

# ENHANCED MASS TRANSFER OF DEUTERIUM EXTRACTED FROM FALLING LIQUID Pb-17Li DROPLETS

Fumito Okino,<sup>a\*</sup> Kazuyuki Noborio,<sup>b</sup> Ryuta Kasada,<sup>b</sup> Satoshi Konishi.<sup>b</sup>

<sup>a</sup> Graduate School of Energy Science, Kyoto University, Uji, Kyoto 611-0011, Japan.

<sup>b</sup> Institute of Advanced Energy, Kyoto University, Uji, Kyoto 611-0011, Japan.

[fumito.okino@iae.kyoto-u.ac.jp](mailto:fumito.okino@iae.kyoto-u.ac.jp)

Release of deuterium from falling droplets of Pb-17Li in vacuum is experimentally studied. By comparing different diameter nozzle data each other, the effect of ambiguous solution is eliminated, and reliable result is attained. The amount of deuterium that is dissolved into Pb-17Li, followed by the release from the liquid droplets in vacuum, is measured with four different diameter nozzles ranging from 0.4 mm-1.0 mm under an initial velocity of 3.0 m/s and four temperatures between 375 °C and 450 °C. The resultant mass transport, represented by quasi-dispersion-coefficient is  $3.4 \times 10^{-7} \text{ [m}^2/\text{s]}$ , which is approximately two orders of magnitude faster than previous studies under static condition. It also revealed different temperature dependency. Cyclic deformation of the sphere shape is observed with a high speed movie camera. These results show the falling droplets of liquid Pb-17Li in vacuum follow the mass transfer mechanism under convection prior domain by self-excited oscillation. This result suggests that the tritium recovery method from a breeding liquid Pb-17Li blanket is viable when using multiple nozzles in vacuum for the extraction.

## I. INTRODUCTION

We have proposed the use of a high temperature liquid Pb-17Li blanket and a process for recovery of tritium from falling liquid droplets by mass transport in a vacuum-sieve-tray (VST) device, as shown in Fig. 1.

The extraction ratio of tritium from the liquid Pb-17Li is a critical design parameter for the VST device. Many previous studies were performed to identify the hydrogen isotopes transport property of liquid Pb-17Li, E. Mas de les Valls et al. (Ref. 1), F. Reiter (Ref. 2), and many other researchers. These previous measurements were performed under static condition, even though reported values were deviated approximately one order of magnitude. The main reason of deviation was assumed that measurements were affected by the ambiguous solubility condition. Considering that a falling droplet in VST is moving, the mass transport will be larger due to the dynamic mechanism. Although, deviation of the result will be larger, and results will be less reliable, if some countermeasure is not taken to eliminate the effects of solubility.

In this study, the mass transport of deuterium from falling liquid droplets of Pb-17Li is measured without the ambiguous solubility effects. Additionally the tritium extraction capability of the VST device is verified.

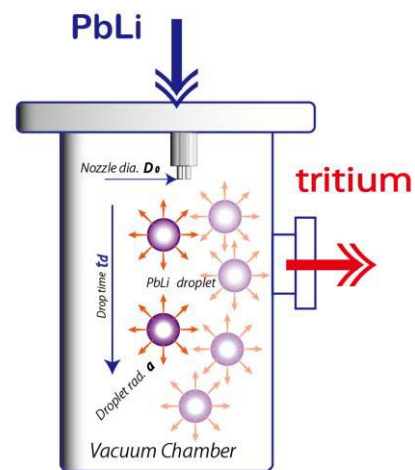


Fig. 1. Schematic of tritium extraction device (VST) Tritium is released from Pb-17Li droplets while falling in vacuum.

## II. EXPERIMENTAL

### II. A. Evaluation Criterion

Dynamic mass transport is considered, in general, a combination of diffusion and motion of fluid such as convection. Mass transfer coefficient and Sherwood number are commonly used to describe this transport phenomenon. But this falling droplet in VST, the experimental result was not well analyzed by previously developed criteria, due to the following reasons. Generally used analytical mass transfer coefficient is defined to describe the solute mass flux through the interface of a solvent as  $N_1 = k (c_{i1} - c_1)$ . The flux is assumed not so high, so the bulk concentration is regarded as constant. On this droplet case, the amount of mass transport is more than half of the total solute in a droplet, but not 100%, within a short dropping period of approximately 0.1 second. So bulk concentration is not

constant, it must be analyzed as unsteady-state mass transfer. A simple analytical mass transfer coefficient to describe the release phenomenon of droplet in VST, is inherently difficult, and simple coefficient is not prevalent.<sup>3</sup>

To evaluate this experimental result by simple criterion, in this study, the concept of axial-dispersion-coefficient is introduced.

It was reported by G. Taylor (Ref. 4) and modified by R. Aris (Ref. 5). A solute was introduced in a constant solvent flow within a tube over a very short period. The mass transport was theoretically studied by using the axial dispersion coefficient which has a same dimension with diffusivity coefficient of  $M^2/T$ . Solute in a flow was proved to follow the form of Fick's law equation. Different from the diffusion coefficient, this axial dispersion coefficient depends on the flow velocity and diffusion coefficient.<sup>6-7</sup> Due to the short release period, and a solvent flowing with solute, the mass transport of deuterium from Pb17Li falling droplets, resembles with Taylor's precondition. So the concept of axial dispersion coefficient is applied to correlate the experimental results of falling droplets in a VST. To avoid confusion, in this study, the quasi-dispersion-coefficient,  $D_{qua}$  which has the same function with the axial dispersion coefficient, is used.

As it follows the Fick's law, the many previous analyses developed for the "diffusion" can be correlated with this study. Due to the dropping is in vacuum, and within a short period, the overall mass transport is assumed as equal with that of at the interface, in this study.

**II.B. Experimental Setup and Measurements**

A schematic and photograph of the experimental setup are shown in Fig. 2-a. and Fig. 2-b. respectively. This setup consists of a reservoir, nozzles with various radii, sight windows, the lower chamber, a gas accumulation pump, a vacuum pump and measuring instruments. Due to regulations, deuterium gas is used instead of tritium for this experiment.

The dimensions of the components are described as follows: the upper reservoir is  $\phi 150 \text{ mm} \times 100 \text{ mm}$  height, the diameters of the four nozzles are 0.4 mm, 0.6 mm, 0.8 mm and 1.0 mm, and the lower chamber is  $\phi 150 \text{ mm} \times 500 \text{ mm}$  height with observation windows. Deuterium and argon gas are supplied for the release measurements. The nozzles are formed by drilling and finished by wire-cut for the Pb-17Li droplet. The deuterium gas is supplied via valve V1 to the Pb-17Li in upper chamber and the soluble pressure is controlled by the digital manometer-1.

To examine the mechanism of mass transport, falling droplets were observed with a high speed movie camera, VW-9000 Keyence at 4000 [frames /s].

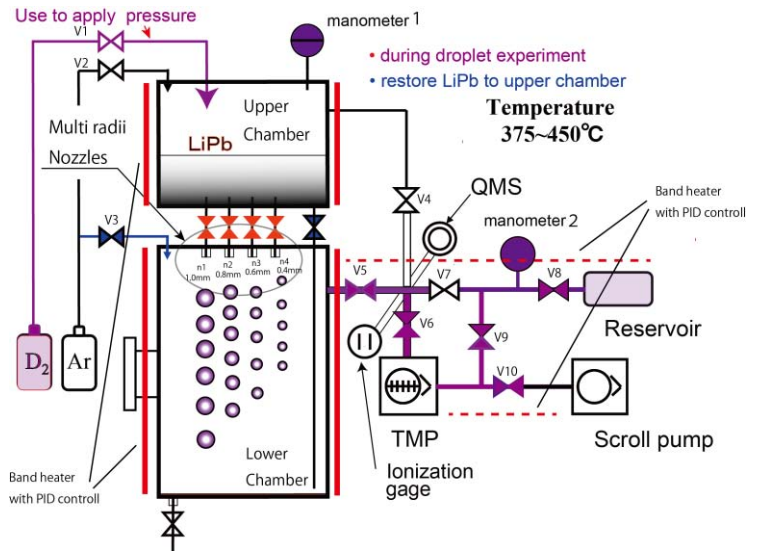


Fig. 2-a. Schematic of the experimental setup Deuterium gas is well absorbed into Pb17Li at the upper chamber, released from falling droplets at the lower chamber in vacuum, in a short period of 0.1 second, and collected at the reservoir. Four different size nozzles are equipped to eliminate the solubility effect.



Fig. 2-b. Photograph of the above mentioned experimental setup. Heat insulators are removed for picture taking.

**II.C. Amount of Released Deuterium**

*II.C.1. Precondition*

The deuterium released from chain of droplets in a vacuum was accumulated into the reservoir through TMP. The amount of released deuterium was calculated by

using the pressure reading on manometer-2, pre-measured reservoir capacity and temperature. The total volume of falling Pb-17Li droplets was calculated by using the pressure difference of upper chamber by manometer-1, with pre-measured capacity and temperature.

Heating was done with eight band type heater, each was independently temperature PID controlled. Experimental was performed under four different temperature conditions of 375 °C, 400 °C, 425 °C and 450 °C. Deviation of temperature after the equilibrium condition was within 2 °C, by data logger monitor reading. Accumulated reservoir area, between V5 and reservoir of Fig. 2-a, was always kept at 120 °C due to the temperature limitation of measurement devices.

The concentration of absorbed deuterium for the experiment was controlled by the policy that, at least within one set of experiments, which means one condition with four different nozzle drops, the concentration of deuterium must be same. Although absolute solubility was uncertain due to the effects of surfactants, progress of oxidization, etc., in longer periods. As shown in Fig. 4, the deviation of released gas ratio was rather small, so above policy was believed to be effective. Detail procedures of absorption were as follows. a) The absorption time was fixed as 150 minutes for each experiment, which is 1.5 times that of pre-examined test result. The convection caused by the side heating was believed to reduce absorption time. b) Concentration difference of absorbed deuterium at the upper, middle and lower portion of the liquid Pb17Li in upper chamber was negligible, by the comparison of each released gas. The convection by side heating was also believed to enhance well dilution.

### II.C.2 Released Deuterium Measurement

A measurement example is shown in Fig. 3. The horizontal axis is the duration time. The vertical axis is the pressure reading from the manometer-2. Both time and pressure are offset to zero at the start of dropping.

The pressure rise, which corresponds to the point < b > to < e > on Fig. 3 hereafter written as < b-e >, only occurred when falling droplets of Pb17Li were equilibrated with deuterium at upper chamber for the same long time. When the droplets were fallen without deuterium absorption (dry-run), no pressure rise occurred, and it showed just background level rise, same with < a-b >. By this pre-examination, the pressure rise < b-e > was confirmed by the deuterium absorption. It was also confirmed by QMS measurement.

The background gas while falling was regarded < b-c >, it is assumed as same rate with < a-b >.

The deuterium released from the deposition of Pb17Li at the bottom of lower chamber, corresponds < d-d' >, and continues to release while < d'-e > after falling stops. Number of droplets per one experiment condition is

about  $1 \times 10^5$ , so the release rate is constant, it must be plotted as the linear line on the Fig. 3. The overshoot deviation from this linear line, which corresponds to < d-d' > is considered as the release from the deposition.

The net amount of deuterium, released while a falling period, correspond < c-d >, by removing the release from background and deposition.

Then, the measured extracted deuterium was rescaled with the following equation to obtain the atomic fraction of extracted gas per liquid Pb-17Li for non-dimensional evaluation.

$$E(d_n(i)) = \frac{M^{de}(d_n(i))}{M^{LiPb}(d_n(i))} \quad (1)$$

The measurement results for the extraction ratio  $E(dn(i))$  as a function of the nozzle diameter  $dn(i)$  at 450 °C is shown in Fig. 4.

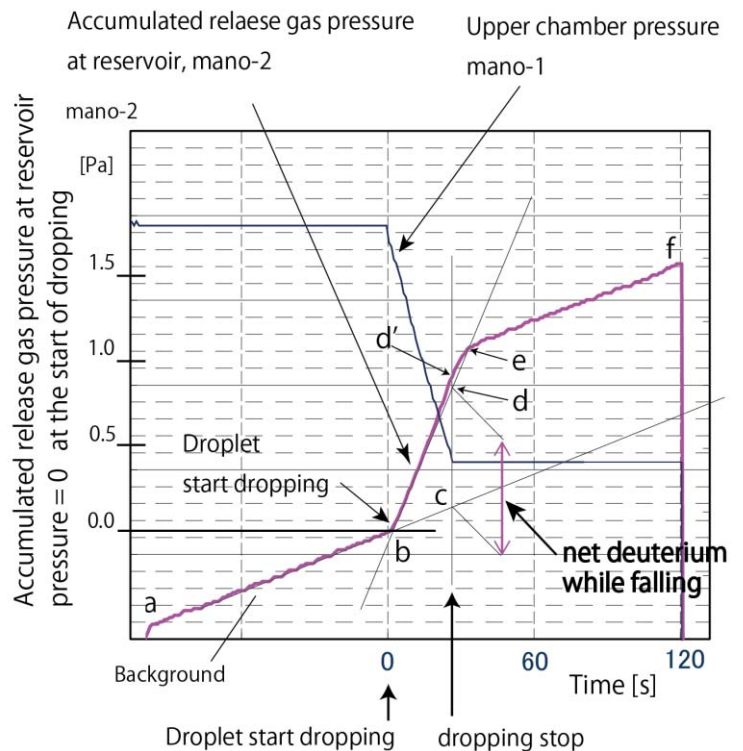


Fig. 3. An example of the accumulated released deuterium from chain of droplets, measured as the pressure evolution at the reservoir. The upper chamber pressure is also plotted to specify the start and stop of dropping time. < c-d > is the net deuterium release from falling droplets, while falling. < b-c > is background gas while dropping period. < d-d' > is released gas from deposited Pb-17Li liquid at the bottom of lower chamber. Gas release from deposited Pb-17Li continues until < e >.

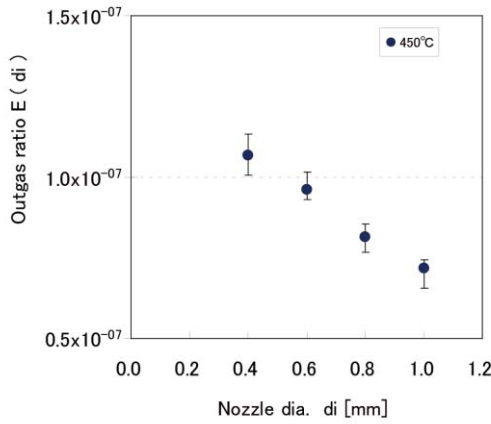


Fig. 4. Plots of normalized amount of released deuterium shown as atomic fraction, at 450 °C. The horizontal axis is the diameter of nozzles used for experiment.

### II.D. Survey of Quasi Dispersion Coefficient

The amount of deuterium released from a droplet is described as the product of solution and release processes, and the theoretical extraction ratio is described as equation (2) under constant temperature, soluble pressure and dropping time conditions.

$$S \cdot Th(D_{qua}, t_d, d_n(i)) \tag{2}$$

As shown in above equation, to identify the reliable quasi dispersion coefficient, without the influence of ambiguous solubility, at least two different nozzle diameter data are required. To eliminate solubility, the function *TTh* which is defined as equation (3), is introduced. By dividing the numerator and denominator of each different diameter nozzle data under same soluble condition, solubility is eliminated. By this processing, the quasi dispersion coefficient is an only unknown parameter as shown below.

$$TTh(D_{qua}, d_n(i), d_n(j)) = \frac{S \cdot Th(D_{qua}, t_d, d_n(i))}{S \cdot Th(D_{qua}, t_d, d_n(j))} \tag{3}$$

$$= \frac{Th(D_{qua}, t_d, d_n(i))}{Th(D_{qua}, t_d, d_n(j))}$$

The amount of deuterium released from a sphere droplet within a short period, can be described by using quasi dispersion coefficient as an equation (4) (Ref. 8) based on the discussion at Sec. II.A.

$$Th(D_{qua}, t_d, d_d(i)) = 1 - \frac{6}{\pi^2} \sum_{n=1}^{\infty} \frac{1}{n^2} \exp \left[ \frac{-D_{qua} n^2 \pi^2 t_d}{d_d/2^2} \right] \tag{4}$$

The function *EE* which divides each experimental result from the four different nozzles, is also defined as an equation below.

$$EE(d_n(i), d_n(j)) = \frac{E(d_n(i))}{E(d_n(j))} \tag{5}$$

By comparing of *TTh* to *EE* for the four different nozzles, if there is a unique quasi dispersion coefficient to fulfill following condition, it is the specific value of this experiment.

$$TTh D_{qua}, d_n i, d_n j = EE d_n i, d_n j$$

To perform a complete parametric survey, the following equation is prepared,

$$Err(D_{qua}) = \frac{1}{16-4} \sum_{i,j=1}^4 1 - \delta_{ij} \left( \frac{EE d_n i, d_n j}{TTh D_{qua}, d_n i, d_n j} \right)^2 \tag{6}$$

where  $\delta_{ij}$  is the Kroenecker's delta and is used to eliminates the division with same data.

A parametric survey of the quasi dispersion coefficient was performed, and the survey results at 450 °C are shown in Fig. 5. The horizontal axis is the parametric value of quasi dispersion coefficient, and the vertical axis is the resultant error value of the parameter survey. For all four temperature cases ranging between 375 °C and 450 °C, the existence of a unique *D<sub>qua</sub>* value was confirmed within 0.003 to 0.013 errors.

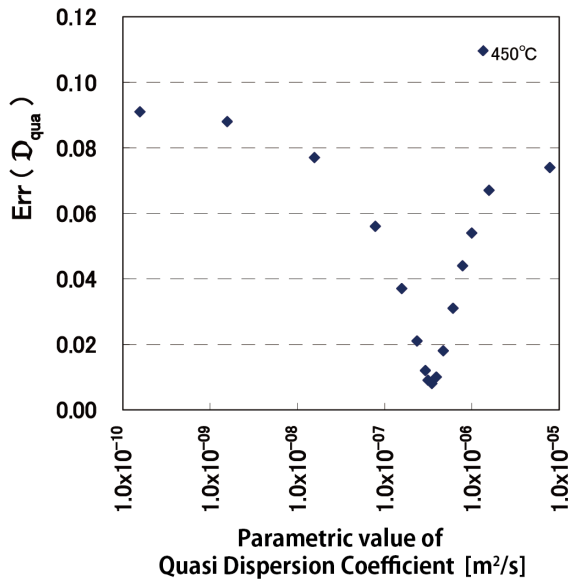


Fig. 5. Result of the parameter survey of quasi dispersion coefficient of deuterium in falling Pb-17Li droplet at 450 °C. Horizontal axis shows the range of parametric survey value. Vertical axis is the corresponding error by Eq. (6). Unique value was determined as  $3.4 \times 10^{-7}$ .

**II.E. Observation of Falling Droplets with a High Speed Movie Camera**

As shown in Fig. 6, a cyclic deformation between a spherical-shape and an ellipsoidal-shape was observed for the falling droplet. The deforming frequency of a droplet from the 0.6 mm diameter nozzle was approximately 200 Hz. The amplitude of deforming radius was 13 % , and the size effect for mass transport was estimated between plus 7 % and minus 8 % by the calculation, so the effect of deformation from sphere can be neglected, due to the accuracy of this study.



Fig. 6, Oscillating deformation of one droplet, recorded with high speed video camera. Frequency was approximately 200 Hz.

**III. RESULTS AND DISCUSSIONS**

From this experiment, the resultant quasi dispersion coefficient as a function of temperature is shown in Fig. 7, and a previously reported diffusivity example is plotted in the same chart for reference. The attained value was  $D_{app} = 3.4 \times 10^{-7}$  [m<sup>2</sup>/s] at 400 °C in this study, and was approximately three hundred times larger ( $1.2 \times 10^{-9}$  [m<sup>2</sup>/s]) in a study by Reiter.

The temperature dependency of this experiment showed less dependency than those of Reiter, which was described by an empirical equation [ $D = 4.03 \times 10^{-8} \exp(-19500/RT)$ ]. The cyclic deformation, frequency of approximately 200 Hz, on the droplet was confirmed.

These results show the mass transport process, which includes convection and diffusion, also occur at the release of deuterium from falling liquid Pb-17Li droplets in a vacuum.

The elimination of the ambiguous solubility was well functioned by this method, considering the deviation of the attained results.

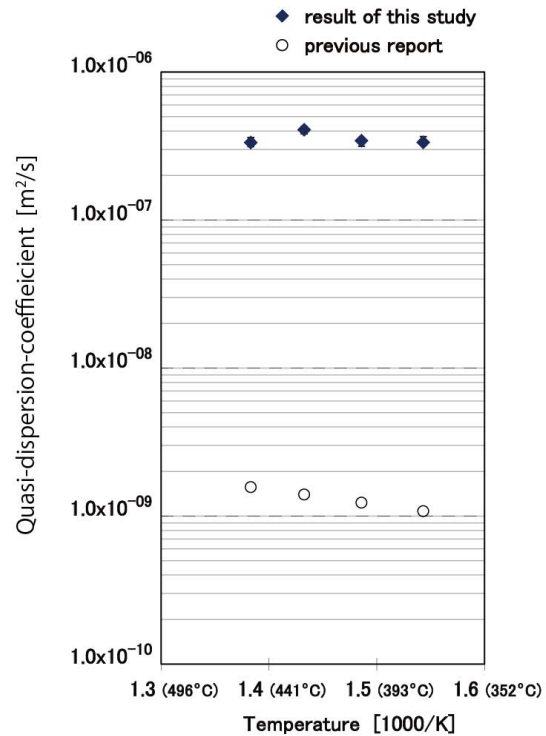


Fig. 7. Plots of the quasi dispersion coefficient of deuterium in the falling Pb-17Li droplets, versus the inverse of experimental temperature. The previously reported diffusivity values by Reiter are also plotted for reference. The attained results show two orders of magnitudes higher value. The less temperature dependency is considered that the convective prior mass flow also occurs in the Pb-17Li droplet.

## IV. CONCLUSIONS

The released deuterium from the falling liquid droplets of Pb-17Li in vacuum was experimentally measured without the ambiguous effects of solubility. Experiments were performed to compare different diameter nozzle data, to eliminate solubility effects. The mass transport of deuterium in Pb-17Li droplet, represented by quasi dispersion coefficient, was identified as  $D_{qua} = 3.4 \times 10^{-7}$  [m<sup>2</sup>/s] at 400 °C and is two orders of magnitude faster than the previous studies of under static condition. The attained results had less temperature dependency than previous studies. Cyclic deformation of the droplet shape was observed. These phenomena show that, mass transport mechanism under convective domain, also occur for the release of deuterium from falling liquid Pb-17Li droplets in vacuum. It contributed the higher mass transport. This result suggests that the tritium recovery method from the breeding liquid Pb-17Li blanket and VST device is viable.

## ACKNOWLEDGMENTS

This investigation was supported by funding from the Kyoto University GCOE program. The high speed movie was provided by Mr. Ochi of the KEYENCE CORPORATION.

## NOTATION

$k$	Mass transfer coefficient	[m / s]
$c_{1i}$	Concentration of solute #1 at the interface	[mol / m <sup>3</sup> ]
$c_1$	Concentration of solute #1 at the bulk	[mol / m <sup>3</sup> ]
$D_{qua}$	Quasi dispersion coefficient; defined on this study as	[m <sup>2</sup> / s]
$d_d(i)$	Diameter of the droplet from the nozzle (i)	[m]
$M^{de}(d_n(i))$	Amount of released net deuterium from droplets through nozzle (i), while a falling period.	[mol]
$M^{LiPb}(d_n(i))$	Mass of Pb17Li droplets from a nozzle (i) while one measurement	[mol]
$N_1$	Flux of solute #1	[mol / m <sup>2</sup> s]
$S$	Solubility	[mol / mol]
$t_d$	Dropping period of one Pb17Li droplet in VST	[s]

## REFERENCES

1. E.MAS DE LES VALLS, L.A.SEDANO, L.BATET, I.RICAPITO, A.AIELLO, O.GASRALDI, F.GABRIEL, "Lead-Lithium Eutectic Material Database for Nuclear Fusion Technology," *J. Nucl. Mater.*, **376**, 353-357 (2008).
2. F. REITER, "Solubility and Diffusivity of Hydrogen Isotopes in Liquid Pb-17Li," *Fusion Eng. Des.*, **14**, 207-211 (1991).
3. M. ADEKOHO WAHEED, MARTION HENSCHKE, ANDREAS PFENNIG, "Mass Transfer by Free and Forced Convection from Single Spherical Liquid Drops," *Int. J. Heat Mass Transfer*, **45**, 4507-4514 (2002).
4. G. TAYLOR, "Dispersion of Soluble Matter in Solvent Flowing Slowly Through A Tube," *Proc. R. Soc. A.*, **A219**, 186-203 (1953).
5. R. ARIS, "On the Dispersion of a Solute in a Fluid Flowing Through a Tube," *Proc. R. Soc. A.*, **A235**, 67-77 (1956).
6. R.B. BIRD, W.E. STEWART, E.N. LIGHTFOOT, *Transport Phenomena 2<sup>nd</sup>*, pp. 643-646, John Wiley & Sons, Inc., ISBN 978-0-470-11539-8.
7. E.L. CUSSLER, *Diffusion Mass Transfer in Fluid Systems-3rd*, pp. 95-107, 2009, Cambridge University Press, ISBN 978-0-521-87121-1.
8. J. CRANK, *The Mathematics of Diffusion. -2<sup>nd</sup> ed.*, pp. 89-91, Oxford University Press, Oxford , U.K. (1979). ISBN 978-0-19-853411-2.