}+\text{M}$	$1.96\cdot 10^{16}$	-1.62	59700	$2.25\cdot 10^{10}$	-2	0	508.82
$\text{O}_2+\text{M}\rightarrow\text{O}+\text{O}+\text{M}$	$1.58\cdot 10^{11}$	-0.5	59472	$6.17\cdot 10^3$	-0.5	0	505.4
$\text{H}_2\text{O}+\text{O}\rightarrow\text{OH}+\text{OH}$	$2.21\cdot 10^3$	1.4	8368	$2.1\cdot 10^2$	1.4	200	72.59
$\text{OH}+\text{H}\rightarrow\text{O}+\text{H}_2$	$2.64\cdot 10^{-2}$	2.65	2245	$5.08\cdot 10^{-2}$	2.67	3166	8.23
$\text{OH}+\text{M}\rightarrow\text{O}+\text{H}+\text{M}$	$4.72\cdot 10^6$	-1	0	$4.66\cdot 10^{11}$	-0.65	51200	436.23
$\text{H}_2\text{O}+\text{OH}\rightarrow\text{H}_2\text{O}_2+\text{H}$	$1.41\cdot 10^5$	0.66	12320	$4.82\cdot 10^7$	0	4000	64.32
$\text{HO}_2+\text{OH}\rightarrow\text{H}_2\text{O}_2+\text{O}$	$4.62\cdot 10^{-3}$	2.75	9277	9.55	2	2000	56.06
$\text{O}+\text{O}_2+\text{M}\rightarrow\text{O}_3+\text{M}$	4.1	0	-1057	$2.48\cdot 10^8$	0	11430	-109.27
$\text{OH}+\text{O}_2\rightarrow\text{O}_3+\text{H}$	4.4·10	1.44	38600	$2.3\cdot 10^5$	0.75	0	96.2
$\text{H}+\text{O}_3\rightarrow\text{O}+\text{HO}_2$	$9.0\cdot 10^6$	0.5	2010	0	0	0	135.65
$\text{OH}+\text{OH}+\text{M}\rightarrow\text{H}_2\text{O}_2+\text{M}$	$9.0\cdot 10^{-1}$	0.9	-3050	$1.2\cdot 10^{11}$	0	22900	217.89
$\text{HO}_2+\text{H}\rightarrow\text{OH}+\text{OH}$	$1.69\cdot 10^8$	0	440	$1.08\cdot 10^5$	0.61	18230	-162.26
$\text{HO}_2+\text{O}\rightarrow\text{OH}+\text{O}_2$	$1.81\cdot 10^7$	0	-200	$3.1\cdot 10^6$	0.26	26083	231.77
$\text{H}_2\text{O}+\text{HO}_2\rightarrow\text{H}_2\text{O}_2+\text{OH}$	$2.8\cdot 10^7$	0	16500	$1.0\cdot 10^7$	0	900	-128.62
$\text{O}_2+\text{O}_2\rightarrow\text{O}_3+\text{O}$	$1.2\cdot 10^7$	0	49800	$5.2\cdot 10^6$	0	2090	396.0
$\text{H}_2+\text{HO}_2\rightarrow\text{H}_2\text{O}_2+\text{H}$	$1.41\cdot 10^5$	0.66	12320	$4.82\cdot 10^7$	0	4000	64.32
$\text{O}+\text{OH}\rightarrow\text{H}+\text{O}_2$	$7.18\cdot 10^5$	0.36	-342	$1.92\cdot 10^8$	0	8270	69.17
$\text{OH}+\text{H}_2\rightarrow\text{H}+\text{H}_2\text{O}$	$2.18\cdot 10^2$	1.51	1726	$1.02\cdot 10^3$	1.51	9370	64.35
$\text{OH}+\text{HO}_2\rightarrow\text{H}_2\text{O}+\text{O}_2$	$1.45\cdot 10^{10}$	-1	0	$2.18\cdot 10^{10}$	-0.72	34813	-304.33
$\text{H}+\text{O}_2+\text{M}\rightarrow\text{HO}_2+\text{M}$	$2.0\cdot 10^3$	0	-500	$2.46\cdot 10^9$	0	24300	204.8
$\text{HO}_2+\text{H}\rightarrow\text{H}_2+\text{O}_2$	$6.63\cdot 10^7$	0	1070	$2.19\cdot 10^7$	0.28	28390	239.67
$\text{H}_2+\text{M}\rightarrow\text{H}+\text{H}+\text{M}$	$4.58\cdot 10^{13}$	-1.4	52500	$2.45\cdot 10^8$	-1.78	480	444.47

The initial conditions of the simulations are as follows. The bubble was always initiated from equilibrium state, thus $R(0) = R_E$ and $\dot{R}(0) = 0$. The corresponding equilibrium pressure is

$$p_E = P_\infty + \frac{2\sigma}{R_E}. \quad (30)$$

Similarly, the initial value for the pressure derivative in Eq. (29) is 0. In the beginning, we assumed only O_2 and vapour molecules inside, with the partial pressure of vapour being the saturated vapour pressure (p_v^*) at ambient temperature. This yields the partial pressure of O_2 to be $p_{\text{O}_2,0} = p_E - p_v^*$. Here, the index 0 denotes the initial value. Assuming ideal gas, the initial number for oxygen is

$$N_{\text{O}_2,0} = p_{\text{O}_2,0} \frac{V_0 N_A}{R_g T_0}, \quad (31)$$

and for vapour, it is

$$N_{\text{H}_2\text{O},0} = \frac{p_v^*}{p_{\text{O}_2,0}} N_{\text{O}_2,0}. \quad (32)$$

Finally, the energy that belongs to equilibrium state is

$$E_E = \frac{T_0}{N_A} \sum_i N_{i,0} c_{v_{i,0}} - \left(\frac{N_{t,0}}{N_A} \right)^2 \frac{a_0}{V_0}. \quad (33)$$

In the case of diffusion, we assumed that the liquid has only dissolved O_2 [64] and that the saturated O_2 concentration is 10% of the initial concentration inside the bubble. This yields

$$c_{0,O_2} = \frac{10^3 \rho_L N_A}{M_{O_2} K_B} \frac{N_{O_2,0}}{N_{t,0}} p_0 \cdot 0.1. \quad (34)$$

Concentration of every other chemical substances in the liquid is supposed to be 0.

Due to the high number of parameters and the complexity of our system, the numerical solving process can be extremely slow. In order to accomplish the above described tasks, we decided to employ High Performance Computing (HPC) by utilizing the massively parallel architecture of Graphic Processing Units (GPUs). They have high amount of computational performance and cheap prize relative to that of CPUs. GPUs have thousands of parallel computational units that can work simultaneously; thus, they are suitable for making high resolution parameter studies, which is the main goal here. However, the biggest difficulty of using GPUs is that the user needs deep knowledge of the hardware architecture in order to write an efficient code and fully utilise the processing units. For detailed description of GPU programming to solve large number of independent ODE systems, the reader is referred to publications [65, 66]. The calculations are performed on an NVIDIA GeForce GTX Titan Black graphics card having a peak double precision processing power of 1707 GFLOPS.

3. The chemical output and the collapse strength of a bubble

Initiating a given system from the aforementioned initial conditions and performing the simulation for a given time, one can obtain the time curves of the bubble radius, wall velocity, internal energy and molecule numbers of each chemical species.

Figure 3 shows the results at $f = 100$ kHz, $p_A = 1.5$ bar and at $R_E = 10 \mu\text{m}$ for the first 8 excitation cycles (the time axes are always in dimensionless form $\tau = t \cdot f$, thus $\tau = 1$ relates to one excitation period). On chart (A), the time curves of the bubble radius (blue line) and the temperature (red dotted line) are displayed. The first collapse stage starts slightly after $\tau = 1$, at around $\tau = 1.2$, where the temperature grows above 3000 K. This high temperature indicates the presence of the reactions that dissociate H_2O molecules. Their products also start to be produced (e.g. H_2O_2 , OH^- or H_2 , see the complete list of molecule numbers on chart (B)). In the expansion phase, the number of the vapour molecules tends to grow by two orders of magnitude as a result of high amount of net evaporation rate from the liquid due to the low internal pressure. In the collapse phase, most of the vapour molecules dissociate and the molecule numbers of the other products start to increase rapidly. It can be observed from chart (B) that in the first couple of collapses (around 3 or 4), the molecule numbers of the products grow gradually until they saturate at a given level. For instance, the molecule number of H_2 (light blue line) is 0, 10^3 , $7 \cdot 10^4$ and $2 \cdot 10^5$ during the first 4 acoustic cycles, respectively. After this initial transient phase, all molecule numbers — except vapour and H atom —

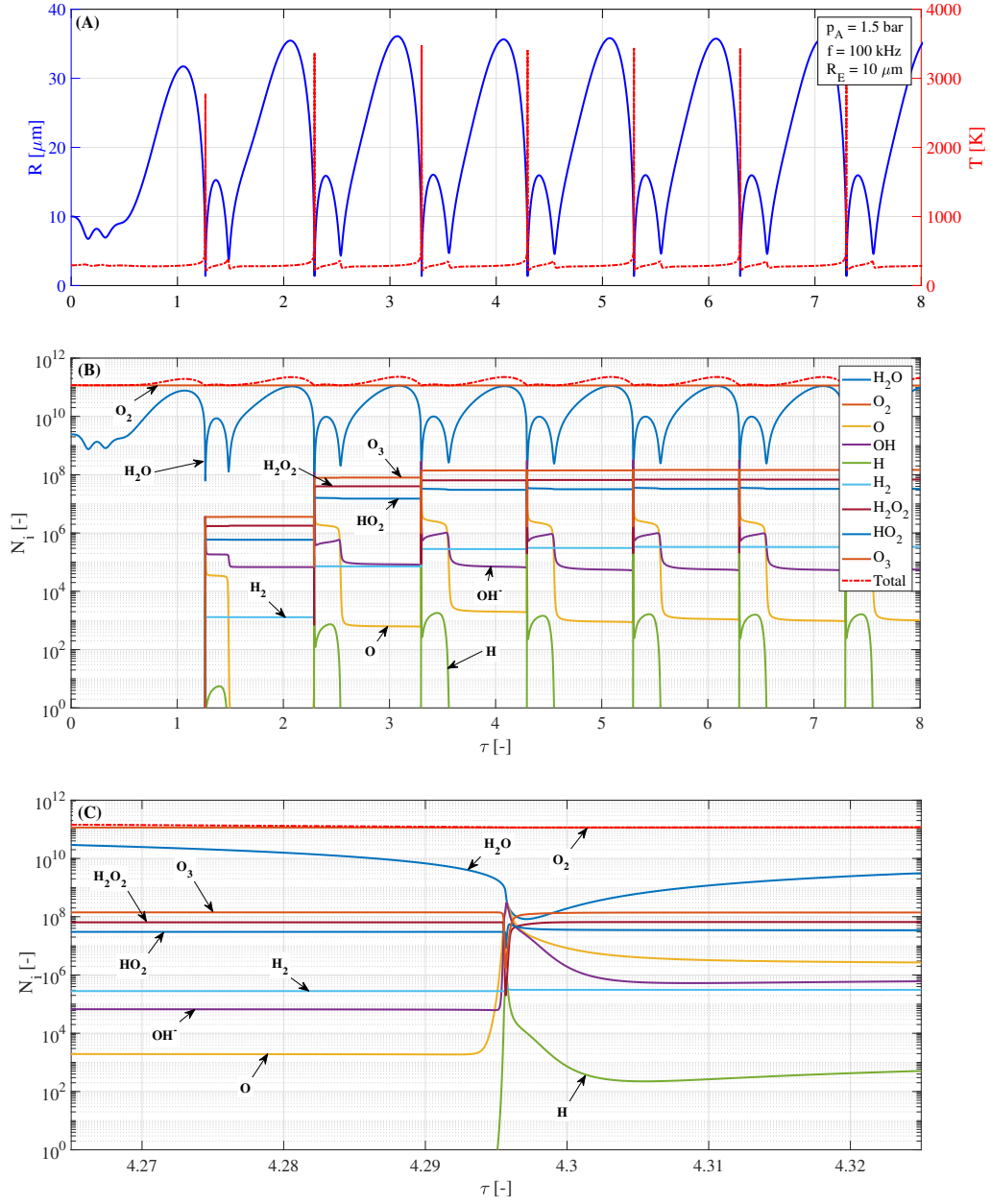


Figure 3: Bubble radius (blue line), temperature (red line) on chart (A) and molecule numbers on chart (B) with $p_A = 1.5$ bar, $f = 100$ kHz and $R_E = 10$ μm , in the first 8 acoustic cycles. A detailed view around the strong collapse is shown on chart (C).

186 stay constant in the expansion phase after the small afterbounces. This means that the
 187 net production rate becomes nearly zero as these products tend to dissociate on lower
 188 temperatures.

189 Although it is hard to identify from the time curves, diffusion into the liquid always
 190 presents, meaning that a certain amount of molecules are continuously leaving the bubble.
 191 Eventually, this could result in an overall decrease of the number of the molecules inside
 192 the bubble, but the evaporating vapour molecules always tend to “replace” this loss.
 193 Therefore, this procedure keeps diffusion of the chemical species into the liquid domain
 194 a rather constant process maintaining a dynamical equilibrium. It must be emphasized
 195 that the time scale of the diffusion process is orders of magnitudes higher than that of
 196 the characteristic time scale of the radial dynamics of the bubble. This phenomenon
 197 has a severe consequence. During the initial transient (see Fig. 3), the production of the
 198 chemical species is high, their concentration increases rapidly inside the bubble. However,
 199 after the initial transients (few cycles), the concentration of the species saturates at
 200 a certain level and the net production rate becomes nearly zero. The existence of a
 201 small positive production rate is due to the slowly diffusing molecules into the liquid
 202 domain. This is the reason why Mettin et al. [16] observed orders of magnitude higher
 203 sonochemical output when the acoustically driven bubble was spherically unstable. In
 204 their case, during the non-spherical collapse phase, the produced chemical components
 205 are released into the liquid also via a complex mixing procedure besides the slow diffusion
 206 process. Nevertheless, the present paper assumes spherical stability and focuses only on
 207 the released chemical species by diffusion.

208 On chart (C), an enlarged view around the fourth strong collapse of chart (B) is
 209 presented. It can be observed that the number of the molecules of vapour drops signifi-
 210 cantly here due to the high rate of dissociation of H_2O . Similarly, the production of e.g.
 211 H atom, OH^- or O atom increase drastically in the collapse phase. In the forthcoming,
 212 much slower, expansion phase, the amount of these types of molecules tend to decrease
 213 considerably keeping the dynamical equilibrium. However, some substances (e.g. O_3 ,
 214 HO_2 , H_2O_2) are reduced in the collapse phase but they are regained in the expansion
 215 phase. In general, Fig. 3 suggests that the production of chemical species are gener-
 216 ally caused by the dissociation of H_2O ; the O_2 molecules are mainly stay at a constant
 217 amount. This implies that it is the amount of vapour that influences chemical output
 218 rather than the initial gas content (pure oxygen here).

In order to connect the chemical activity of the bubble to its radial dynamics, proper
 characterisation of the collapse strength of a bubble is required. In the literature, the
 strength of the collapse is usually quantified by the following formulae [67–69]:

$$CS_1 = \frac{R_{max} - R_E}{R_E} \quad (35)$$

$$CS_2 = \frac{R_{max}}{R_{min}} \quad (36)$$

$$CS_3 = \frac{R_{max}^3}{t_c}, \quad (37)$$

219 where CS refers to collapse strength, R_{max} is the maximum value of $R(t)$, R_{min} is the
 220 subsequent minimum value of $R(t)$ and t_c is the collapse time, which is the elapsed time

between the maximum and minimum values of $R(t)$. In the literature, CS_1 and CS_2 is referred to as relative expansion and compression ratio, respectively. Observe that all these three quantities are derived only from the $R(t)$ curve; therefore, they represent the dynamical nature of the system around the collapse phase. It is commonly accepted [43, 70] that the threshold for sonochemical activity is $CS_1 = 2$; however, the relationship between the chemical output and the magnitude of the above defined collapse strength is rarely investigated.

As we assume spherical stability, we define the chemical production as the number of diffusing molecules into the liquid during one acoustic cycle as an average of 10 cycles. It is written mathematically as

$$CP_i = \frac{1}{10} \int_0^{10T} A \left(\frac{dm}{dt} \right)_i dt, \quad (38)$$

where $T = 1/f$ is the period of the excitation, A stands for the surface area of the bubble and CP_i means the chemical production of component i . It is clear that chemical production can be constructed for all types of diffusing species. The higher the chemical production of certain substances, the better the sonochemical output of the stable bubble for a given application. It depends on the specific application which chemical product is useful in a process; that is, maximizing the chemical production of certain species is keen interest of sonochemistry.

It is emphasized here that in our calculations, every simulation is run up to 30 periods of excitation, hence the effects of aforementioned initial transients could be neglected. Consequently, chemical production is estimated for the last 10 acoustic cycles, and the necessary quantities for collapse strengths (R_{max} , R_{min} , t_c) are determined from the last cycle.

4. The global overview of the chemical output in the 3D parameter space

High resolution numerical simulations are performed on the excitation amplitude - excitation frequency ($p_A - f$) parameter plane at different bubble sizes R_E . The exact values of these control parameters are given in Tab.3. Again, the total number of the parameter combination is $512 \times 512 \times 7 = 1835008$. For the details of the simulation set up at every parameter set, see the detailed description of the previous section.

Table 3: The control parameters, their resolutions and their scales.

Parameter	Limits	Resolution	Scale
f [kHz]	50 - 1000	512	log
p_A [bar]	0 - 2	512	lin
R_E [μm]	2 - 14	7	lin

After each simulation, one can calculate the values of the different collapse strengths CS_i and the chemical production of every substance CP_i using Eqs.(35)-(38). The strategy is to create a series of high resolution bi-parametric plots over the $p_A - f$ parameter plane at different values of R_E . Fig.4 summarises the chemical productions of $\text{OH}^\cdot$ radical, H_2O_2 , H_2 and O_3 for two different sizes ($R_E = 6$ and $12 \mu\text{m}$). The frequency

and the chemical production are on logarithmic scale. The black lines denote several iso-lines of the relative expansion CS_1 . From all charts of Fig. 4, it can be concluded that chemical production rates grows quickly with increasing pressure amplitude p_A and with decreasing the frequency f for every substances. However, employing a constant pressure amplitude, some peaks in the chemical production can be found at several values of the frequency due to the harmonic resonances of the system [71–73] (for example on chart (B), at $p_A = 1.5$ bar, local maxima of chemical production can be noticed at $f \approx 75$ kHz and at $f \approx 138$ kHz, marked by red squares). It can be observed that lowering the bubble size makes these resonances denser, compare for instance charts (C) and (D). On chart (C), at $R_E = 6 \mu\text{m}$, there are around 6 resonances in contrast to chart (D) where there are only 3. It should be noted here that the trends of chemical production are qualitatively the same for every chemical species. Thus, from now on, we only discuss the case of the production of OH^- radicals as the other components behave in a similar way.

The most important aspect in Fig. 4 is that the iso-lines of CS_1 correlate quite well with the chemical production. This observation suggests that a clear relationship exists between the collapse strength and the chemical production. In order to reveal the nature of this dependence, at every parameter combination, the values of the collapse strength CS_i and the values of the corresponding chemical production CP_i are collected in a single diagram. Figure 5 demonstrates this condensed representation where the chemical production of OH^- radical is plotted as a function of the three different collapse strengths. It is apparent that, particularly on Figs. 5A and B (CS_1 and CS_2), the points related to a specific bubble size show a clear trend. Especially on smaller equilibrium radii (up to approx. $8 \mu\text{m}$), the dependence is clear. However, over $10 \mu\text{m}$, the points gradually start to exhibit scattered behaviour. Nevertheless, an obvious trend can still be visible. In the case of CS_3 , a clear trend between the collapse strength and the chemical output is hard to be recognised compared to those of CS_1 and CS_2 . Due to such a poor correlation, the further discussion of CS_3 is omitted from now on.

From Fig. 5, it can be stated that the incidence of the chemical processes is approximately between $CS_1 \approx 2 - 3$. However, the exact value is less clear for smaller bubbles. This confirms the typically accepted rule-of-thumb in the literature (e.g. [43]) that the chemical reactions occur approximately over the value of $CS_1 \approx 2$. Nevertheless, such a threshold value does not provide information about the actual reaction rates. Observe that at $R_E = 2 \mu\text{m}$, the chemical production of OH^- remains relatively low as it does not exceed $3 \cdot 10^7$ molecules/cycle with a value of 12 for CS_1 . In contrast, the highest chemical production occurs at around $R_E = 12 \mu\text{m}$ of bubble size with a collapse strength of only $CS_1 \approx 6$. This is due to the fact that a larger bubble can contain more molecules; that is, larger reactive volume means higher total production. Moreover, a larger bubble has obviously bigger surface, thereby increasing diffusion rate as well (see Eq. (38)). As a consequence, the collapse strength alone cannot determine the sonochemical production quantitatively, the effect of the bubble size has to be included in such a description. This is the main topic of the next section.

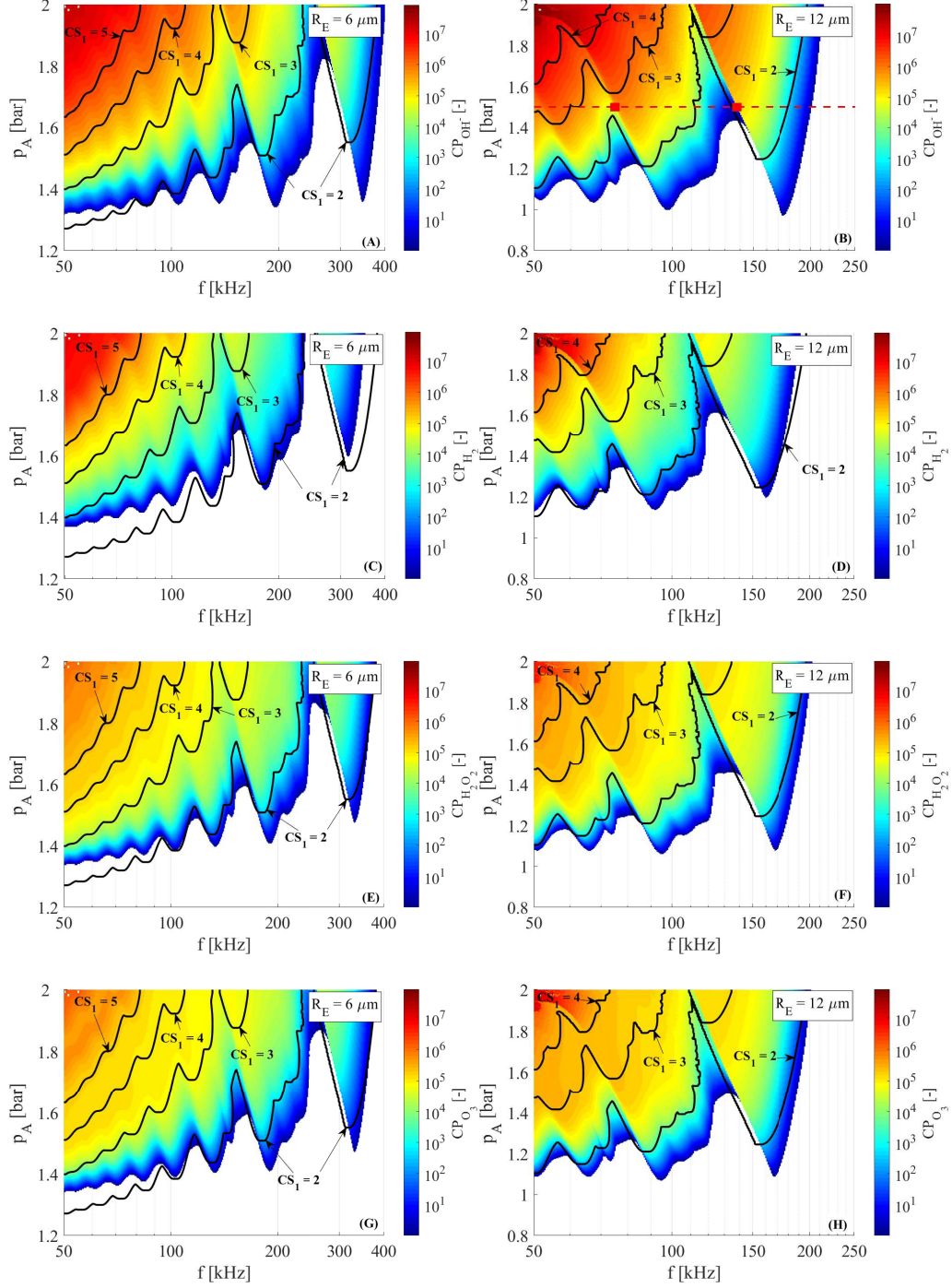


Figure 4: Bi-parametric plots of chemical productions of OH^- radical, H_2O_2 , H_2 and O_3 at two different bubble sizes ($R_E = 6$ and $12 \mu\text{m}$). The black lines are the iso-lines of relative expansion CS_1 for some specific values.

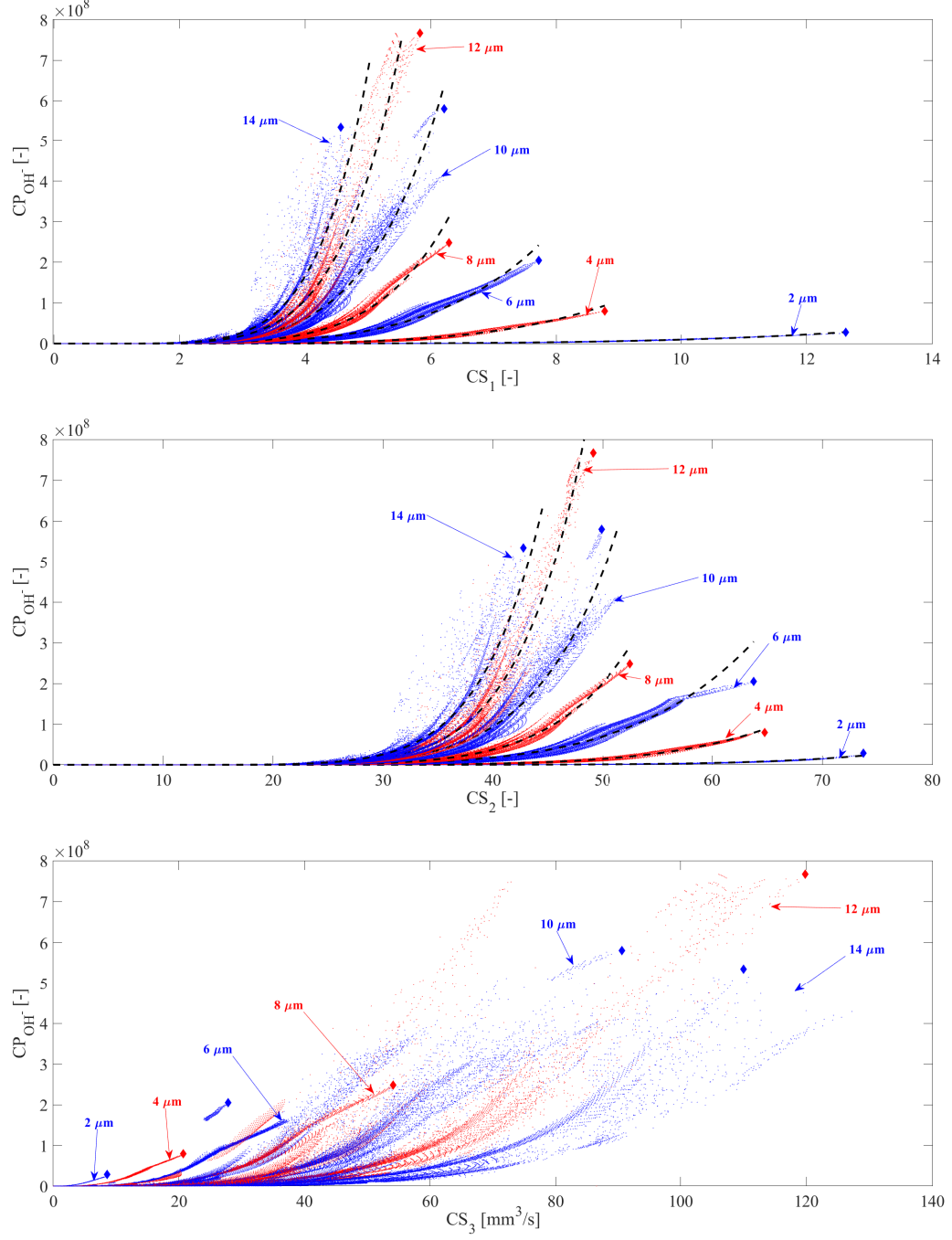


Figure 5: Chemical production of OH^- as a function of the three different collapse strengths. The values of R_E are denoted by the arrows. The black dashed lines belong to the fitted curves via Eq. (39).

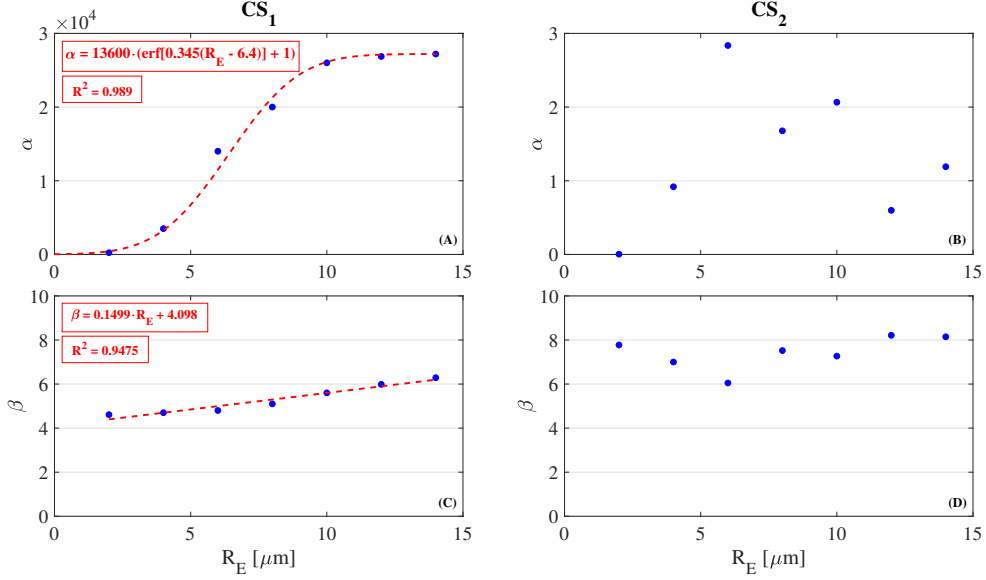


Figure 6: The values of parameters α and β for CS_1 and CS_2 , as a function of R_E . In the case of CS_1 , the fitted curves are also indicated together with the corresponding equations as well.

5. Mathematical description of the chemical production as a function of the collapse strength

As a final step, we aim to construct a mathematical formula between the collapse strength and the sonochemical production. With the least squares method, we fitted curves of the form

$$CP = \alpha \cdot CS_i^\beta \quad (39)$$

to the (CS_i, CP) points. Here α and β are the fitting parameters. The fitted curves are shown in Fig. 5 with the black dashed lines. The curves go through the (CS, CP) points sufficiently well (the R^2 values are above 0.9 for every values of R_E). The exact values of the fitting parameters for CS_1 and CS_2 as a function of R_E are shown in Fig. 6 (α and β are in the first and second rows, respectively). For the case of CS_2 , the values of α and β do not follow an exact trend. For CS_1 , on the other hand, it seems that the parameters have a clear dependence on the equilibrium radius. In the following, we focus only on CS_1 as it has the best visible correlation between collapse strength CS and the chemical production CP and it has the most clear trend in the fitted parameters as a function of the bubble size R_E .

From a viewpoint of applications and simplicity, it could be really useful if one can estimate the chemical activity and the production only from the dynamical properties of the bubble dynamics; for instance, only from CS_1 . Therefore, we aimed to construct a rather simple mathematical formula, that can help to predict the sonochemical production of a single bubble using only the collapse strength CS_1 without the simulation of

the complex chemical system. The fitted curves to the values of α and β as a function of R_E , which are also presented in Figs. 6A and C are

$$\alpha(R_E) = 13600 \cdot (\text{erf}[0.345(R_E - 6.4)] + 1) \quad (40)$$

and

$$\beta(R_E) = 0.1499 \cdot R_E + 4.098. \quad (41)$$

Here, the function $\text{erf}(x)$ stands for the error function, which is defined by [74]

$$\text{erf}(x) = \frac{2}{\pi} \int_0^x e^{-t^2} dt. \quad (42)$$

308 The R^2 values of the two fits are 0.989 and 0.9475 for $\alpha(R_E)$ and $\beta(R_E)$, respectively. It
 309 should be noted that R_E has to be in μm in Eqs. (40)) and (41). For the approximation of
 310 $\alpha(R_E)$, we intended to use a formula that tends to zero with decreasing R_E , and saturates
 311 with increasing R_E while it is still a reasonable good fit. We are aware of the fact
 312 that there is no specific physical meaning of the error function used here. Substituting
 313 the R_E values into Eqs. (40) and (41), then applying the results to approximate the
 314 chemical production via Eq. (39), one can get slightly different curves compared to the
 315 ones obtained by the individual fitting of α and β . This difference is shown in Fig. 7 where
 316 the black dashed lines are the curves fitted independently to the points corresponding to
 317 the different values of R_E , while the orange dashed lines is the ones obtained by using
 318 the approximated coefficients α and β calculated from Eqs. (40) and (41). The difference
 319 is not negligible; however, the orange lines also go through sufficiently well between the
 320 (CS-CP) points. Thus they are quite good approximations to estimate the chemical
 321 production of a single bubble.

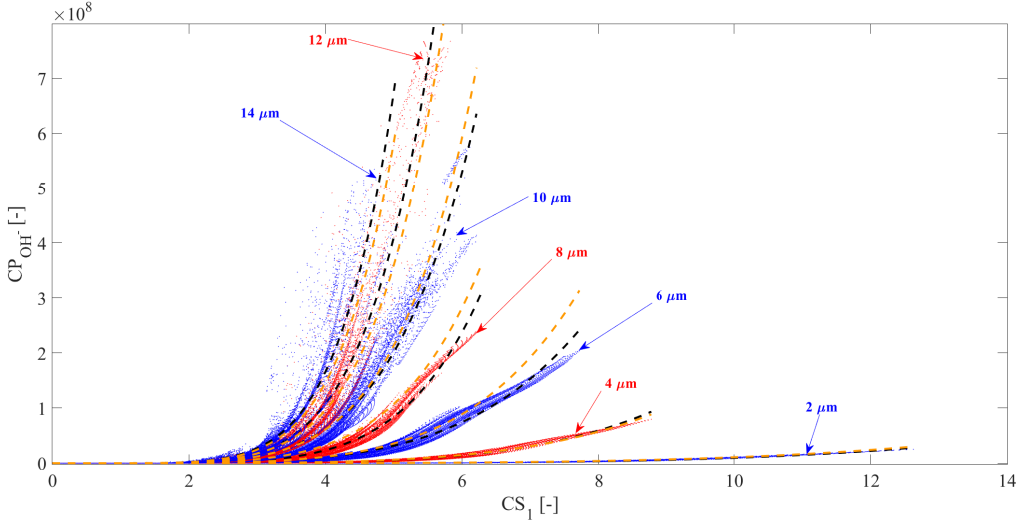


Figure 7: The difference between the two kind of fitted curves: individually fitted α and β (black curves); and approximated α and β from Eqs. (40) and (41) (yellow curves).

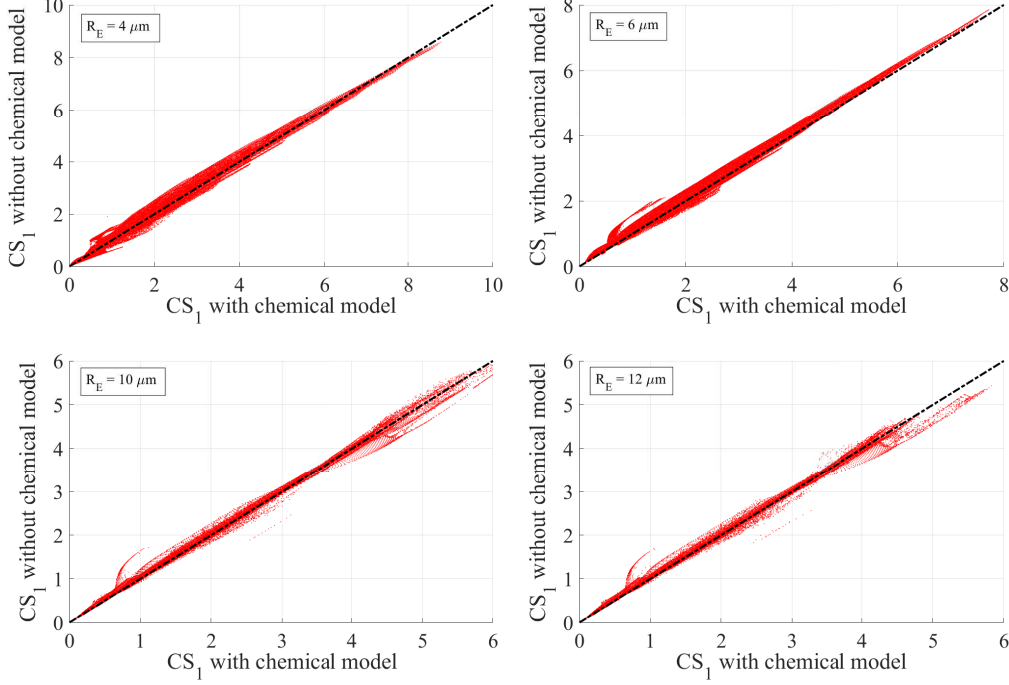


Figure 8: Values of CP_1 estimated from the Keller–Miksis model as a function of those of the full chemical model on the same parameter regime (see Table 3.). The bubble sizes are $R_E = 4, 6, 10$ and $12 \mu\text{m}$, respectively. The points are located fairly well around a straight line with the slope of 45 degrees (denoted with black lines).

By employing Eqs. (39)–(41), it becomes possible to estimate the chemical production of the system without explicitly calculating the chemical reactions. It is a remarkable result, because by solving the Keller–Miksis equation, the dynamics of a single bubble is relatively simple to calculate and it is the widely applied method in the investigation of bubble dynamics. This can be helpful during the optimisation of sonochemical reactors.

For validating this kind of application, it has to be proven that the values of collapse strength CS_1 are approximately the same with both the full-model (including chemical kinetics) and the sole Keller–Miksis equation (without chemical reactions). Therefore, we repeated the simulations on the parameter regime shown in Tab. 3 using only the Keller–Miksis equation (Eqs. (1)–(3)). The two type of collapse strengths are presented as a function of each other in Fig. 8 for four different bubble sizes. It can be clearly seen that the points are located around a straight line with a slope of 45 degrees. This implies that the values of the collapse strengths are approximately the same for both models. Thus, no correction is necessary during the estimation of the chemical yield from the bubble radius vs. time curves using the simple Keller–Miksis equation.

It should be emphasised, however, that the conclusions made above are based on many assumptions/simplifications: e.g. uniform bubble interior, spherical stability, simplified diffusion modelling and the approximations via curve fitting. Nevertheless, we believe

that our model can provide a good estimation for the chemical production; and to the best knowledge of the authors, it is the first study that successfully established a quantitative description between the collapse and the chemical production.

6. Summary and discussion

One of the most important aspects of sonochemical applications is the chemical production of some certain elements, such as oxidants, catalysts, etc. Maximizing the output of specific chemical species is the key to optimize the operation of sonochemical reactors. In the literature, several threshold values for different definitions of the collapse strength of a bubble exist to characterise the incidence of chemical activity. However, detailed parameter study has not been performed previously to correlate the exact chemical production and the collapse strength.

In the present study, chemical production of a harmonically excited bubble was investigated in the parameter space of the excitation pressure amplitude (p_A), driving frequency (f) and bubble size (R_E). The dynamics of the bubble wall was modelled by the Keller–Miksis equation and the internal pressure was calculated by van der Waals equation of state. Evaporation/condensation, heat transfer, diffusion and altogether 44 different chemical reactions for 9 different chemical species were taken into account. Chemical production was defined as the average number of molecules diffusing into the liquid during one acoustic cycle.

It was shown that the chemical production increases with increasing pressure amplitude and with decreasing frequency in the examined parameter domain. Accordingly, the maximum values of the chemical production observed at the biggest value of p_A and lowest value of f for each bubble size. These points are indicated with large diamonds in Fig. 5. This low frequency domain is often mentioned as the “giant response region” [1, 75], where the bubble grows drastically during the expansion phase. As a consequence, the speed of the collapse is extremely fast and the temperature tends to grow intensely high.

It was also found that the collapse strength alone cannot characterise the intensity of chemical processes, but there is an explicit dependence on the bubble size. Nevertheless, it was confirmed numerically that the widely used threshold $CS_1 \approx 2-3$ for sonochemical activity is reasonably valid. From Fig. 5, it turned out that despite of the high collapse strength, smaller bubbles have less chemical output compared to the larger ones at the same collapse strength. It has also been shown that there is an optimal bubble size for the chemical production at around $R_E = 12 \mu\text{m}$. However, the effect of spherical stability was not examined here. Bubbles tend to loose their spherical stability as their size increases [76–79]. It was proven numerically by Klapcsik and Hegedűs [80] that an increasing viscosity of the liquid domain (for instance by using glycerine) can stabilise the spherical shape. That is, the present finding can be valid by employing high viscosity fluids [81].

Finally, a mathematical formula was created that describes sufficiently well the sonochemical production as a function of relative expansion as a measure of the collapse strength. With the aid of these equations, it became possible to make estimations on chemical production qualitatively, based only on the dynamical features of bubbles.

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391 Appendix A. Dimensionless system

392 As already mentioned in Sec. 2, the Keller–Miksis-equation (1) and the energy equa-
393 tion (7) are solved in dimensionless form. Dimensionless time is denoted by τ and it is
394 defined as

$$\tau = \frac{t}{T} = tf. \quad (\text{A.1})$$

395 Dimensionless bubble radius and wall velocity are

$$x_1 = \frac{R}{R_E} \quad (\text{A.2})$$

and

$$x_2 = \dot{R} \frac{T}{R_E} = \frac{\dot{R}}{R_E f}. \quad (\text{A.3})$$

396 The derivatives with respect to dimensionless time are

$$\square' = \frac{d\square}{d\tau} = \frac{d\square}{dt} \frac{dt}{d\tau} = \frac{d\square}{dt} \frac{1}{f}, \quad (\text{A.4})$$

397 where prime denotes the derivative with respect to τ and $\square$ means any time-dependent
398 variable. Making a first order system from the second order Keller–Miksis-equation yields

$$x_1' = x_2, \quad (\text{A.5})$$

$$x_2' = \frac{N_{KM}}{D_{KM}}, \quad (\text{A.6})$$

399 where the nominator N_{KM} and denominator D_{KM} are

$$N_{KM} = \frac{p_L - p_\infty}{C_1 x_1} + \frac{(p - p_\infty)x_2}{C_2 x_1} + \frac{\frac{dp}{dt} - \dot{p}_\infty}{C_3} - (1 - C_4 x_2) \frac{3}{2} \frac{x_2^2}{x_1} \quad (\text{A.7})$$

and

$$D_{KM} = 1 - C_5 x_2 + \frac{C_6}{x_1}. \quad (\text{A.8})$$

400 Here the needed quantities with the dimensionless variables are

$$p_\infty = C_7 \sin(2\pi\tau), \quad (\text{A.9})$$

$$\dot{p}_\infty = C_8 \cos(2\pi\tau), \quad (\text{A.10})$$

$$p_L = p - \frac{C_9}{x_1} - C_{10} \frac{x_2}{x_1}, \quad (\text{A.11})$$

$$\left. \frac{dp}{dt} \right|_n = \frac{p_n - p_{n-1}}{\tau_n - \tau_{n-1}} C_{11}. \quad (\text{A.12})$$

401 The equation constants denoted by C are given in Tab. A.4. Here, the equilibrium energy (E_E) is calculated from the initial values described in Sec. 2.

Table A.4: The equation constants for the dimensionless system.

C_1	$\rho_L (R_E f)^2$
C_2	$\rho_L R_E f c_L$
C_3	$\rho_L R_E f^2 c_L$
C_4	$\frac{R_E f}{3c_L}$
C_5	$\frac{R_E f}{c_L}$
C_6	$\frac{4\mu_L}{c_L \rho_L R_E}$
C_7	p_A
C_8	$2\pi f p_A$
C_9	$\frac{2\sigma}{R_E}$
C_{10}	$4\mu_L f$
C_{11}	f
C_{12}	$\frac{1}{E_E} \frac{4\pi}{f} R_E^2$
C_{13}	$R_E f$
C_{14}	$\frac{R_E}{3}$
C_{15}	$\frac{4\pi}{f} R_E^2$
C_{16}	$\frac{R_E}{3} N_A$

402
403

With the dimensionless variables, the energy equation takes the form of

$$x_3' = C_{12}x_1^2 \left(-C_{13}px_2 + \sum_i \left(\frac{dm}{dt} \right)_i e_i + \kappa \frac{\partial T}{\partial r} \Big|_{r=R} + \dot{m}_{eva}e_{eva} - \dot{m}_{con}e_{con} + C_{14}x_1 \sum_{\gamma} (r_{\gamma,b} - r_{\gamma,f}) \Delta H_{\gamma,f} \right). \quad (\text{A.13})$$

404 The rate of molecule changes are

$$x_4' = C_{15}x_1^2 \left(\dot{m} + C_{16}x_1 \sum (r_{\text{H}_2\text{O},\text{prod}} - r_{\text{H}_2\text{O},\text{destr}}) \right) \quad (\text{A.14})$$

for water and

$$x_i' = C_{15}x_1^2 \left(\left(\frac{dm}{dt} \right)_i + C_{16}x_1 \sum (r_{i,\text{prod}} - r_{i,\text{destr}}) \right) \quad (\text{A.15})$$

405 for every other substance.

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