A critical review of measuring technique based on laser induced fluorescence and its future application in verification and validation of advanced light water reactor simulation

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Abstract: LIF (Laser Induced Fluorescence) is a non-intrusive, whole-field and instantaneous measurement technique that has been applied in many measurement fields. Embarking from the present research situation, this work presents an introduction of principles and the early application of LIF technique to measuring visual mixing and concentration distribution, temperature fields and liquid film thickness. Analyzing the advantages and disadvantages of the technique, LIF technique can be used as the method of validation and verification of computational fluid dynamic (CFD) models. According to analysis of applicability, LIF technique has a wide range of application. It can measure the temperature fields and observe mixing phenomenon in the rod bundles, boron dilution in the downcomer and the liquid film thickness of containment. In conclusion, LIF technique can verify and validate advanced light water reactor simulation in the future.

Keyword: laser induced fluorescence; application; validation and verification; advanced light water reactor

1 Introduction

The numerical simulation, compared with the experimental research, has advantages of lower price, shorter period and easier to repeat. Besides, the method has less hypotheses and a wider range of applications. The method can be able to simulate intricate flow fields, micro scale structure and developing process. It is pretty important for modern design and modification of commercial or laboratorial application. Recently, numerical simulation has been developing very fast and has been applied to a variety of research field. However, reasonable confidence in CFD results must be built through validation with reliable experimental data. Obtaining reliable experimental data needs effective measurement methods.

With the development of numerical simulation, reliable experimental data is essential to verify and validate the confidence of number simulation. The traditional contact measurement methods have many limitations. Traditional measurements have been typically carried out with intrusive devices such as cold wires, thermocouples and RTD's sensors and so on, which are limited by point measurements, significantly perturb local velocity and temperature fields and suffer from limited time response due to a significant probe thermal inertia.

Hence, new requirements of measurement technique are put forward. These are whole field, indirect, non-intrusive and real-time measurement techniques. Laser diagnostic technique with the advantage properties mentioned above, is the new measurement technique which would have promising application in recent years.

Laser diagnostic technique includes laser-scanning pressure-sensitive paint, molecular tagging velocimetry/thermometry, particle image velocimetry, laser doppler velocimetry, laser induced fluorescence. In essence, laser diagnostic measuring is the optical measuring, based on scattering, reflection, refraction, fluorescence, and phosphorescence. In this paper, we only concern the technique of laser induced fluorescence.

LIF technique, as the non-intrusive experimental techniques to study visual mixing and local concentration distribution[1], temperature fields[2-4], and liquid film thickness[5-10] with good spatial and temporal resolution, can be used as the method for

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validation and verification of computational fluid dynamic (CFD) models.

In the present study, the future application of LIF technique in verification and validation of advanced light water reactor simulation is proposed. With the development of LIF technique, it is predicted that the technique be applied to measure the temperature fields and mixing in the rod bundles, boron dilution in the downcomer, and the thickness films of containment, and then to verify and validate the applicability of the CFD analysis.

2 Principles of LIF

2.1 Principles of fluorescence emission

As is known to all, the energy can be transferred to or pass through the material in a variety of forms. When a beam of light passes through a material, the light can be absorbed, reflected, transmitted, and scattered[11]. The absorption of photons of light will excite the molecules of the material to an electronically excited state where the absorbed energy can be converted to rotational, vibrational (*i.e.* heat), or chemical energy, or re-emitted as photons with lower energy. This is a process known as photoluminescence.

When dye molecules absorb electromagnetic radiation and subsequently re-radiate photons of lower energy, the dye emits two different types of photoluminescence, including fluorescence and phosphorescence. The time of the re-radiation process is the major difference between them two. For fluorescence, the absorption of light of a particular wavelength is followed by the emission of light of longer wavelengths within a few nanosecond. However, phosphorescence is relatively long-lived, whose emission lifetime range from milliseconds to minute[12]. This paper only concerns the principles of fluorescence. The process of fluorescence can be schematically illustrated by a simple diagram of the energy states as shown in Fig.1. Fluorescence is the result of a three-stage process: absorption, existence at an excited state, and emission.

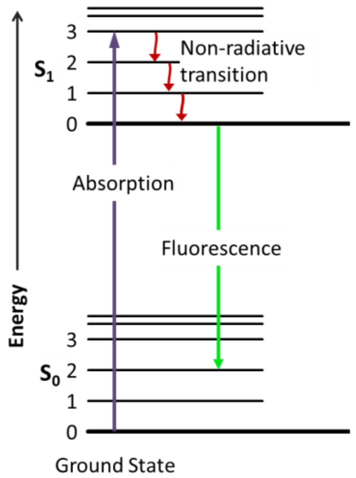

Fig.1 A schematic diagram illustrating the processes involved in optical absorption and subsequent emission of fluorescence[12]

The process of fluorescence is generated as shown below:

Excitation:

$$S_0 + h\nu_{ex} \rightarrow S_1 \tag{2-1}$$

Fluorescence emission:

$$S_1 \rightarrow S_0 + h\nu_{ex} + heat \tag{2-2}$$

Here $h\nu$ is a generic term for photon energy with h is Planck's constant and ν is frequency of light. State S_0 is called the ground state of the fluorophore and S_1 is its first excited state[13, 14]. The specific frequencies of exciting and emitted light are dependent on the particular system.

When photons of energy $h\nu_{ex}$, where ν_{ex} is the frequency of the excitation, incidents upon the fluoroscope from an external light source such as an incandescent lamp or a laser. This absorbed photon excites an electron in the fluorophore from the ground state to a higher energy state. The electron then exists in this excited state for a very brief time, known as the excited-state lifetime, which is typically picosecond to nanosecond.

In general, fluorescence emission will occur if the excited photon energy is greater than the minimum energy corresponding to the band gap, so the emission spectrum is typically independent of the excitation wavelength.

2.2 Principles of the measurement technique

LIF is a new measurement technique and has the advantage properties such as of whole field, indirect and non-intrusive. The method has been successfully applied to the full fields of temperature measurement, concentration distribution and liquid thickness. Many scholars at home and abroad have also gradually carried out research.

The fluid which would be measured adds some dye/dyes under the laser with appropriate wavelength, and then the fluorescence with special intensity would be emitted. The main principle of LIF is shown as Eq. (2-3) and detail was described by Lemoine [14].

$$I = K_{opt} V_c I_0 \Phi \varepsilon_1 C e^{-C(\varepsilon_1 b+\varepsilon_2 e)} \qquad (2\text{-}3)$$

Where K_{opt} is an optical calibration constant, V_c is the collection volume, and I_0 is the incident intensity of the laser beam. ε_2 is the molar extinction coefficient of the fluorescence signal, which is much lower than the coefficient ε_1 due to the spectral shift between the laser radiation and the fluorescence emission. C is the dye concentration and Φ is quantum yield of dye which defined as the ratio of the number of photons emitted to the number of photons absorbed. Φ changes with pH and temperature[13, 14].

The expression for the fluorescence signal takes into account the attenuation of the incident laser beam intensity, caused by absorption, when the laser pass through the adsorbing medium (distance b). Besides, fluorescence emission is attenuated while passing along the distance e in the absorbing medium (see Fig.2).

We can see from the Eq.2-3, if the term $C(\varepsilon_1 b+\varepsilon_2 e)$ is weak enough, the influence of Beer's absorption on the laser beam path can be ignored. In other words, the concentration is low enough and the path is short enough, the measure fluorescence signal turns out to be directly proportional to the concentration as other parameters keeping constant or known[13]. The Eq.2-3 can be simplified as:

$$I = K_{opt} V_c I \varepsilon_{10} C \Phi(T, pH) \qquad (2\text{-}4)$$

Besides, the molecular transition time and the lifetime of the excited state are very short, generally less than $10^{-9} s$ [15]. Therefore, the florescence technique is able to follow high frequency concentration, temperature and pH. Generally, the frequency response of the system is limited by the detection device.

By summarizing above, we can assume that LIF can be used as the measurement of pH[16], concentration[15, 17] and temperature [18, 19] if other parameters keep constant or known.

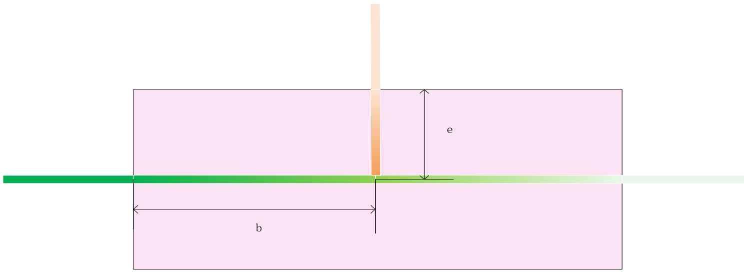

Fig.2 The process of emitting fluorescence.

3 Design consideration

Although several studies pertaining to fluid mechanics have used LIF technique as a diagnostic technique for both flow visualization and mixing measurements, concentration and temperature, the development of the technique are limited by four factors which are properties of dyes, optical limitations, calibration limitations, and Practical applicability.

3.1 Properties of dyes

Photo-stability, equilibrium concentration and combination of dyes are the major conflicts in the respect of properties of dyes.

A common problem quoted in the literature about using fluorescein is lack photostability or problems due to "photobleaching" [20-24]. Photobleaching (or photodegredation) is a result of continued exposure of the dye to the chosen excitation frequency. It refers to the decay of the emission of light intensity over a period of time due to photodecomposition or collisional quenching of the dye[25]. For example, Saylor[21, 23] recommended using pulse times much less than characteristic photobleaching times, which are in the millisecond time scale, to minimize photobleaching effects. Although Nd:YAG lasers have instantaneous energy densities that are much higher than continuous wave sources, they also have pulse lengths on the order of a few nanoseconds, which are approximately $10^{-5}s$ times less than previously measured photobleaching time scales.

It's scarcely possible to eliminate the influence of the photobleaching. In the design of experiments, photobleaching can be minimized by frequent calibration, minimizing the exposure time of the dye to the excitation light, and using the longest excitation wavelength possible to excite the dye[26]. The difficulty of the experiments are greatly increased.

The fluorescence intensity was linearly related to the dye concentration until fluid reaches the equilibrium concentration[11, 27]. As is known to all, different dye has different equilibrium concentration. The concentration should be selected under the equilibrium concentration when using the methods for flow visualization and mixing measurements. Besides, if the concentration is high enough, the influence of absorption along the laser beam path can't be ignored[13, 28]. This will increase the unevenness of light intensity distribution. Tan[29] exposed that the change of concentration also has an influence on the temperature sensitivity, especially for two different temperature tendency dyes, and it is obvious in mixed band when the two-color/two dyes technique measured temperature fields.

In conclusion, the choice of concentration is very essential in the experiments of Laser Induced Fluorescence.

3.2 Optical limitations

Optical limitations to the accuracy of the system can arise from curvature in the container or of the fluid, from light source contamination, and from variations in the refraction index due to temperature variations[25]. Firstly, measurements in containerless fluid flows will require correction for the image refraction due to the curvature of the fluid itself. The same is true for fluid moving in a curved container. Secondly, since ambient light often contains a number of possible dye excitation frequencies, one must take care to minimize the possibility of alternative excitation sources. Finally, when mapping temperature variations in a fluid, the changes of the fluid index of refraction will change. Since the changes are almost identical in both ratio frequencies, the only error here is due to inaccuracies in spatial information. Although the temperature measured will be correct, the image will be blurred or distorted thereby reducing spatial resolution. Besides, the design of optical circuits is influenced by external environment.

3.3 Calibration limitations

For LIF technique, the greatest uncertainty results from the camera response[25]. For the demonstrations, we used 6 bits of an 8 bit camera (linear region). This corresponds to less than 100 different possible pH or temperature states. These errors can be reduced by using a higher grade camera (12 bit linear gives 4096 possible states). One can choose dyes to maximize the response for the specific application to optimize the system for the number of possible states. Further, one must calibrate for spatial variations in camera response and, for analog cameras, one must account for frame grabber and line noise as well.

Besides image processing technology is one of important factors to limit the LIF technique. Crosley[30] pointed out that the nonuniformity of light intensity of light sheet is obvious, and the peak of light intensity is 1.5 times more than the average. Kychakoff[31] measured the radical mole concentration of OH component concentration of combustion gas and the nonuniformity of light sheet intensity is corrected by programmed software. Due to little absorption along the direction of optical path, variation of fluorescence intensity can be ignored. Since then measurement accuracy is greatly increased.

3.4 Practical applicability

So far LIF technique has been only used in low-parameter and visual experiment measurements. These parameters include ambient temperature and pressure and suitable pH. Too higher or too lower temperature can affect the temperature sensitivity and even fail some important properties of dyes. Besides, some dyes can be dependent on pH and spectral characteristic curve will be changed.

What is more, experimental facilities must be transparent and visual, which makes it difficult to manufacture. Experimental facilities with complex construction needs to take into account refractive error and so on. Furthermore, because of complex optical path, it is more accurate and better. Small changes in light intensity and incident angle can even introduce large errors. So each experiment need calibration measurements again.

These factors mentioned above will be a large challenge for LIF technique to make application and dissemination.

4 The application of LIF technique

LIF technique has had a wider application in many fields. In the early application, it can be used to measure temperature fields, concentration distribution and liquid thickness in the single-phase flow model. Afterwards, the technique is used as the method for validation and verification of computational fluid dynamic (CFD) models. The details are introduced as follows.

4.1 Early application of LIF technique

According to our research, many studies have been studied about the application of LIF technique. Most of these studies used a single dye, fluorescein, as a fluorescent tracer. Fluorescein is ubiquitous in LIF studies because its physical properties are ideal; excitable with both the 488 nm and 514 nm lines of an argon ion laser, water soluble, pH dependent emission, high quantum efficiency and low cost[15].

LIF can be the method to study visual mixing and local concentration distribution. When the concentration is low enough and the path is short enough, the measure fluorescence signal turns out to be directly proportional to the concentration as other parameters keeping constant or known[13]. So LIF technique can be used to measure concentration distribution[15, 17].

LIF technique has been applied to measuring temperature fields as well. At first, some scholars[13, 32-34] used only one temperature sensitive dye as fluorescer. However, the laser intensity distribution was always varying and behave according to a two-dimensional Gaussion distribution. In order to eliminate the influence of laser intensity, Coppeta *et al*.[25, 35] put forward ratiometric technique where the fluorescence emission intensity ratio chooses two different emission bands. Both bands can belong to one dye or two dyes. As we can see from the Eq. 4-1, not only the incident laser intensity but also the influence of concentration ratio, probe volume size, laser intensity, and Beer's absorption on the laser beam path are eliminated by processing the ratio of the fluorescence emission on the two color bands.

$$R=\frac{I_1}{I_2}=\frac{\Phi_1\varepsilon_1 C_1}{\Phi_2\varepsilon_2 C_2} \tag{4-1}$$

Thereafter, two-color/single-dye technique, were developed[18, 35-37]. By the way, two spectral detection bands with different temperature sensitivities of dye were chosen. To overcome re-absorption of the fluorescence in larger optical paths, Lavielle.[38] promoted three color techniques, which eliminated the influence of optical path and concentration. The method appears to promote the development of temperature measurement with LIF technique.

LIF measurement technique has also been applied to measuring liquid thickness. LIF technique is method with a very high sensitivity, very good spatial resolution and practical applicability to measure the thickness of liquid films. Beyrau[39] developed a method based on laser-induced fluorescence which can be applied inside a high-pressure, high-temperature vessel to measure the thickness fuel wall films. The liquid thickness in the annular is studied with high-speed LIF technique[39-41]. LIF measurements use a fluorescing material within the liquid phase, a monochromatic laser light, and a camera for observation of the resulting fluorescence[40, 41]. Schbring[40, 41] used planar laser-induced fluorescence to provide direct visualization of the liquid film in upward vertical air-water annular flow. It indicates that the measurement of fluid films is practicable and will have promising application.

As mentioned earlier， many researches about the measurements of temperature fields, concentration distribution and liquid thickness have been studied. The non-intrusive experimental technique to measure local temperature and velocity, with good spatial and temporal resolution is desirable, and is still the focus and hotspot in the research of the later.

4.2 Verification and validation numerical simulation

A cost effective optimization process requires a numerical simulation tool such as CFD methodology. Reasonable confidence in numerical simulation tools' results must be built through validation with good experimental data. LIF technique, as the non-intrusive experimental technique to study visual mixing and local concentration, temperature, and liquid thickness with good spatial and temporal resolution, can be used as the method for validation and verification of computational fluid dynamic (CFD) models.

In recent decades years, Many scholars have investigated the LIF technique as the method of visual mixing and measurement of quantitative concentration. McDaniel[42, 43] addressed the issue of use of LIF as a quantitative, steady state diagnostic for compressible flows. Their work found particular application in the mixing systems of high speed engines where the ability measure local quantities over small volumes was particularly useful for diagnosing mixing rates in these systems and creating multidimensional datasets for validation and verification of CFD models. Given the analysis performed by McDaniel[42, 43] in developing the LIF for compressible flow application, the use of this model on CFD datasets was an attractive extension of the theoretical development.

Hartfield[44] performed such work, applying the model as a computational fluid imaging technique to steady state, 3-D CFD results and comparing directly to LIF imaging of high speed mixing system experiments. As mentioned earlier, LIF is an obvious candidate for application to COIL flowfields due to the use of I_2 as a reactant. Recently, Madden[45] proposed a method for comparison of computational fluid dynamic simulation and planar laser induced fluorescence images for a supersonic flowfield. They obtained the experimental date by the fluorescence of I_2 in the presence of a pulsed laser source and then designed the CFD model utilizing a numerical solution of the Navier-Stokes continuity equations for mass, momentum, and energy, with individual mass continuity equations. The model and the experiment PLIF images were very good agreement. So it is predicted that future work will focus on detailed examinations of the quantification of CCD pixel saturation, precise quantification of the laser sheet positioning and width, and parameterizations of mixing studies.

Besides, a few scholars have investigated temperature with the LIF technique to validate and verify CFD, and experimental temperature fields is very similar to that of the theoretical one.

Dahikar[46] performed the experiments for the steam injected centrally at the bottom of a vertical rectangular water vessel. Temperature fields near the plume as well as in the downstream have been measured in a vertical plane through the central axis by PLIF. Besides, they provided CFD simulation by employing $k-\varepsilon$ and large eddy simulation (LES) turbulence models. Comparison of experiment data with CFD models, experimental dimensionless temperature is very similar to that of the theoretical one.

Jaworski[47] investigated heat transfer in oscillatory flow conditions , which are typically found in thermoacoustic devices. In the experimental part, PLIF were applied to obtain spatially and temporally resolved temperature fields within the parallel-plate heat exchangers (HX) channels. The numerical part deal with the implementation of CFD modelling is implemented to analyze the time-averaged temperature fields. They pointed that the experimental data obtained from the measurement of the temperature fields provide an important reference for validation of a CFD code implemented on the basis of the classical thermoacoustic.

Narayanaswamy[48] performed experiments to explore the use of two-photon planar laser-induced fluorescence (PLIF) of krypton gas for applications of scalar imaging in supersonic flows. Experiments were performed in an underexpanded jet of krypton, which exhibited a wide range of conditions, from subsonic to hypersonic. The data were used to infer the distribution of gas density and temperature by correcting the fluorescence signal for quenching effects and using isentropic relations. The centerline variation of the density and temperature from the experiments agree very well with those predicted with an empirical correlation and a CFD simulation (FLUENT).

Few scholar has investigated liquid film thickness with the LIF technique to validate and verify CFD. Cheng, Yong-sheng[7] deals with the basic research on the fuel mixture preparation process, and reports the experimental and numerical investigations on characteristics of the wall-wetted fuel film. In the experiments, the film thickness on the wall was measured by using LIF technique. In the numerical simulation, the commercial computational fluid dynamics (CFD) software FLUENT was used. The results were shown that the agreement of two methods was not very good. However, the results can guide the numerical models about quid thickness to be consummate.

5 The future application to verification and validation of advanced light water reactor simulation

As mentioned above, LIF technique is successful to be applied to measurements of concentration distribution, temperature fields and liquid thickness. It has also been used as the method for validation and verification of computational fluid dynamic (CFD) models. In the future, we can predict that the technique will have wide application prospects in verification and validation of advanced light water reactor simulation. This paper will research from following several aspects.

5.1 Boron dilution in the downcomer

In recent years, inhomogeneous boron dilution has recently become one of the most important issues in PWR safety. Rohde[49] performed the experiments on slug mixing and gained the concentration distribution determined by measuring the conductivity at two test facilities, modelling different reactor types in scale 1:5, the Rossendorf test facilities. Sodium chloride simulating the boron concentration is used as tracer in the test facility. The experiment data helped in the clarification of the mixing mechanisms and should form a data basis for the validation of computational fluid dynamics (CFD) codes. However, mesh sensors perturbed the local flow state to some degree.

So non-intrusive diagnostics are attractive because of the inherent ability provided local measurements with minimal perturbation of the flow. LIF technique is attractive for a variety of reasons as described earlier. Gavelli[50] presented the application of the LIF technique to the study of liquid mixing in the downcomer of a pressurized water reactor. In the experiments, the boron concentration is simulated by an aqueous solution of a fluorescent tracer. High resolution quantitative information on the mixing phenomena occurring in the reactor vessel downcomer during the slug injection was gained by using a non-intrusive approach. The results can be applied to the study of liquid mixing in the complex geometry of a reactor vessel downcomer.

Kiger[51] presented quantitative measurements of scalar dilution transients within a scaled model of a PWR downcomer. The injection transients correspond to the case of a single pump start-up injecting into an initially stagnant system. The mixing measurements were conducted by means of LIF technique. The results are validate computational fluid dynamics codes that are starting to be used to simulate the mixing phenomena encountered in such complex systems.

5.2 Temperature and flow distribution in rod bundle

As is known to all, the temperature and flow distribution in the coolant and along the rod bundles is one of major importance in nuclear reactor design and safety assessment. Therefore, experimental investigation of the flow and heat transfer in rod bundle geometry is one of the hotspots in the field of nuclear research. In the numerical part, many scholars have done a lot of research. Amongst the many studies performed involving CFD simulations of rod bundles.

Recently, Toth and Aszodi[52] developed a CFD model of VVER-440 fuel assembly to perform steady-state and transient calculations. Effects of the spatial resolution, turbulence models, difference schemes and different inlet boundary conditions were also investigated. Using the STAR-CD code, Conner[53] had presented the CFD methodology for a specific

5×5 rod bundle. This CFD model had been validated by benchmark experiments.

Liu [54] developed a three-dimensional (3-D) CFD model with the Reynolds stresses turbulence model to simulate the thermal–hydraulic characteristics in a rod bundle and to investigate the effects of different types of grid on the turbulent mixing and heat transfer enhancement.

Liu.[54] also studied effects of the numerical methods such as mesh refinement, wall treatment, and appropriate definition of boundary conditions on the CFD predicted results for a rod bundle.

However, few related experimental studies are investigated deeply in recent years. As is known to all, reasonable confidence in CFD results must be built through validation with good experimental data. Some verification and validation calculations are necessary to confirm its applicability to a relevant aspect before adoption of the CFD analysis to the real design case. With the development and maturity of the LIF technique, we can predict that the technique be applied to measure the temperature flied and mixing in the rod bundles to verify and validate the applicability of the CFD analysis.

5.3 Falling films measurements

Vertically free falling films, have a wide range of application in passive safety containment cooling system of AP1000, nuclear seawater desalination system, and gas-liquid separation system of steam generator, so it is very important to find a suitable method to measure thickness of falling films.

Excepting its advantages of high heat transfer coefficient and low energy consumption[1], vertically free falling films' nonliner flow characteristic, oscillation instability and break behavior under thermal effect or stress conditions will have great influence on the performance of industry equipments. Some examples can be the second carry-on problem of SG and local drain out of passive containment cooling system (PCCS). So it is necessary to conduct comprehensive researches on oscillation behavior of liquid thickness[2-4] and spacial-temperal evolution pattern. How to precisely measure fluctuation liquid films has been a key problem of fluid dynamics wave characteristics.

The traditional contact measurement methods can be easily affected by liquid surface tension, and probe would surely sabotage the boundary conditions of measurement point to interfere the flow characteristic of liquid films. Meantime, the measurement has low accuracy, because it's only an average value of a wide area. Besides, capacitance measurement system has high output resistance, so it is sensitive to outside perturbation and tends to have instability phenomenon. Most normal optical methods analyze light to measure film thickness. Taking the light-absorption method as an example, it's more effective for smoother interface with smaller perturbation. But it may have false measurement for film with surface oscillation due to multi-refraction of light inside liquid.

LIF technique, with the advantage properties such as of whole field, indirect non-intrusive and instantaneity, has been used to measure thickness thickness fuel wall films[39] and liquid thickness in the annular[40, 41]. According to the researches we mentioned above, we would predict that the measurement of liquid films with the technique will have the extensive application of nuclear industry such as passive safety containment cooling system of AP1000, nuclear seawater desalination system, and gas-liquid separation system of steam generator.

6 Conclusion

So far, measuring technique based on laser induced fluorescence and its future application in verification and validation of advanced light water reactor simulation is one of the hotspots. This work provides a brief introduction of laser induced fluorescence about the principles and application. LIF technique can be used as the measuring method of visual mixing and concentration distribution, temperature fields and liquid thickness and so on. Besides, it can also apply to validation and verification of computational fluid dynamic (CFD) models. Many researches show that experimental results based on LIF technique are very similar to that of the theoretical one. Judging from the analysis above, LIF technique has broad application foreground. A few studies have been carried out that the technique has been applied to measuring the temperature fields and observing mixing phenomenon in the rod bundles. Also, it has been used to measure boron dilution in the downcomer. Furthermore, it may be adopted to measure the liquid film thickness of containment and gas-liquid separation system of steam generator. In conclusion, LIF technique has been verifying and validating advanced light water reactor simulation in the future. However, the applications of LIF technique will be limited by the choice of dyes, optical limitations, calibrations and practical applicability mentioned in the third part. The problems are that we have to consider and overcome in the future applications of LIF technique.

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