

Micro-explosion and autoignition of composite fuel/water droplets

D.V. Antonov¹, G.V. Kuznetsov¹, P.A. Strizhak¹, O. Rybdylova²,
S.S. Sazhin^{2*}

¹*National Research Tomsk Polytechnic University 30, Lenin Avenue, Tomsk, 634050, Russia*

²*Sir Harry Ricardo Laboratories, Advanced Engineering Centre, School of Computing, Engineering and Mathematics, University of Brighton, Brighton, BN2 4GJ, UK*

August 29, 2019

Abstract

The results of detailed experimental investigations of puffing, micro-explosions and autoignition of composite fuel/water droplets free falling in a high temperature air are presented. The measurements were performed for non-mixed and premixed Diesel fuel/water and rapeseed oil/water droplets. Air temperature was in the range 850-1100°C, initial droplet radii were in the range 0.62-1.34 mm, and relative volume fractions of fuel (Diesel fuel or rapeseed oil) were in the range 3-97%. It is shown that the time to micro-explosion decreases with increasing temperature, weakly depends on the volume fraction of fuel, and increases with increasing droplet sizes. The latter trend is shown to be consistent with the prediction of the previously developed simple model for droplet puffing/micro-explosion based on the assumption that the water component forms a spherical sub-droplet located in the centre of a fuel droplet. It is shown that the autoignition delay time for composite droplets, close to the time to micro-explosion for both fuels under consideration, is almost an order of magnitude less than that for pure fuel droplets.

Key words:

Micro-explosions, puffing, Diesel fuel, rapeseed oil, water, autoignition

*Corresponding author. E-mail: S.Sazhin@brighton.ac.uk

1. Introduction

It is well known that micro-explosions in water-in-fuel droplets, leading to rapid disintegration of these droplets, can potentially reduce nitrogen oxide particulate emission and improve engine performance [1, 2, 3, 4] (see the first paragraph in the Introduction in [5] for further details). The term ‘micro-explosions’ used in the literature refers to a wide range of phenomena with different underlying physics. For example, micro-explosions investigated in [6] were triggered by pockets of air formed during droplet collisions. Micro-explosions studied in [7] were produced by exothermic reactions between aluminum and a polymer and were used to prevent metal fuel particle agglomeration. Our study will be specifically focused on micro-explosions in immiscible liquids (e.g. Diesel fuel/water droplets). Following [8], a micro-explosion can be defined as ‘the bursting of the continuous phase (i.e. Diesel fuel or rapeseed oil) by the energy of evaporation of dispersed sub-droplets (i.e. water)’. Puffing (rapid increase in the droplet volume) usually precedes micro-explosions and the two processes are often modelled together [5].

A number of teams have presented the results of experimental studies of micro-explosions. In [9] composite droplets of water and sunflower oil emulsion were used. These droplets were representative of emulsions used in applications of biofuels. Micro-explosions of composite droplets of water and various oil products were studied in [10, 11]. The authors of [8] focused their research on water-in-oil emulsion droplets. Puffing and micro-explosions of water-in-Diesel fuel emulsion droplets exposed to high temperatures were studied in [12, 13]. The authors of [14] considered micro-explosions in kerosene, Diesel, gasoline, oil, turbine and automobile oil droplets with water sub-droplets within them (see [11, 12] for a more detailed overview of the earlier experimental studies).

A number of models for puffing and micro-explosions have been suggested. These models were either relatively simple [15, 16, 17] or very advanced, based on the direct numerical simulation of the processes involved [18, 19]. Although

30 the models suggested in [18, 19] are certainly very useful for understanding
the background of the mechanisms involved in the processes of puffing/micro-
explosions in droplets, the applicability of these models for practical engineering
applications is not obvious. Practitioners interested in these phenomena focus
mainly on such characteristics as the time to puffing/micro-explosion, but not
35 on the details of the processes inside the exploding droplet. In [5] a simple
model of the processes specifically focused on a crude approximation of this
time was developed. This model will be used in this paper for the analysis of
experimental data.

The research to be presented focuses primarily on experimental investiga-
40 tion of puffing and micro-explosions in free falling water-liquid fuel droplets
in a hot air. This investigation is different from those presented earlier when
various droplet supports were used (e.g. [8, 11]) or when high speed droplets
were injected from a nozzle (e.g. [13]). In contrast to most previous studies of
this phenomenon, in our case micro-explosion of droplets will be investigated
45 together with their autoignition. Some experimental results will be compared
with the predictions of a simplified model for puffing/micro-explosions earlier
suggested in [5].

2. Description of experiments

2.1. Composition of droplets

50 Three components (water, rapeseed oil and Diesel fuel) were used in our ex-
periments. Two types of droplets were considered: non-mixed two-component
(water and liquid fuel) droplets, produced during sequential preparation of the
components, and premixed droplets prepared using TWEEN 85 surfactant. In
the case of non-mixed two-component droplets, a single water sub-droplet was
55 located inside the fuel droplet. In the case of premixed droplets many small wa-
ter sub-droplets were distributed almost homogeneously inside the fuel droplet.

There are three reasons for our primary interest in non-mixed droplets.
Firstly, premixed emulsified fuels tend to segregate after long storage; in this

case, an interface is formed inside the droplet, and this droplet turns into a
60 non-mixed droplet. Secondly, the presence of an interface inside a non-mixed
droplet allows us to investigate all stages of the micro-explosion/autoignition
process and estimate the integral characteristics. Thirdly, for the preparation
of non-mixed droplets there is no need to use TWEEN 85 surfactant. The prop-
erties of the latter have a strong effect on the conditions and characteristics of
65 micro-explosions.

The combustible liquids were selected primarily due to their wide use as
currently available and prospective fuels. Diesel fuel is widely used for inter-
nal combustion engines because of its reliability and efficiency. Rapeseed oil
is widely used as a biofuel. Biodiesel produced from rapeseed oil is believed
70 to be ecologically clean. Typical concentrations of these combustible liquids in
prospective fuels are expected to be between 90% and 100% [20, 21]. Using the
results presented in numerous research papers (e.g. [8, 5, 14] and the references
therein), we expect that the addition of water to fuel (Diesel fuel or rapeseed
oil) will lead to the intensification of fuel droplet disintegration and autoignition
75 of the fuel vapour/air mixture. This occurs due to rapid increase in the liquid
surface area during the micro-explosions, which speeds up the evaporation pro-
cess and decreases the physical ignition delay that is controlled by the breakup,
heating and evaporation of droplets [22].

2.2. Experimental setup

80 A scheme of the setup used in our experiments is shown in Fig. 1. A muffle
furnace was used to heat air in the chamber up to about 1200°C. The distri-
bution of the air temperature inside the furnace was recorded using low inertia
thermocouples with errors of temperature measurement less than $\pm 3^\circ\text{C}$ and
errors of time measurement less than 0.1 s. The results inferred from thermo-
85 couple measurements were collected using National Instruments Thermocouple
Input Module NI 9219.

The temperature in the enclosure was measured using three thermocouples
as schematically shown in Fig. 1. Typical differences in the readings from these

thermocouples were in the range 6-9°C; the reading from the upper thermocou-
90 ple was always the maximal one while the reading from the lower thermocouple
was the minimal one. This gradient in temperature in the enclosure was ignored
in our analysis and the temperature in the enclosure was assumed equal to the
one inferred from the reading from the middle thermocouple. The temperature
of the furnace was maintained in the range 800-1200°C; the droplet radii were
95 in the range 0.62-1.34 mm.

The axis of the furnace was vertical which meant that contact was avoided
between the free falling droplets and the inner walls (the cases when this con-
tact took place were excluded from the analysis). The fragmentation of falling
droplets was recorded using a high speed video camera, capable of operating at
100 speeds of up to 10^5 frames per second. The parent and disintegrated droplets
were illuminated from below.

The experiments were performed in three stages. Firstly, a two-component
droplet of the required size was formed with the help of a weigher mounted on a
holder, as shown in Fig. 1. Secondly, a special device was mechanically brought
105 into contact with the holder to release the droplet leading to its free fall in a hot
air. After that, the high speed video camera mentioned above (here operating
at 5400 frames per second) recorded the droplet and its fragmentation products.
The droplet remained in the enclosure for less than 1 s.

2.3. Analysis of images

110 Typical recorded images of disintegrating droplets are presented in Fig. 2.
Fig. 2a shows non-mixed droplets, while Fig. 2b shows premixed droplets (emul-
sions). All images, including the ones in Fig. 2, were processed using Phantom
Camera Control software[23]. These images were used for the analysis of droplet
fragmentation times.

115 All images were scaled using the inner diameter of the furnace (50 mm).
Droplet diameters were scaled using the initial droplet diameters inferred from
their volumes, assuming that the droplets were initially spherical. Also, the
radii of the vapour bubbles (R_b) inside droplets were inferred from the analysis

of images. Finally the effective diameters of the aerosol clouds generated after
120 the micro-explosions were estimated as $D_c = (D_1 + D_2)/2$, where D_1 and D_2
are maximal and minimal diameters of cloud ellipsoids (see Fig. 3b). In Fig. 3b
the images of the autoignition of fuel vapour/air mixture are shown alongside
the images of fragmented droplets.

The number of child droplets and their sizes were calculated using the algo-
125 rithm described in [24]. Then the total area of child droplets was calculated and
compared with the initial area of the parent droplet. This allowed us to estimate
the change in the total area of the liquid surface during micro-explosions.

Altogether, more than 1000 experiments were performed, and about 460
images showing micro-explosions and the following autoignition were obtained.
130 Those images for which the droplet transit time inside the furnace was not
sufficient to collect the integral characteristics of micro-explosions and estimate
the number and sizes of droplet fragments after micro-explosion were not used
in our analysis. This approach allowed us to gather reliable statistics for the
experimental results in identical conditions.

135 **3. Results**

3.1. Times to micro-explosion

The plots of the observed times to micro-explosion versus the air temperature
for droplets with volume fractions of water 10% and volume fractions of Diesel
or rapeseed oil of 90% are shown in Fig. 4. In all cases, the droplet radii were
140 0.84 mm (which corresponds to volume $2.5 \mu\text{l}$). Air temperature varied between
 850°C and 1100°C . The reason for selecting this range of temperatures was that
at temperatures less than 850°C micro-explosions were not observed, while at
temperatures greater than 1100°C the time to micro-explosion was too short
to perform measurements (sometimes micro-explosions took place before the
145 droplets were released).

As can be seen in Fig. 4, the time to micro-explosion decreases with increas-
ing air temperature for both droplets, and this decrease is nonlinear. When

air temperature increased from 850°C to 1100°C, this time decreased by 55% for water/Diesel fuel droplets and only by 14% for rapeseed oil/water droplets. The reason for the difference in the behaviour of these droplets is not clear to us. The trend for the time to micro-explosion to decrease with increasing air temperature is related to an exponential decrease in the evaporation time of both droplet components with temperature.

The plots of the observed time to micro-explosion versus the volume fraction of fuel in two-component droplets are shown in Fig. 5. The droplet sizes were the same as in the case shown in Fig. 4. Air temperature was 950°C, and the fuel volume fractions varied from 3% to 97%. As one can see from Fig. 5, minimal times to micro-explosion were observed when the difference between the volume fractions of fuel and water was maximal, which corresponded to fuel volume fractions 3% and 97%. The behaviour of the times to micro-explosion for the intermediate values of fuel volume fractions turned out to be rather complex. As follows from our video recording of the process, the super-heating of the water/Diesel fuel interface takes place in very localised regions.

The results shown in Figs. 4 and 5 and the dependence of the time to micro-explosion on droplet radii, to be shown later, make it possible to control the times to micro-explosion in specific regions of the combustion chamber. This result is important for practical applications including those in internal combustion engine designs.

3.2. Formation and dynamics of child droplets

The time dependence of the normalised droplet diameter squared versus time for representative non-mixed and premixed droplets is shown in Fig. 6a. In all cases the initial droplet radii were 0.84 mm, but air temperatures varied from one experiment to another. The images of parent and child droplets at various stages of puffing and micro-explosion are shown in Figs. 6b-6f.

The dynamics of a non-mixed droplet with volume fractions of water 10% and Diesel fuel 90% are shown in Fig. 6b. Air temperature in this case was 950°C. Three stages of the dynamics of the parent droplet can be clearly seen

in this figure. The slow growth of droplet diameter during Stage I could be attributed to thermal swelling and the slow growth of vapour bubbles. The evaporation at this stage is expected to be weak. At Stage II, approximately 0.35 s after droplet release, the droplet diameter increases rapidly showing the development of the puffing process which can be clearly seen on the droplet image taken during this period. This puffing is followed by a rapid development of the micro-explosion and formation of many child droplets during Stage III.

The dynamics of a droplet similar to the one shown in Fig. 6b but for a volume fraction of water 10% and of rapeseed oil 90% are shown in Fig. 6c. In this case we can see not three but four stages in the development of the process. Initially, during Stage I, droplet diameter remains almost constant which shows the balance between swelling and evaporation processes. After this initial stage, a slow growth in the droplet can be seen during the remaining part of Stage I, as shown in Fig. 6b. The droplet dynamics in the following stage (Stage II) look rather different. In Fig. 6c we can clearly see a reduction, rather than growth, in droplet size. This is attributed to droplet breakup leading to a detachment of small sub-droplets from the droplet surface as illustrated by the images taken at approximately 0.35 s and 0.45 s. Stages III and IV show the rapid growth of droplets and their micro-explosion as in the cases of Stages II and III shown in Fig. 6b.

The dynamics of a droplet similar to the one shown in Fig. 6b but with volume fractions of water 97% and Diesel fuel 3% are shown in Fig. 6d. In this case, during Stage I, the droplet diameter remains almost constant which indicates the balance between droplet swelling and evaporation, as shown in Fig. 6c. At Stage II, a slow growth in droplet diameter, attributed to the formation of a vapour bubble, can be seen. This growth is followed by a rapid decrease in the droplet diameter until it reaches approximately 0.6 of the droplet initial diameter, which is attributed to bubble release leading to droplet fragmentation (not micro-explosion). Then we see another increase in the droplet diameter, attributed to the growth of another bubble, followed by a decrease in the diameter associated with the development of a micro-explosion.

The dynamics of a premixed (emulsion) droplet with a volume fraction of
210 water 10% and of rapeseed oil 90% are shown in Fig. 6e. Air temperature in
this case was 1050°C. As one can see from this figure, Stage I is characterised
by a visible decrease in droplet diameter due to evaporation without distorting
the shape of the droplet. During Stage II, droplet diameter remains almost
constant which indicates a balance between droplet evaporation and the growth
215 of internal bubbles. Finally, during Stage III, droplet diameter rapidly decreases
due to micro-explosion development and the formation of fine aerosols.

The dynamics of a droplet similar to the one shown in Fig. 6e but with
a volume fraction of water 10% and of Diesel fuel 90% are shown in Fig. 6f.
Air temperature was 1000°C. In this case, Stage I is characterised by a visible
220 decrease in droplet diameter due to evaporation that does not distort the shape
of the droplet. At Stages II and III one can see very large oscillations in droplet
diameter, attributed to the growth and collapse of bubbles. Eventually a micro-
explosion takes place (Stage IV).

The time evolution of the total area of the droplets and aerosol cloud, de-
225 veloped after the micro-explosion, is shown in Fig. 7. Droplets with a volume
fraction of water 10% and of Diesel fuel 90% (Fig. 7a) and volume fraction of
water 10% and of rapeseed oil 90% (Fig. 7b) were studied. Air temperature was
1100°C in the case shown in Fig. 7a and 980°C in the case shown in Fig. 7b.
In both cases the initial droplet radii were 0.84 mm. As one can see from this
230 figure, the total surface area of the aerosol cloud increases 100-200 times com-
pared with that for the parent droplet. In both cases shown in Fig. 7 this area
continued to grow after the time instant when a micro-explosion took place.

The time evolutions of the bubble radii for non-mixed droplets with a volume
fraction of water 10% and of rapeseed oil 90%, and volume fraction of water
235 97% and of rapeseed oil 3% are shown in Fig. 8. Air temperature in the first
case was 850°C, while in the second case it was 980°C. In both cases, the initial
droplet radii were 0.84 mm. As can be seen from this figure, bubbles formed
and collapsed during very short time periods in both cases. In the case of water
rich droplets, bubble formation and collapse took place later than in the case

240 of fuel rich droplets. We could not observe any bubbles in premixed (emulsion) droplets as these droplets are not transparent. The presence of these bubbles in the droplets, however, can be inferred from the results and images shown in Fig. 6f.

3.3. *Micro-explosion and autoignition*

245 In the experiments described above, micro-explosions were often accompanied by autoignition of the fuel vapour mixture. Since the total autoignition delay time is commonly considered to be the sum of the physical and chemical delay times [22], the micro-explosions, strongly affecting the physical delay, are expected to have a strong influence on this time. Hence, the importance of
250 studying both processes together. This is done in the current section.

In our experiments, the autoignition of the fuel vapour/air mixture in the presence of child droplets and aerosol clouds was observed both in air flow and during contact of these child droplets and clouds with the inner furnace walls. At the same time, autoignition was not observed during the contact of parent
255 droplets with these walls. In the latter case droplet rapid fragmentation was typically observed. In all cases, autoignition was seen only at temperatures greater than 950°C. It was observed that micro-explosions typically triggered autoignition in the whole domain, while puffing led to local autoignition. For flows with premixed droplets, autoignition was not observed.

260 Typical images showing the development of micro-explosions and autoignition of aerosol clouds, formed as a result of the micro-explosion of a droplet with a volume fraction of water 10% and of Diesel fuel 90%, are shown in Fig. 9. The droplet radius was 0.84 mm, while the air temperature was 1100°C. As one can see from this figure, the spread of the aerosol cloud follows the micro-explosion rather quickly (1.7 ms in the case shown in Fig. 9). Then after about 0.5 ms
265 the first signs of autoignition can be clearly seen. During the following 3.2 ms the flame spreads over a large region around the droplet.

Using images similar to those shown in Fig. 9 the autoignition delay times for composite droplets were obtained. These were compared with the autoignition

270 delay times for pure Diesel fuel and pure rapeseed oil droplets. In the latter
cases we could not use the setup shown in Fig. 1 as the droplet transfer time
was much less than the autoignition delay time. Instead, the experimental
methodology described in [25] was used. In this methodology, a furnace similar
to the one shown in Fig. 1 was used, but the axis of this furnace was not
275 vertical but horizontal. A droplet of liquid fuel was placed in the centre of
the furnace on a tiny holder of diameter 0.62 mm. This holder was introduced
into the furnace with the help of a special mechanism. Video recording of the
autoignition process was performed with the help of a high-speed video camera.

The plots of time to micro-explosion and autoignition delay times for Diesel
280 fuel based and rapeseed oil based droplets versus air temperature are shown in
Fig. 10. The following droplets were considered: droplets with a volume fraction
of water 10% and of Diesel fuel 90% and pure Diesel fuel droplets (Fig. 10a),
and droplets with a volume fraction of water 10% and of rapeseed oil 90% and
pure rapeseed oil droplets (Fig. 10b). The initial droplet radii in all cases were
285 0.84 mm.

As follows from Fig. 10a, the autoignition delay for both Diesel fuel and
Diesel fuel/water droplets decreases with temperature, but the autoignition de-
lay of Diesel fuel/water droplets is almost an order of magnitude shorter than
that of pure Diesel fuel droplets. Moreover, the autoignition delay time for
290 Diesel fuel/water droplets turned out to be almost the same as the time to
micro-explosion for these droplets. This is an expected result as the total au-
toignition delay is equal to the sum of the physical and chemical autoignition
delays. The former is reduced dramatically as a result of the micro-explosion.
The chemical ignition delay for Diesel fuel is expected to be very small (a frac-
295 tion of a millisecond, see [22]).

Essentially the same trends as shown in Fig. 10a can be seen in Fig. 10b
for rapeseed oil and rapeseed oil/water droplets. The main differences between
these two cases are: 1) the autoignition delay times for rapeseed oil and rapeseed
oil/water droplets are longer than those for Diesel fuel and Diesel fuel/water
300 droplets; 2) the difference between the autoignition delay times and the time to

micro-explosion is visibly larger for rapeseed oil/water than for Diesel fuel/water droplets. This might be attributed to the different thermophysical properties of the two fuels and different chemical ignition delays, although this problem has not been investigated so far to the best of our knowledge.

305 4. Modelling of the phenomena

As mentioned in the Introduction, the most advanced models of puffing and micro-explosion processes were suggested and developed in [18, 19]. These models were based on the Direct Numerical Simulation of the processes involved with tracking the interfaces between water and fuel, water and water vapour, 310 water vapour and fuel, and fuel and air. The main difficulty with the application of these models to the analysis of the experimental data, described earlier, is that they require the identification of the exact initial position of the water sub-droplet in the fuel droplet. In the case of a supported droplet this could be done using the methodology described in [14]. Unfortunately, it would be very 315 difficult to do this in our experiments with free falling droplets.

Among simplified models of the phenomenon we can single out the model described in [16]. This model is focused on the late stage of the development of puffing/micro-explosion when water inside fuel droplets has evaporated and the problem reduces to the interaction between water vapour and the surrounding 320 liquid fuel shell. The stability analysis of the system was performed assuming that this fuel shell is initially spherical. This allowed the author to predict the number and average diameter of the droplets, generated as a results of the shell rupture, as a function of the initial thickness of the shell. Thus, this model could potentially predict typical sizes of the droplets generated during micro- 325 explosions but not the times to puffing/micro-explosions.

One of the predictions of the model, which could be potentially compared with experimental data, is that the most probable diameter of the child droplets is approximately equal to 0.27 of the initial thickness of the fuel shell surrounding water vapour. We cannot infer the value of this thickness from our experiments,

but we can anticipate that it is proportional to the original thickness of this shell surrounding water. Considering the case when water in the composite droplet occupies 10% of the overall volume of the droplet and remembering the key assumption of the model described in [16] that the water sub-droplet is spherical and is located in the centre of a spherical composite droplet, the radius of the water sub-droplet can be estimated as:

$$R_w = 0.1^{1/3} R_d.$$

This allows us to estimate the original thickness of the fuel shell as

$$\delta_{fs} = R_d - R_w = \left(1 - 0.1^{1/3}\right) R_d \approx 0.536 R_d. \quad (1)$$

The plots of the observed numbers of child droplets N_{droplet} versus the radii of the child droplets for 3 droplets, 5 μl , 10 μl and 15 μl , are shown in Figure 11. These droplet volumes correspond to droplet radii 1.06 ± 0.05 mm, 1.33 ± 0.05 mm and 1.53 ± 0.05 mm, respectively. In all cases, ambient gas temperature was 400°C and water occupied 10% of the overall volume of the droplets.

In all three cases in this figure one can see clear maxima for N_{droplet} at certain values of the radii of child droplets R_{cd} . These maxima were observed at $R_{cd} = 0.095$ mm, $R_{cd} = 0.095$ mm and $R_{cd} = 0.085$ mm for droplets with volumes 5 μl , 10 μl and 15 μl , respectively. This indicates no visible increase in δ_{fs} with R_d , in contrast to the prediction of Equation (1).

There are several factors which might contribute to this discrepancy between the prediction of the model suggested by [16] and experimental data. The most obvious is the key assumption of this model that the water sub-droplet is spherical and is located in the centre of a spherical composite droplet. This assumption is not compatible with our observations.

Similarly to the model described in [16], a more recent model described in [5] is also based on the assumption that the fuel shell is spherically symmetric. In contrast to [16], the authors of [5] assumed that liquid water (not water vapour) is surrounded by the fuel shell. The focus of the model described in [5] is on the heating of both water and the fuel shell which makes it complementary to

the model described in [16].

One of the main advantages of the model developed in [5] is that it is able to give a crude prediction of the time to micro-explosion, which is an easily measurable parameter. This model is based on an application of the analytical solution to the heat transfer equation for the temperature inside a composite (fuel/water) droplet as a function of the distance from its centre and time. A composite droplet involves a small spherical water sub-droplet placed in the centre of a fuel (n-dodecane) droplet. The temperature at the surface of the droplet is assumed to be equal to the fuel boiling temperature or slightly below this temperature. Based on the above-mentioned analytical solution, the time instant when the temperature at the water/fuel interface reaches the water boiling temperature is found. This temperature indicates the start of the puffing process which is believed to quickly lead to a micro-explosion.

This model was applied to the analysis of the experiments described in [13], where the micro-explosion of relatively small (< 0.25 mm) droplets was studied. Our model predicted an increase in the time to puffing/micro-explosions when the fuel droplet diameters increased and water and fuel mass fractions were constant. This result agreed with experimental findings.

In what follows we attempt to apply the model described in [5] to the analysis of experimental data presented in the previous section. We approximated Diesel fuel with n-dodecane, but used a range of boiling temperatures typical of realistic Diesel fuels (489.47 K – 620 K). The values and approximations of thermophysical properties of rapeseed oil, taken from [26] and used in our analysis, are shown in Table 1.

Experimental data for air temperature 950°C and droplet initial room temperature 20°C were used in the modelling. Two types of composite non-mixed droplets were considered: 1) droplets with a volume fraction of water 10% and of Diesel fuel 90%; 2) droplets with a volume fraction of water 10% and of rapeseed oil 90%. The droplet surface temperature was taken equal to 477 K (boiling temperature of rapeseed oil) for droplets with rapeseed oil, and three values of droplet surface temperature (489.47 K, 513 K and 620 K) were used

Table 1: Rapeseed oil thermophysical properties and approximations.

Parameter	Value (T is in $^{\circ}\text{C}$)
Boiling temperature [$^{\circ}\text{C}$]	204
Density [kg/m^3]	$-0.689 T + 921.72$
Specific heat capacity [$\text{J}/(\text{kg } ^{\circ}\text{C})$]	$1.9 T + 1962$
Thermal conductivity [$\text{W}/(\text{m } ^{\circ}\text{C})$]	$0.0003 T + 0.1736$

for modelling droplets with Diesel fuel. 513 K is the boiling temperature of n-dodecane.

The experimentally observed times to micro-explosion, and those predicted
 380 by the model for both types of droplets for a range of initial droplet radii 0.6
 mm to 1.4 mm are compared in Fig. 12. As can be seen from this figure, for
 small droplets with initial radii close to 0.6 mm the predictions of the model for
 both types of droplets are close to the experimental results. For larger droplets,
 however, we can see clear deviations between the model predictions and ex-
 385 perimental data, although the predicted increase in this time with increase in
 droplet radii is compatible with the observed increase in this time. Also the
 order of magnitude of the predicted and observed times is similar. The mod-
 elled results for droplets containing rapeseed oil lie between those for droplets
 containing Diesel fuel with boiling temperatures of 489.47 K and 513 K.

The deviation between modelling and experimental results did not surprise
 390 us. The model is essentially based on the assumption that the water sub-droplet
 is located in the centre of the fuel droplet as in the case of the model described
 in [16]. At the same time our observations show that bubbles preceding micro-
 explosions develop away from the centre of the droplets, especially for larger
 395 droplets (cf. the images shown in Fig. 6). In this case, we would expect that
 the temperature at the surface of a water sub-droplet would reach the boiling
 point of water more quickly than predicted by the original model described in
 [5]. This is compatible with the results shown in Fig. 12.

To summarise, we can conclude that the model developed in [5] can predict

400 some correct trends regarding the time to micro-explosion observed experimen-
tally, and correct orders of magnitude for the values of this time. For relatively
small droplets (with initial radii close to 0.6 mm) the values of the time to micro-
explosion predicted by the model and observed experimentally are reasonably
close.

405 To the best of our knowledge, the modelling of the total autoignition delay
is always based on the assumption that it can be estimated as a sum of physical
and chemical autoignition delays. The physical autoignition delay takes into ac-
count droplet breakup, heating and evaporation, followed by the diffusion and
convection of fuel vapour into air. The chemical autoignition is controlled by
410 chemical reactions leading to explosion of the fuel vapour/air mixture. In the
case of the experiments described in the previous section the physical autoigni-
tion delay is controlled mainly by the time to micro-explosion.

The rigorous modelling of chemical autoignition, even for a single hydro-
carbon, would need to take into account hundreds of species and thousands of
415 chemical reactions. For example, the comprehensive kinetic mechanism for au-
toignition of cyclohexane comprises 499 species and 2323 reactions [27]. The
authors of [27] managed to replace this with a reduced mechanism, comprising
50 species involved in 143 reactions. Even this reduction, however, would not
have solved the problem of modelling the autoignition of realistic Diesel fuels
420 comprising more than a hundred components [28]. An alternative approach to
modelling the autoignition of Diesel fuel is based on the application of the Shell
model [29, 30]. The adaptation of this model to modelling the autoignition
of rapeseed oil has not been investigated to the best of our knowledge. Even
simpler models of Diesel spray autoignition based on a single Arrhenius-type
425 chemical reaction have been used in many papers, including [31].

In the case of Diesel fuel, the application of the Shell model was justified
by the fact that the chemical autoignition delay for this fuel is generally much
shorter than the physical autignition delay and this is confirmed by the results
shown in our Fig. 10. Using the latter figure we can take one step further and
430 ignore the contribution of chemical autoignition delay altogether. This delay is

expected to be comparable to the errors in the estimate of the time to micro-explosion.

5. Conclusions

Micro-explosion is shown to be the dominant mechanism of disintegration
435 of non-mixed fuel/water droplets in the range of air temperatures 850-1050°C,
while puffing led to droplet disintegration only in the case when the volume frac-
tion of fuel was small (3%). In the case of premixed droplets, micro-explosions
were observed only at temperatures greater than 1000°C. In this case, they were
preceded by droplet evaporation and a small increase in size of a parent droplet.
440 When the air temperature was increased from 850°C to 1050°C the time before
the start of disintegration via puffing of premixed droplets decreased by 1.1-
1.5 times. Time to puffing for these droplets decreased by 1.6-1.8 times when the
air temperature increased from 800°C to 1000°C. The times to micro-explosion
of these droplets decreased by 1.1-1.4 times when the air temperature was in-
445 creased from 1000°C to 1100°C. When the volume fraction of fuel increased
from 3% to 97%, the time to micro-explosion of premixed droplets decreased by
5-15%.

It was shown that the autoignition delay time for composite Diesel fuel/water
droplets is almost the same as the time to micro-explosion of these droplets.
450 This time is also much shorter than the autoignition delay time of pure Diesel
fuel droplets in similar conditions. The autoignition delay time for composite
rapeseed oil/water droplets was shown to be slightly longer than the time to
micro-explosion of these droplets. This time was seen to be almost an order of
magnitude less than the autoignition delay time of pure rapeseed oil droplets in
455 the same conditions.

The observed increase in the time to micro-explosion of composite droplets
of increasing droplet size is shown to be consistent with the prediction of the
previously developed model for droplet puffing/micro-explosion based on the
assumption that the water component forms a spherical sub-droplet located in

460 the centre of the fuel droplet. Actual values of this time were shown to be noticeably shorter than predicted by the model except for those of relatively small droplets with initial radii close to 0.6 mm.

Acknowledgements

465 Work on this paper was supported by the National Research Tomsk Polytechnic University (project VIU-ISHFVP-60/2019) (contributions by D.V. Antonov, G.V. Kuznetsov, P.A. Strizhak), and the EPSRC, UK (Grant Nos. EP/R012024/1 and EP/M002608/1) (contributions by O. Rybdylova and S.S. Sazhin).

References

- [1] A. M. Ithnin, H. Noge, H. A. Kadir, W. Jazair, An overview of utilizing water-in-diesel emulsion fuel in diesel engine and its potential research study, *Journal of the Energy Institute* 87 (4) (2014) 273 – 288. doi:<https://doi.org/10.1016/j.joei.2014.04.002>.
URL <http://www.sciencedirect.com/science/article/pii/S1743967114200717>
- 475 [2] O. A. Elsanusi, M. M. Roy, M. S. Sidhu, Experimental investigation on a diesel engine fueled by diesel-biodiesel blends and their emulsions at various engine operating conditions, *Applied Energy* 203 (2017) 582 – 593. doi:<https://doi.org/10.1016/j.apenergy.2017.06.052>.
URL <http://www.sciencedirect.com/science/article/pii/S0306261917308000>
- 480 [3] J. K. Mwangi, W.-J. Lee, Y.-C. Chang, C.-Y. Chen, L.-C. Wang, An overview: Energy saving and pollution reduction by using green fuel blends in diesel engines, *Applied Energy* 159 (2015) 214 – 236. doi:<https://doi.org/10.1016/j.apenergy.2015.08.084>.

- 485 URL <http://www.sciencedirect.com/science/article/pii/S030626191501020X>
- [4] B. K. Debnath, U. K. Saha, N. Sahoo, A comprehensive review on the application of emulsions as an alternative fuel for diesel engines, *Renewable and Sustainable Energy Reviews* 42 (2015) 196 – 211.
490 doi:<https://doi.org/10.1016/j.rser.2014.10.023>.
URL <http://www.sciencedirect.com/science/article/pii/S1364032114008405>
- [5] S. S. Sazhin, O. Rybdylova, C. Crua, M. Heikal, M. A. Ismael, Z. Nissar, A. R. B. Aziz, A simple model for puffing/micro-explosions in water-fuel emulsion droplets, *International Journal of Heat and Mass Transfer* 131 (2019) 815 – 821. doi:<https://doi.org/10.1016/j.ijheatmasstransfer.2018.11.065>.
495 URL <http://www.sciencedirect.com/science/article/pii/S0017931018338936>
- 500 [6] C. H. Wang, W. G. Hung, S. Y. Fu, W. C. Huang, C. K. Law, On the burning and microexplosion of collision-generated two-component droplets: miscible fuels, *Combustion and Flame* 134 (2003) 289–300.
- [7] M. A. Rubio, I. E. Gunduz, L. J. Groven, T. R. Sippel, C. W. Han, R. R. Unocic, V. Ortalan, S. F. Son, Microexplosions and
505 ignition dynamics in engineered aluminum/polymer fuel particles, *Combustion and Flame* 176 (2017) 162 – 171. doi:<https://doi.org/10.1016/j.combustflame.2016.10.008>.
URL <http://www.sciencedirect.com/science/article/pii/S0010218016303078>
- 510 [8] O. Moussa, D. Tarlet, P. Massoli, J. Bellettre, Parametric study of the micro-explosion occurrence of w/o emulsions, *International Journal of Thermal Sciences* 133 (2018) 90 – 97. doi:<https://doi.org/10.1016/j.ijthermalsci.2018.07.016>.

- URL <http://www.sciencedirect.com/science/article/pii/S1290072917317970>
515
- [9] D. Tarlet, E. Mura, C. Josset, J. Bellettre, C. Allouis, P. Massoli, Distribution of thermal energy of child-droplets issued from an optimal micro-explosion, *International Journal of Heat and Mass Transfer* 77 (2014) 1043 – 1054. doi:<https://doi.org/10.1016/j.ijheatmasstransfer.2014.06.054>.
520
- URL <http://www.sciencedirect.com/science/article/pii/S0017931014005262>
- [10] P. A. Strizhak, M. V. Piskunov, R. S. Volkov, J. C. Legros, Evaporation, boiling and explosive breakup of oil - water emulsion drops under intense radiant heating, *Chemical Engineering Research and Design* 127 (2017) 72 – 80. doi:<https://doi.org/10.1016/j.cherd.2017.09.008>.
525
- URL <http://www.sciencedirect.com/science/article/pii/S0263876217304616>
- [11] D. Antonov, J. Bellettre, D. Tarlet, P. Massoli, O. Vysokomornaya, M. Piskunov, Impact of holder materials on the heating and explosive breakup of two-component droplets, *Energies* 11 (12) (2018) 3307. doi: [10.3390/en11123307](https://doi.org/10.3390/en11123307).
530
- URL <https://www.mdpi.com/1996-1073/11/12/3307>
- [12] M. Y. Khan, Z. A. A. Karim, A. R. A. Aziz, M. R. Heikal, C. Crua, Puffing and microexplosion behavior of water in pure diesel emulsion droplets during leidenfrost effect, *Combustion Science and Technology* 189 (7) (2017) 1186–1197. arXiv:<https://doi.org/10.1080/00102202.2016.1275593>, doi:[10.1080/00102202.2016.1275593](https://doi.org/10.1080/00102202.2016.1275593).
535
- URL <https://doi.org/10.1080/00102202.2016.1275593>
- [13] M. A. Ismael, M. R. Heikal, A. R. A. Aziz, C. Crua, M. El-Adawy, Z. Nissar, M. B. Baharom, E. Z. Zainal A., Firmansyah, Investigation of puffing and micro-explosion of water-in-diesel emulsion spray using shadow imaging,
540

Energies 11 (9) (2018) 2281. doi:10.3390/en11092281.

URL <https://www.mdpi.com/1996-1073/11/9/2281>

- 545 [14] D. Antonov, R. Volkov, P. Strizhak, An explosive disintegration of heated fuel droplets with adding water, *Chemical Engineering Research and Design* 140 (2018) 292 – 307. doi:<https://doi.org/10.1016/j.cherd.2018.10.031>.

URL <http://www.sciencedirect.com/science/article/pii/S0263876218305616>

- [15] H. Watanabe, Y. Matsushita, H. Aoki, T. Miura, Numerical simulation of emulsified fuel spray combustion with puffing and micro-explosion, *Combustion and Flame* 157 (5) (2010) 839 – 852. doi:<https://doi.org/10.1016/j.combustflame.2010.01.013>.

555 URL <http://www.sciencedirect.com/science/article/pii/S0010218010000246>

- [16] O. G. Girin, Dynamics of emulsified fuel drop micro-explosions, *Atomization and Sprays* 27 (5) (2017) 407–422.

- [17] Y. Zhang, Y. Huang, R. Huang, S. Huang, Y. Ma, S. Xu, Z. Wang, A new puffing model for a droplet of butanol-hexadecane blends, *Applied Thermal Engineering* 133 (2018) 633 – 644. doi:<https://doi.org/10.1016/j.applthermaleng.2018.01.096>.

560 URL <http://www.sciencedirect.com/science/article/pii/S1359431117330405>

- [18] J. Shinjo, J. Xia, L. C. Ganippa, A. Megaritis, Physics of puffing and microexplosion of emulsion fuel droplets, *Physics of Fluids* 26 (10) (2014) 103302. arXiv:<https://doi.org/10.1063/1.4897918>, doi:10.1063/1.4897918.

URL <https://doi.org/10.1063/1.4897918>

- 570 [19] J. Shinjo, J. Xia, Combustion characteristics of a single decane/ethanol

emulsion droplet and a droplet group under puffing conditions, Proceedings of the Combustion Institute 36 (2) (2017) 2513 – 2521. doi:<https://doi.org/10.1016/j.proci.2016.06.191>.

URL <http://www.sciencedirect.com/science/article/pii/S154074891630253X>

575

[20] T. Kadota, H. Yamasaki, Recent advances in the combustion of water fuel emulsion, Progress in Energy and Combustion Science 28 (5) (2002) 385–404.

580

[21] H. Watanabe, T. Harada, Y. Matsushita, H. Aoki, T. Miura, The characteristics of puffing of the carbonated emulsified fuel, International Journal of Heat and Mass Transfer 52 (2009) 3676–3684.

[22] E. M. Sazhina, S. S. Sazhin, M. R. Heikal, V. I. Babushok, R. J. R. Johns, A detailed modelling of the spray ignition process in diesel engines, Combustion Science and Technology 160 (1) (2000) 317–344.

585

[23] <https://www.phantomhighspeed.com/resourcesandsupport/phantomresources/pccsoftware>.

[24] D. Antonov, O. Vysokomornaya, M. Piskunov, N. Shlegel, Analysis of statistical data on drop collisions in an aerosol flow during experiments, EPJ Web Conf. 196 (2019) 00013. doi:[10.1051/epjconf/201919600013](https://doi.org/10.1051/epjconf/201919600013).

URL <https://doi.org/10.1051/epjconf/201919600013>

590

[25] P. A. Strizhak, M. V. Piskunov, R. S. Volkov, J. C. Legros, Evaporation, boiling and explosive breakup of oilwater emulsion drops under intense radiant heating, Chemical Engineering Research and Design 127 (2017) 72 – 80. doi:<https://doi.org/10.1016/j.cherd.2017.09.008>.

URL <http://www.sciencedirect.com/science/article/pii/S0263876217304616>

595

[26] B. E. Poling, J. M. Prausnitz, J. O’Connell, The Properties of Gases and Liquids, McGraw-Hill, New York, 5th edition, 2001.

- [27] J. Griffiths, R. Piazzesi, E. M. Sazhina, S. S. Sazhin, P. A. Glaude, M. R. Heikal, CFD modelling of cyclohexane auto-ignition in an RCM, Fuel 96 (2012) 192–203.
600
- [28] S. S. Sazhin, Modelling of fuel droplet heating and evaporation: Recent results and unsolved problems, Fuel 196 (2017) 69 – 101. doi:<http://dx.doi.org/10.1016/j.fuel.2017.01.048>.
URL <http://www.sciencedirect.com/science/article/pii/S0016236117300583>
605
- [29] S. S. Sazhin, E. M. Sazhina, M. R. Heikal, C. Marooney, S. V. Mikhalovsky, The Shell autoignition model: a new mathematical formulation, Combustion and Flame 117 (1999) 529–540.
- [30] E. M. Sazhina, S. S. Sazhin, M. R. Heikal, The Shell autoignition model: application to gasoline and Diesel fuels, Fuel 78 (1999) 389–401.
610
- [31] S. S. Sazhin, E. Shchepakina, V. Sobolev, Order reduction in models of spray ignition and combustion, Combustion and Flame 187 (2018) 122 – 128. doi:<https://doi.org/10.1016/j.combustflame.2017.08.025>.
URL <http://www.sciencedirect.com/science/article/pii/S001021801730322X>
615

Figure Captions

Fig. 1 A schematic presentation of the setup used in the experiments.

620 **Fig. 2** Typical images showing bubble growth, puffing, and micro-explosions of droplets and autoignition of the fuel vapour/air mixture: (a) a non-mixed droplet with a volume fraction of water 10% and of rapeseed oil 90%; (b) a non-mixed droplet with a volume fraction of water 10% and of Diesel fuel 90%; in both cases the initial droplet radii were 0.84 mm and air temperature was 980°C.

625 **Fig. 3** Typical video images showing our approach to calculation of key parameters characterising the micro-explosion process: (a) cross-sectional area of the furnace; (b) dynamics of bubble growth inside a non-mixed droplet during the puffing process (the volume fraction of water 10% and volume fraction of rapeseed oil 90%, initial droplet radius 0.84 mm and air temperature 850°C);
630 (c) a non-mixed droplet and aerosol cloud growth (volume fraction of water 10% and volume fraction of Diesel fuel 90%, initial droplet radius 0.84 mm and air temperature 1050°C).

635 **Fig. 4** Average observed times to micro-explosion versus air temperatures for free falling droplets with initial radii 0.84 mm. The solid curve refers to a non-mixed droplet with a volume fraction of water 10% and of rapeseed oil 90%, while the dashed curve refers to a non-mixed droplet with a volume fraction of water 10% and of Diesel fuel 90%. In both cases error bars are shown.

640 **Fig. 5** Average observed times to micro-explosion versus volume fraction of fuel for free falling droplets with initial radii 0.84 mm and air temperature 950°C. The solid curve refers to water/rapeseed oil droplets while the dashed curve refers to water/Diesel fuel droplets. In both cases error bars are shown.

645

Fig. 6 (a) Time evolution of normalised diameters squared for the processes shown in Figs. 6b-6f (letters near the curves refer to the figure where the details of the process are shown; the initial droplet radii in all cases were 0.84 mm); (b) a non-mixed droplet with a volume fraction of water 10% and of Diesel fuel 90% in air with temperature 950°C; (c) a non-mixed droplet with a volume fraction of water 10% and of rapeseed oil 90% in air with temperature 950°C; (d) a non-mixed droplet with a volume fraction of water 97% and of rapeseed oil 3% in air with temperature 950°C; (e) a premixed droplet with a volume fraction of water 10% and of rapeseed oil 90% in air with temperature 1050°C; (f) a premixed droplet with a volume fraction of water 10% and of Diesel fuel 90% in air with temperature 1000°C. Various stages of the processes are described in the paper.

Fig. 7 Time evolution of the area of the liquid/air interface (droplets or aerosol clouds) during a non-mixed droplet micro-explosion: (a) a droplet with a volume fraction of water 10% and of Diesel fuel 90% in air with temperature 1100°C; (b) a droplet with a volume fraction of water 10% and of rapeseed oil 90% in air with temperature 980°C; in both cases the initial droplet radii were 0.84 mm. Inserts show the dependence of the normalised areas on the air temperature.

Fig. 8 Time evolution of bubble radii during a non-mixed droplet puffing/micro-explosion: (a) a droplet with a volume fraction of water 10% and of rapeseed oil 90% in air with temperature 850°C; (b) a droplet with a volume fraction of water 97% and of rapeseed oil 3% in air with temperature 980°C; in both cases the initial droplet radii were 0.84 mm.

Fig. 9 Typical video images showing the autoignition process of a non-mixed droplet with a volume fraction of water 10% and of Diesel fuel 90% in air with temperature 1100°C; the initial droplet radius was 0.84 mm.

Fig. 10 Autoignition delay times versus air temperature for pure fuel droplets (filled triangles) and non-mixed droplets with a volume fraction of water 10% and of fuel 90% (filled circles); times to micro-explosions of non-mixed droplets
680 with a volume fraction of water 10% and of fuel 90% (filled squares). Plots (a) and (b) refer to the cases when the fuels were Diesel fuel and rapeseed oil, respectively. In both cases the initial droplet radii were 0.84 mm.

Fig. 11 The distributions of the number of child droplets by their radii (R_{cd})
685 for three initial volumes of droplets, 5 μl , 10 μl and 15 μl , for a mixture of 90 vol% Diesel fuel and 10 vol% water. Ambient gas temperature was 400°C. Vertical bars show the values of R_{cd} at which the number of droplets was measured and the errors of these measurements.

Fig. 12 The plots of the time to micro-explosion inferred from experimental
690 data for droplets with a volume fraction of water 10% and of Diesel fuel 90% (triangles), and for droplets with a volume fraction of water 10% and of rapeseed oil 90% (squares). Average values of this time for droplets with Diesel fuel (rapeseed oil) points are connected with solid (dashed) lines. Predictions of the
695 model for droplets with a volume fraction of water 10% and of Diesel fuel 90%, and for surface temperatures 489.47 K, 513 K, and 620 K are shown by filled circles, circles, and filled diamonds, respectively. Predictions of the model for droplets with a volume fraction of water 10% and of rapeseed oil 90% for surface temperature 477 K are shown by crosses.

a

b

c

Fig. 3

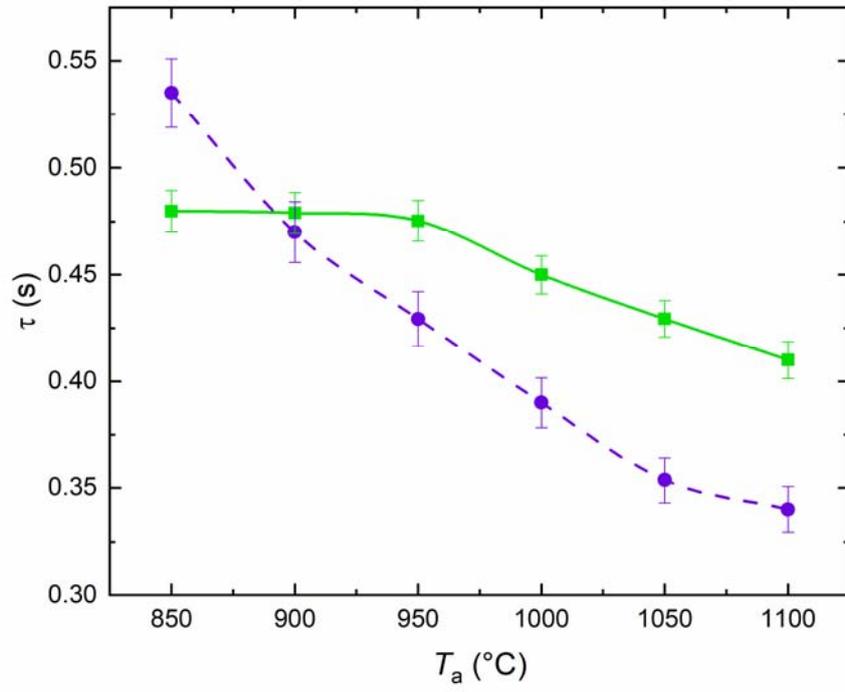


Fig. 4

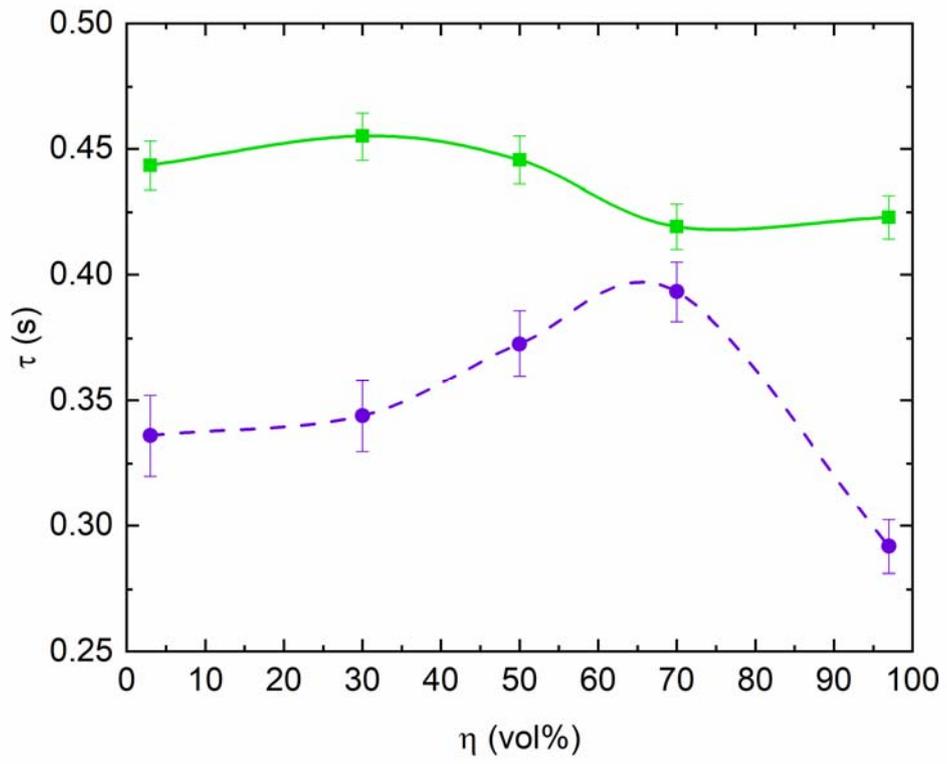


Fig. 5

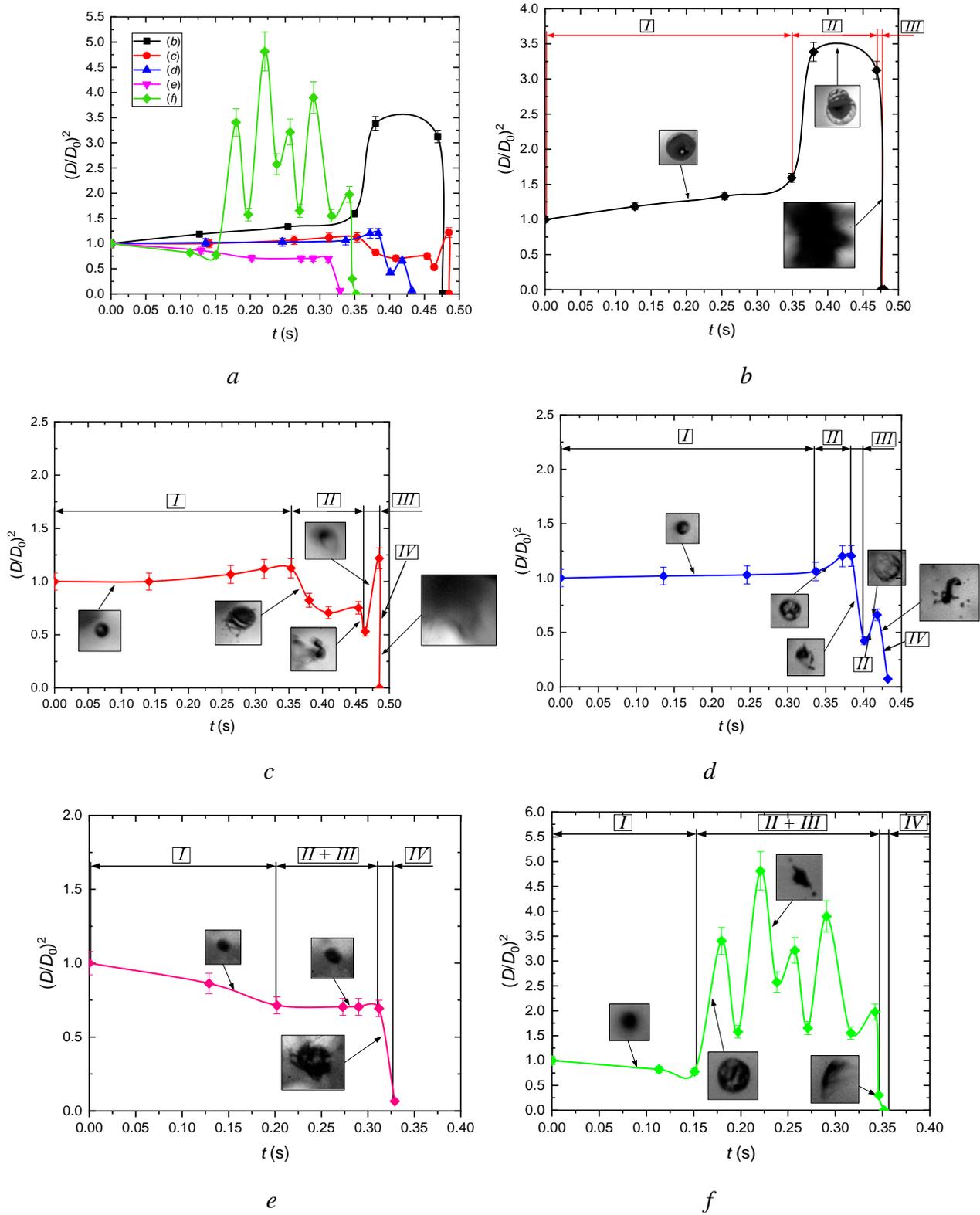
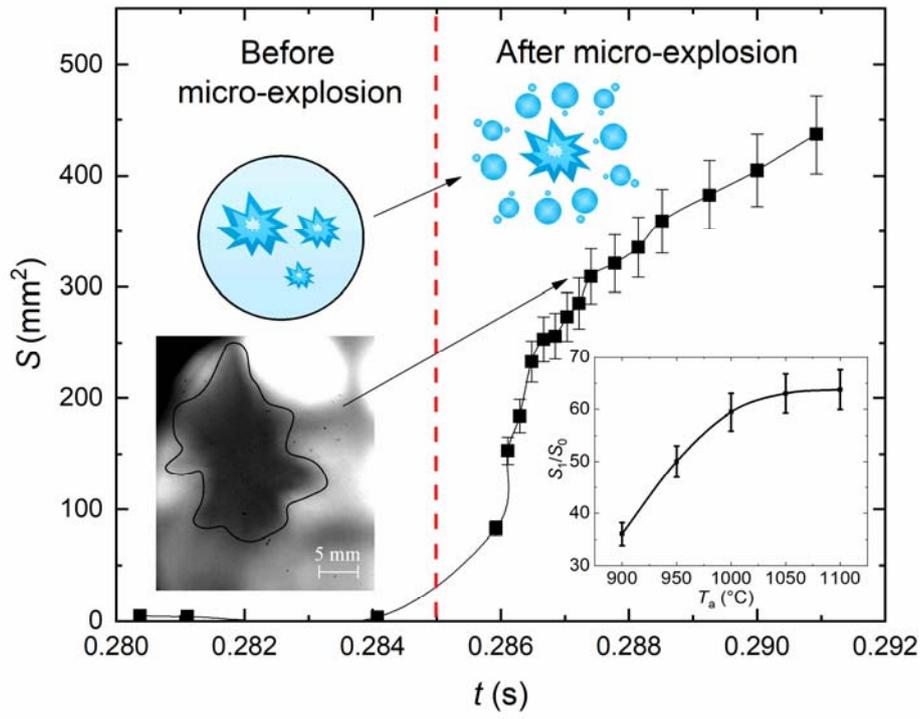
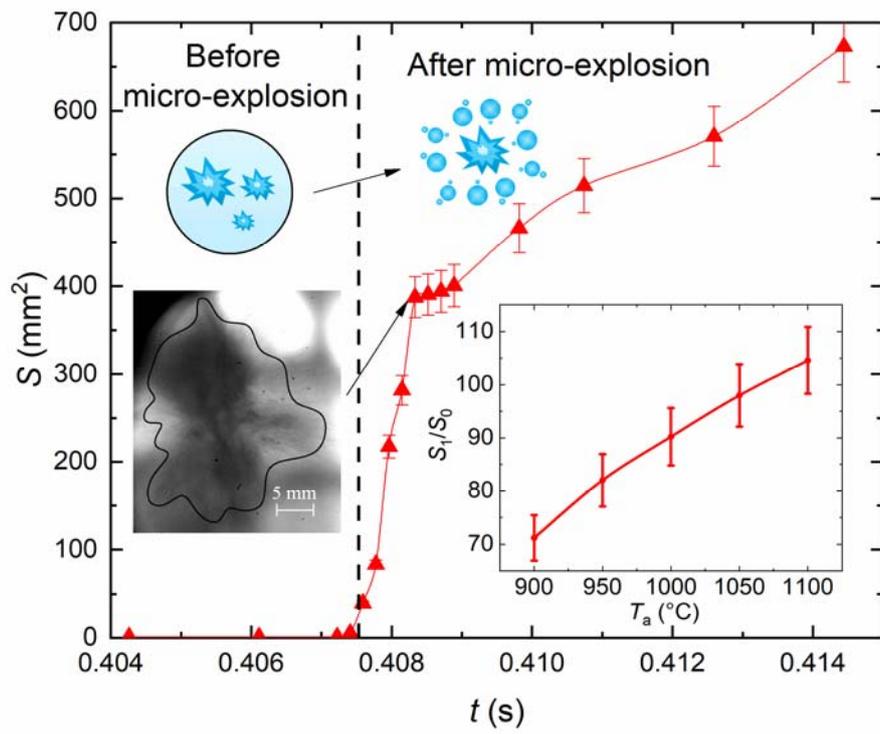


Fig. 6



a



b

Fig. 7

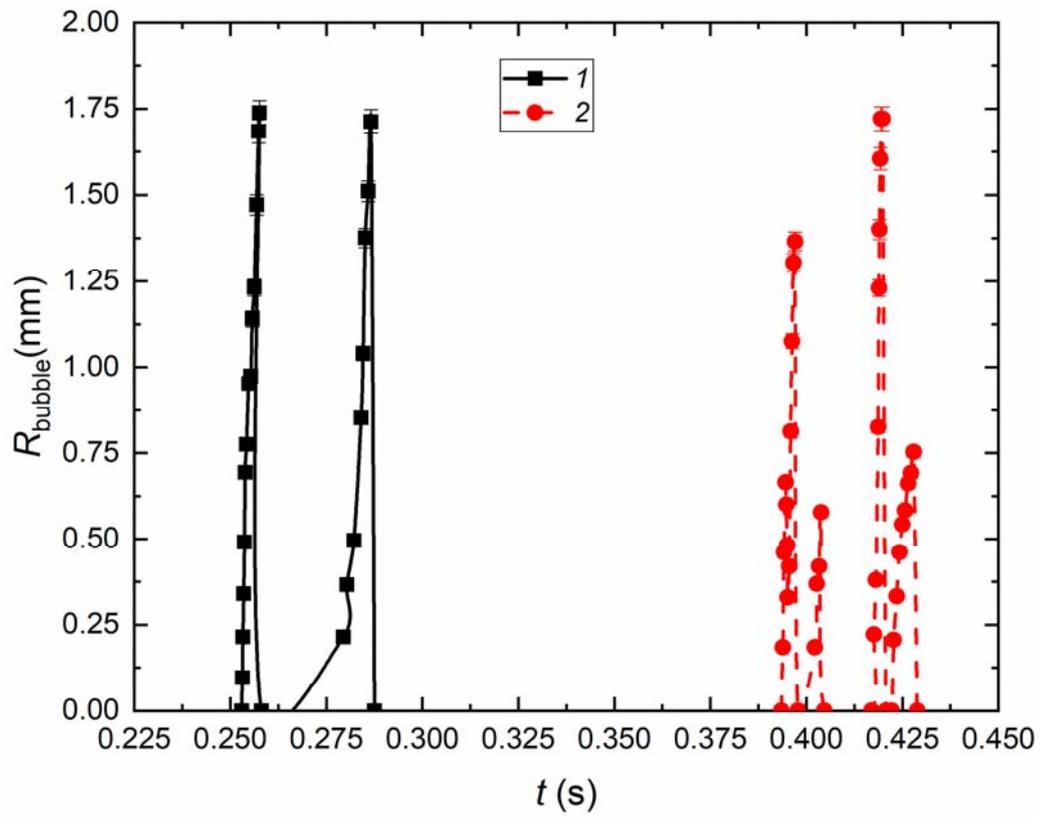


Fig. 8

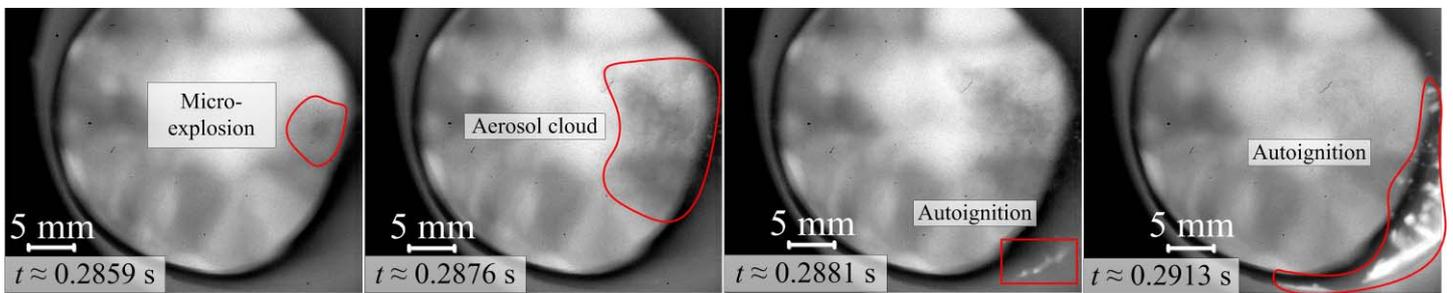
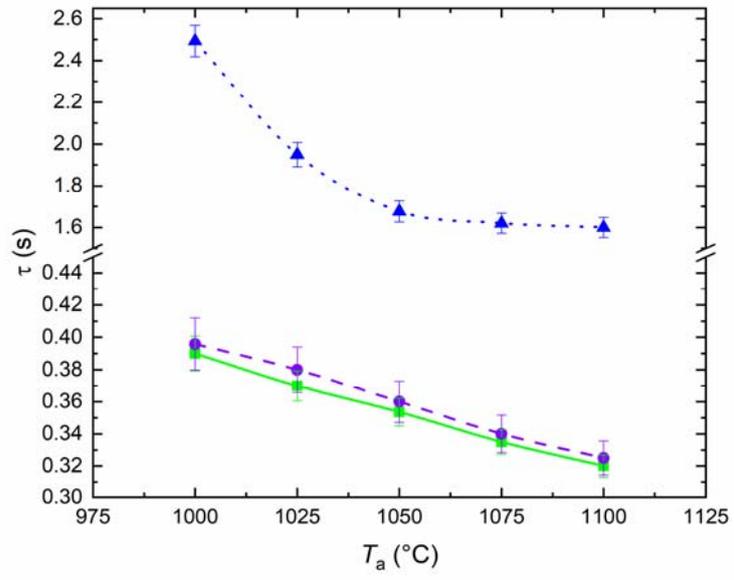
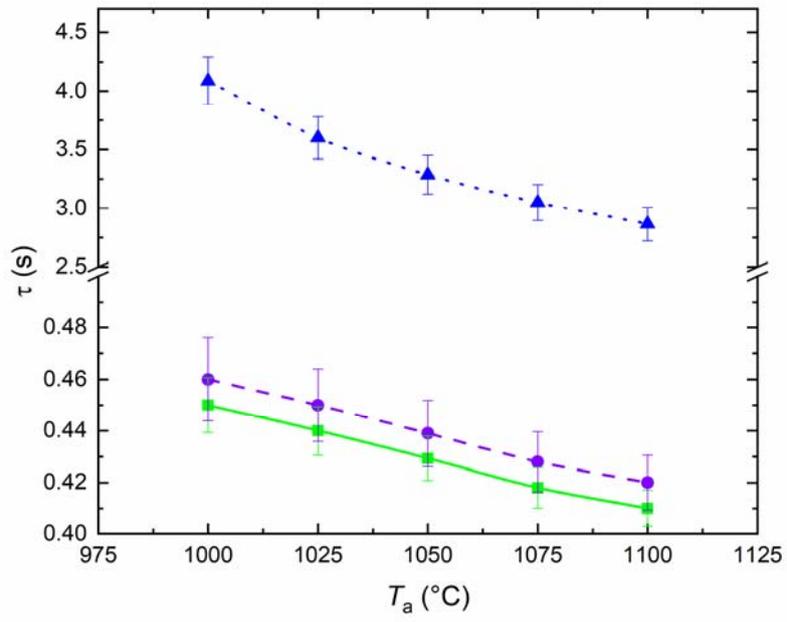


Fig. 9



a



b

Fig. 10

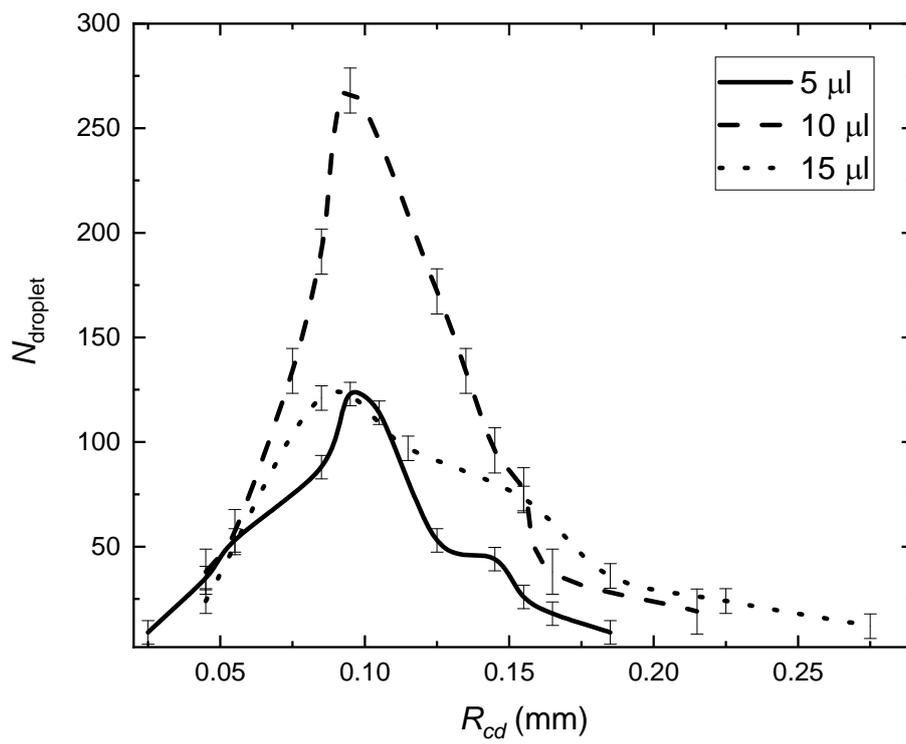


Fig. 11

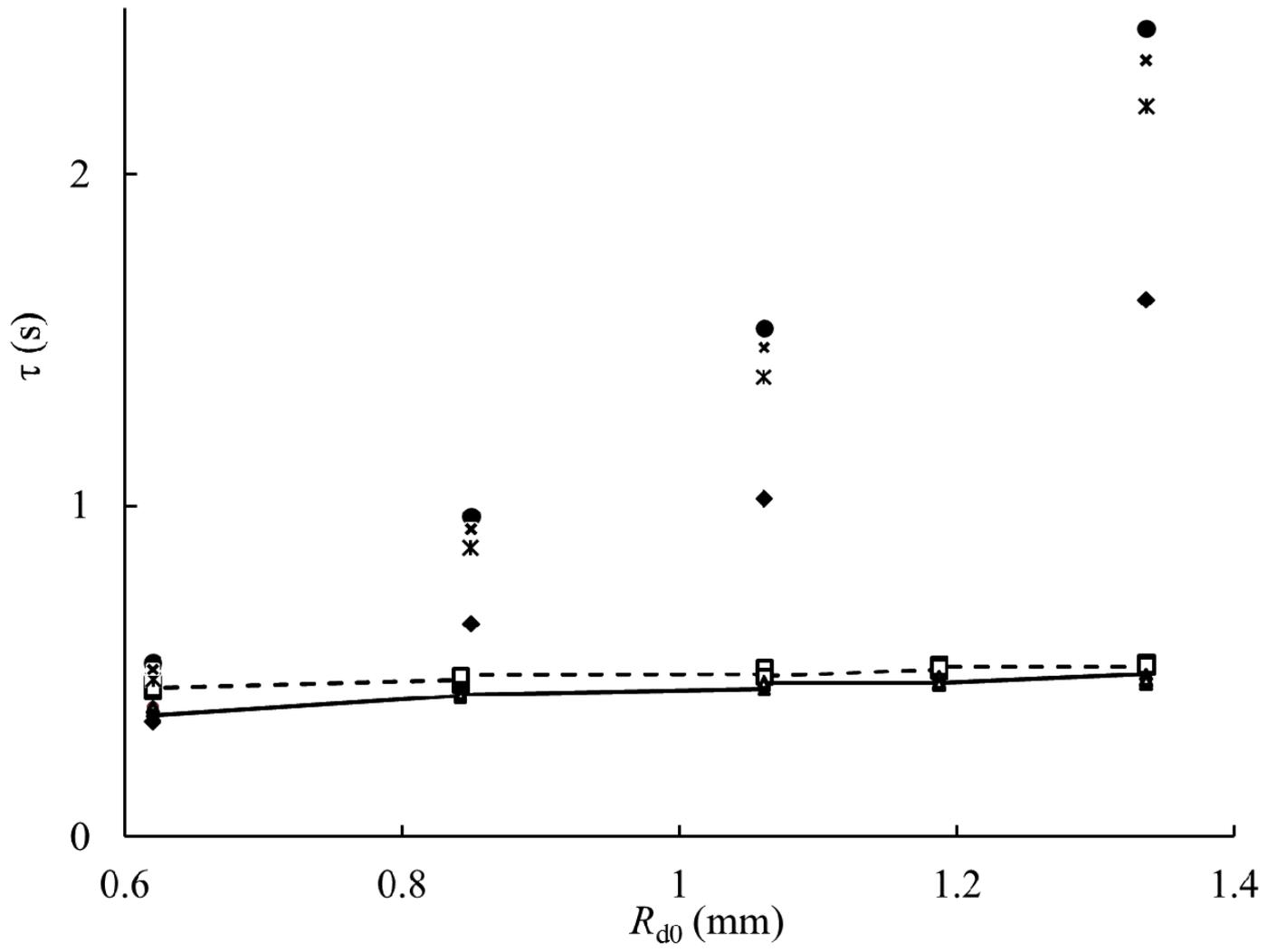


Fig. 12