

Visualization Study on Chemical Reacting Flow using LIF

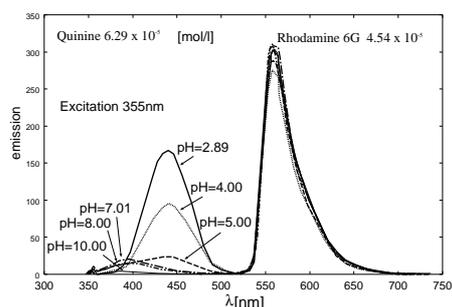
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ABSTRACT

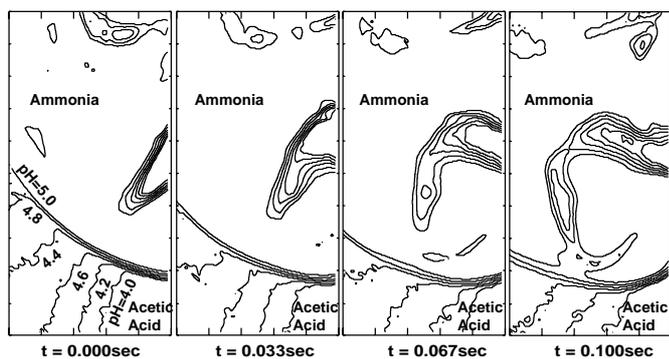
The sodium-water reaction should be precisely evaluated for the safety analysis of the sodium-cooled nuclear power plant. To evaluate these chemical reacting jet, the characteristics of the reaction and the mole fraction distributions of the reacting material should be known. In this study, to evaluate the basic characteristics, two fluid jet and chemical reacting jet was measured by the PIV and DELIF.

The momentum diffusion of the two fluid jet was found to be suppressed under water jet into acetic acid solution. The mechanism was not clarified yet, however, some chemical phenomenon may play an important role on the mixing suppression.

The new dye pair for the dual emission LIF technique was proposed to measure the pH distribution. The Quinine for pH sensitive dye with blue emission and Rhodamine 6G for non-sensitive dye with orange emission, were excited by the third harmonic of Nd:YAG laser (355nm). Left figure shows the emission spectroscopy of the mixed dye solution. The high accurate measurement could be achieved for the range of pH 4.0 to 5.5. The ammonia jet into acetic acid was measured using the proposed dye as shown in right figure. The effectiveness of the present method was demonstrated.



Emission spectrum of Quinine and Rhodamine 6G with 355nm excitation



pH distributions of ammonia jet into acetic acid

1 INTRODUCTION

In the steam generator of the Liquid Metal Fast Breeder Reactor (LMFBR), the heat is exchanged between the high temperature liquid sodium and boiling water. The steam generator is the one of the key devices for LMFBR. If the small hole were generated on the wall of heat transfer tube, high pressure water inside the tube might eject into the liquid sodium pool. This accident, SGTR (Steam Generator Tube Rapture), is the design basis accidents for LMFBR. Since the liquid sodium and water rapidly reacts to generate NaO_2 , NaOH and Hydrogen gas, the accident may cause the steam generator to be damaged. The phenomenon is called Sodium-water reaction. In the LMFBR, to reduce the high pressure of the sodium-water reaction, the pressure release system, e.g., Rapture Disks, were installed.

The scenario of the sodium-water accident was as follows. Initially, the small hole may be generated because of the vibration and/or thermal fatigue. When the raptured hole on the tube wall is enough small, the sodium-water reaction is dominated by the amount of water. The small water jet injected into sodium. The reaction is too small to damage the vessel. However, the generated corrosion medium, e.g., NaO_2 , may impinge the neighbor tubes. Then, the neighbor tubes may be broken to be the huge amount of water may release to the sodium, causing the huge pressure with rapid reaction. Therefore, to escape the huge accidents, the small rapture could be detected quickly. The initial stage of sodium-water reactor should be precisely evaluated.

The reaction may be affected by the turbulence jet and also the reaction itself. To evaluate the such chemical reacting jet, the characteristics of the reaction and the mole fraction distributions of the reacting material should be known. The turbulent mixing may enhance the reaction. Also the mixing may be affected by the reaction. The numerical simulation code for the Sodium-water reaction should be developed. However, the basic characteristics of the chemical reacting jet is not completely known.

Breidenthal and Koochesfahani measured the transition stage of the mixing layer¹⁾. Bennani²⁾ and Nagata³⁾ measured the reaction of two fluids inside the grid turbulence. Sakai et al.⁴⁾ measured the mole fraction using optical fiber. However, these measurement technique was not enough to reveal the chemical reacting jet well. The spatial distributions of the pH should be more precisely measured to evaluate the effects of reaction on the turbulence.

In this study, to provide the verification data for the numerical simulation code, the chemical reacting jet was measured by Laser Induced Fluorescent technique. The new fluorescent dye pair was proposed for measuring the pH from 4.0 to 5.5.

2 EXPERIMENT

2.1 Experimental Setup

In this study, to evaluate the LIF technique, simple reaction between acetic acid and ammonia was selected. Figure 1 shows the schematic view of the test section. As the ambient fluid, Acetic acid solution of $0.1 \text{ mol}/\ell$ was used. The acetic acid solution flew in the duct with $100\text{mm} \times 100\text{mm}$ cross-section. The velocity of the acetic acid solution was fixed to be $5 \text{ mm}/\text{s}$. At the center of the duct, the small circular nozzle with 2mm ϕ was set concentricly. From the nozzle, Ammonia solution of 0.1 or $0.5 \text{ mol}/\ell$ was ejected into the duct. The injection velocity was set to be $U_N = 0.1 \sim 0.5\text{m}/\text{s}$, i.e., $\text{Re} = 200 \sim 1000$. The acetic acid and ammonia reacted in the duct.



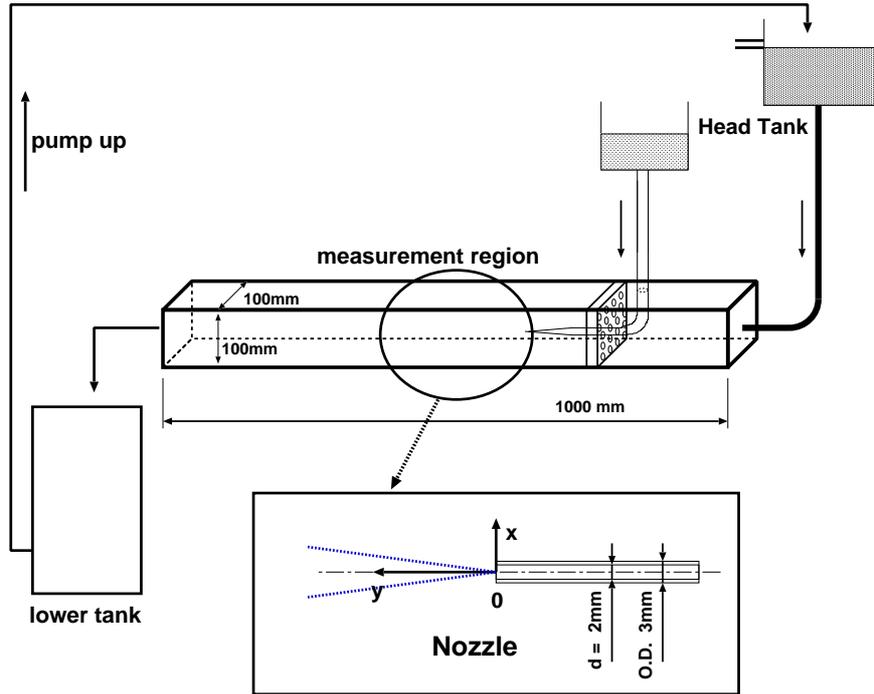


Figure 1: Experimental setup

The reacting factor of the above reaction is $k = 10^8 \text{m}^3/(\text{mol}\cdot\text{s})$. The Schmidt number is $Sc = 250, Da = 2 \times 10^8$. The generation of the reacting heat could be negligible.

To clarify the effects of the reaction, four cases of the experiments were carried out. The fluid for the cases are summarized as follows,

- (1) Water jet into water,
- (2) Water jet into acetic acid (0.1 mol/l),
- (3) Ammonia jet (0.1 mol/l) into acetic acid (0.1 mol/l)
- (4) Ammonia jet (0.5 mol/l) into acetic acid (0.1 mol/l).

2.2 Velocity Distribution

To measure the velocity distribution, Particle Image Velocimetry (PIV) was applied. The small tracer particles with $10 \sim 20 \mu\text{m}$ were inserted into the jet and ambient fluid. The twin Nd:YAG Laser (30Hz) were synchronized with the 60Hz progressive scan CCD camera to capture the particle images. The measured area were nozzle outlet and downstream.

Figure 2 shows the averaged velocity distributions. When the inlet velocity was lower, i.e., 50mm/s ($Re=200$), the injected jet curved to upward direction. Since the density of acetic acid solution was about 1.3 times heavier than that of ammonia solution, the curved jet was caused by the buoyancy effect. Under the lower velocity condition, the effects of buoyancy dominated, causing the effects of mixing to be undetectable. Therefore, the following discussion was focused on the higher velocity condition.

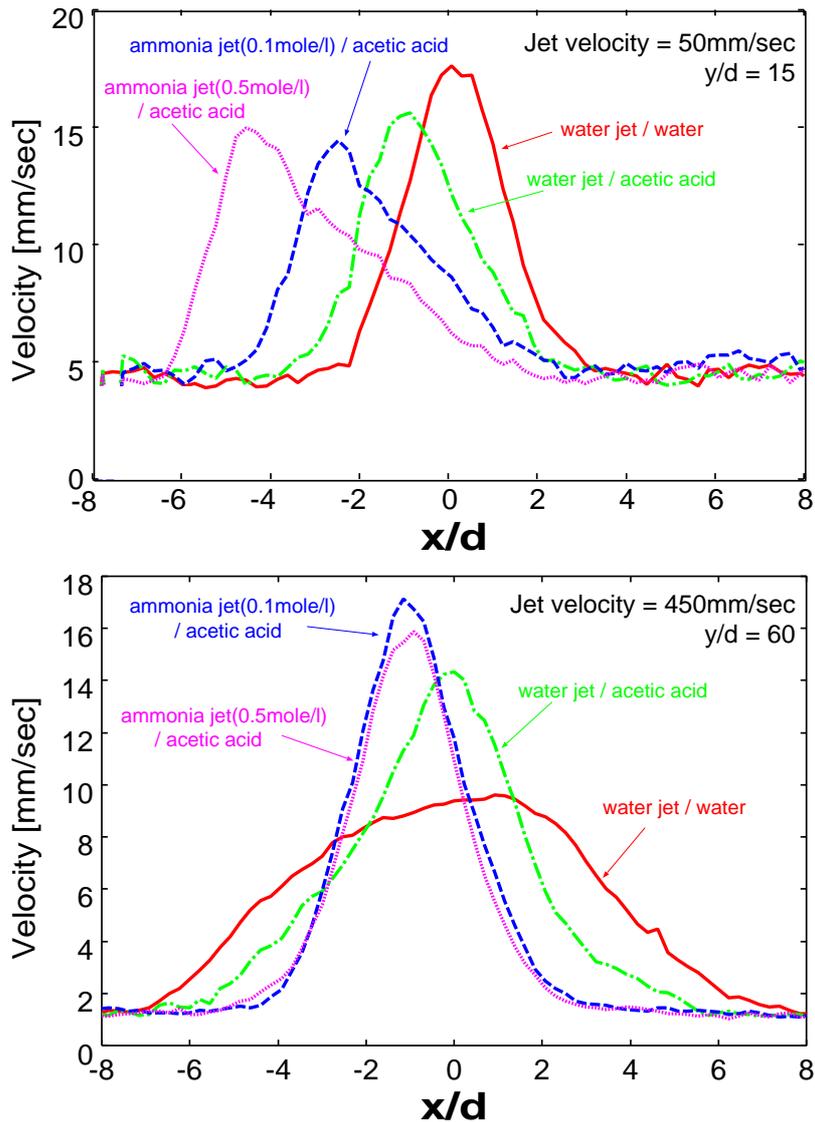


Figure 2: Diffusion estimated by velocity distribution

Figure 2 lower shows the velocity under higher jet inlet velocity condition at downstream, $y/d = 60$. Although the injected jet velocities are the same, the momentum diffusion pattern between jet and ambient fluid were differed. In Case (1), Water jet into Water, the jet expanded to $8d$, because of the jet mixing. While, in the other cases, Water jet into Acetic Acid and so on, the expansion of the jet was about $3d$. Under this condition, the mixing between Water jet and Acetic Acid was suppressed. With the Ammonia jet (cases (3) and (4)), the almost similar results were obtained. The suppression of the mixing was observed not only Ammonia jet case, but also Water jet case. This result means the suppression was caused by the two fluid mixing, not by the chemical reaction.

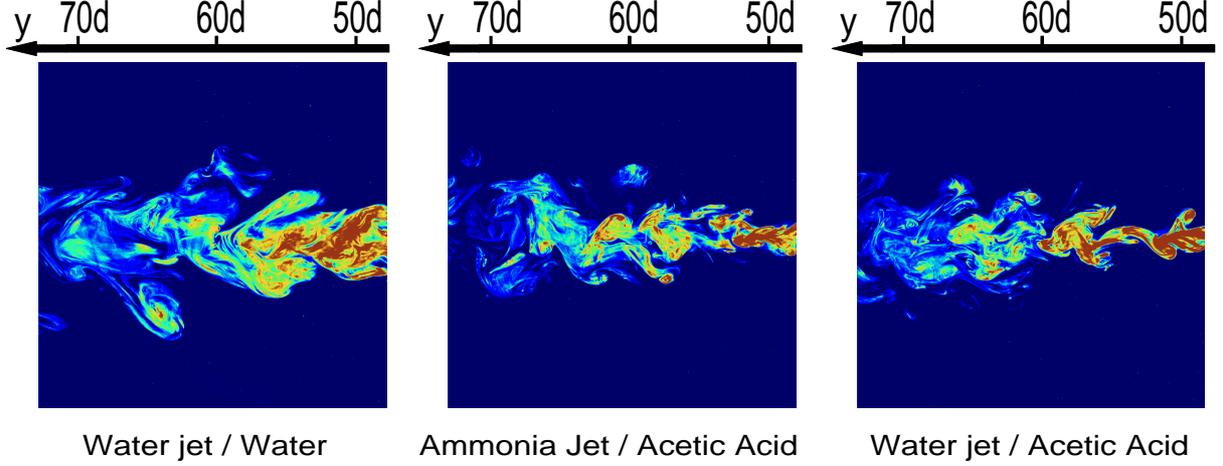


Figure 3: Jet diffusion taken by single emission LIF (pseudo color)

2.3 Momentum Diffusion

To investigate the two fluid mixing more clearly, the single emission Laser Induced Fluorescent (LIF) technique was applied to visualize the diffusion of the injected jet. The Rhodamine B was solved into the jet fluid. Then, the diffusive jet was visualized using the Nd:YAG Laser with high resolution CCD camera (1k x 1k/30Hz) Figure 3 shows the example of the diffused jet. The white area denotes the jet fluids. The diffusion of (1) water/water case was found to be much larger than that of the other cases. The LIF visualization was carried out under the Acetic Acid jet into Acetic Acid, which showed almost similar diffusion to Water/Water case. That is, the momentum diffusion of the same fluid was much larger than that of the different fluids.

These results indicate that the mixing of two different fluid was suppressed comparing with that of same fluid. Such kinds of the diffusion suppression effects were reported for polymer jet and/or surfactant jet. However, in the present experiment, the similar suppression effects were observed for water jet into acetic acid jet, whose molecular characteristics were completely different from polymer solution and/or surfactant solution. The mechanism of the diffusion suppression for the acetic acid was not known. Further investigation should be carried out.

Under the relatively lower Reynolds number, the effects of the chemical reaction was considered to be much smaller than that of fluid mixing.

3 Dual Emission LIF

To clarify the reacting phenomena, the DELIF (Dual emission LIF) technique was applied. The fluorescent of the dye was expressed as follows,

$$I_f(b) = I_e(b)A\phi L\epsilon C \quad (2)$$

where, I_f is the measured intensity at point b , I_e is the laser intensity at point b , A is the fluorescent, ϕ is quantum effect, L is unit length of beam path, ϵ is absorption, C is the mole

fraction of the dye. When the absorption and/or emission of the dye depends on the fluid pH,

$$I_f(b, \text{pH}) = I_e(b, \text{pH})A\phi L\epsilon(\text{pH})C$$

The problem is that the laser intensity I_e depends on the location b . If I_e is uniform, pH distribution could be easily detected with measuring of I_f . However, I_e was affected by the laser intensity, test section refraction, illumination path and so on. To overcome the problems, the dual emission LIF was proposed by many researchers. The pH independent dye was simultaneously dissolved into the fluid. When the fluorescence from pH dependent dye and pH independent dye denote I_{1f} and I_{2f} , respectively, the effects of the laser intensity I_e at location b could be neglected,

$$\frac{I_{1f}(b)}{I_{2f}(b)} = \frac{\epsilon_1(\text{pH}, \lambda)C_1\phi_1}{\epsilon_2(\lambda)C_2\phi_2}$$

The fluorescent ratio is independent of the laser local intensity. Using the ratio, the pH distributions could be accurately measured.

For the DELIF, the two different dye should be selected. These dye should be absorbed the certain wave length of the excited laser. Also, the emission wavelengths of the two dye should be independent. Coppeta and Rogers⁵⁾ used the fluorescein(Green) and rhodamin B(Red) with Ar-ion Laser excitation (Blue). However, the Ar laser is continuous wave laser. To detect the transient, pulsed laser excitation is required. As the pulsed laser, Nd:YAG laser was used, whose emission wavelengths are 532nm (second harmonic, Green) and 355nm (third harmonic, Ultra Violet). No dye pair was reported for the green and ultra violet excitation, in the previous studies.

In this study, 20 kinds of fluorescent dye were analyzed using the spectroscopy. As the results, the Quinine (Blue) and Rhodamin 6G (Orange) with 355nm excitation (Ultra Violet) were selected. Figure 4 shows the emission characteristics of these dye. The Quinine and Rhodamin 6G were dissolved into Acetic Acid, then the emission spectrum was analyzed using the spectro meter. Varying the pH with Ammonia, the emission spectrum was recorded.

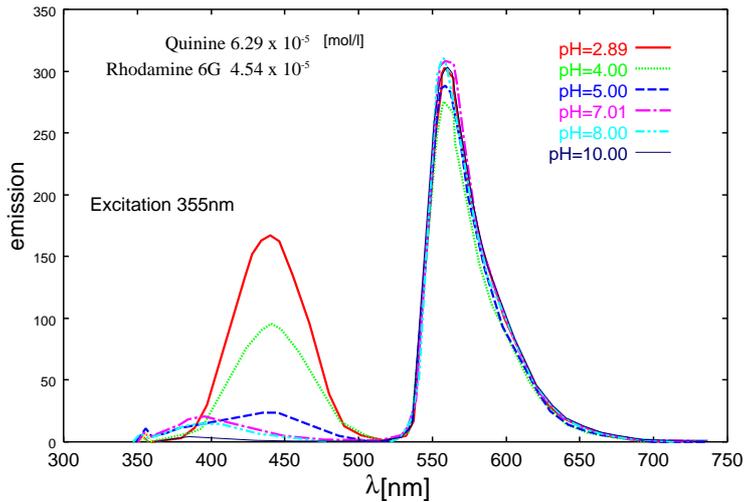


Figure 4: Quinine and Rhodamine 6G relative emission versus wavelength as a function of pH

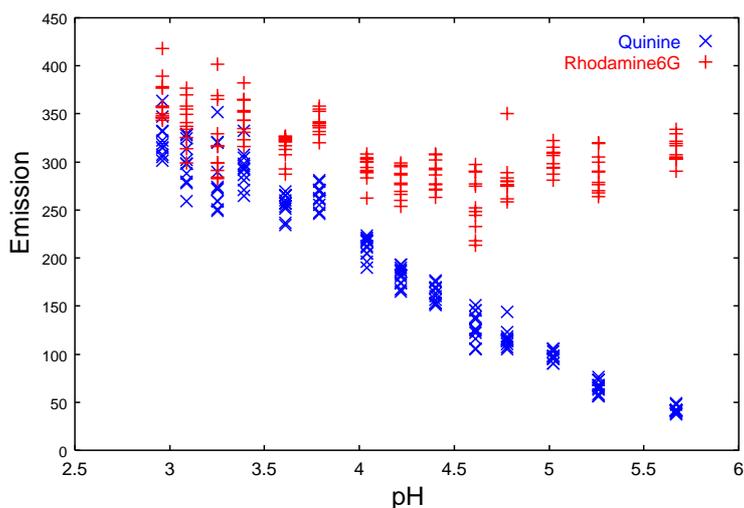


Figure 5: Emission plot versus pH

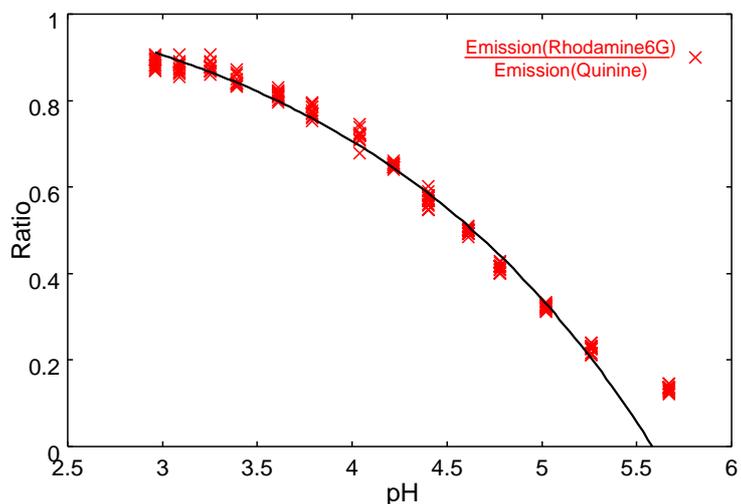


Figure 6: Calibration plot of normalized fluorescence ratios versus pH

The emission of Quinine distributed around 450 nm (Blue) and that of Rhodamine 6G did around 600nm (Orange). These emission peaks could be easily separated with using the optical filter. With decreasing pH, the emission intensity of Quinine also decreases, however, that of Rhodamin 6G keeps almost constant. The emission of Rhodamine 6G and Quinine denote the laser intensity and the pH, respectively.

To detect the emission of these two dye, the stereo viewer was introduced⁶⁾, which can record the two views of the same area with one camera. Two optical filters were set in front of the stereo viewer to separately record the Orange and Blue intensity.

The calibration of the dual emission LIF was carried out using the actual fluids and configurations. The acetic acid solution with Quinine and Rhodamine 6G were poured into the rectangular vessel. Under the constant temperature, the fluorescent intensity were recorded onto the CCD camera with varying the pH using ammonia. The optical setups, e.g., cylindrical

lens system and camera lens, were completely the same with the actual experiment. Figure 5 shows the individual fluorescent intensity for the two dyes. Although the characteristics of the two dyes can be qualitatively observed, (constant for Rhodamin 6G and decreasing for Quinine), the error seems to be relatively large. These larger error is caused by the unstable laser power and reflection of laser at the wall.

However, as shown in Figure 6, the intensity ratio of these two fluorescent, f_Q/f_{R6G} , has very few errors for the pH estimation. This calibration curve can be used for the measurement of pH. The accuracy based on the present calibration curve is about 5% for pH measurement.

Figure 7 shows the measured results of the downstream of ammonia jet into acetic acid solution with every 1/30 second. The location was $x = 60d$ from nozzle and $x = -10d$ from the center line of the nozzle. Since the view area for the present stereo viewer is limited, only the small area of the jet could be measured. The pH was calculated from the fluorescent intensity ratio using the calibration curve (Fig.6). The diffusion of ammonia vortex into acetic acid was clearly quantized.

Although the pH distribution was measured with high accuracy, there were several problems.

The measured pH range was only pH= 4.0 ~ 5.5. The pH at the vortex center region could not be measured, because of the limitation of the calibration curve. As shown in Fig. 6, only the range between pH= 4.0 ~ 5.5 were calibrated. To extend the measurable pH range, the concentration of the dyes and the fluorescent intensity should be controlled.

The measured area is limited to be small. With using the dichromic mirror and two CCD camera, the area could be extended to much larger area.

Because of the CCD camera, temporal resolution is limited to be 30 Hz. With using the double or triple pulse laser, the temporal resolution could be improved. Since the fluorescent delay is only 1ns and laser pulse duration is about 5ns, high speed reaction effects could be captured. The time scale of the turbulence is much slower than the chemical reaction.

4 CONCLUSION

- The momentum diffusion of the two fluid jet was measured using PIV. The mixing was suppressed under water jet into acetic acid solution.
- To measure the pH distribution, the new dye pair for the dual emission LIF technique was proposed. The Quinine for pH sensitive dye with blue emission and Rhodamine 6G for non-sensitive dye with orange emission, were excited by the third harmonic of Nd:YAG laser (355nm). The high accurate measurement could be achieved for the range of pH 4.0 to 5.5. The ammonia jet into acetic acid was measured using the proposed dye, showing the effectiveness of the present method.

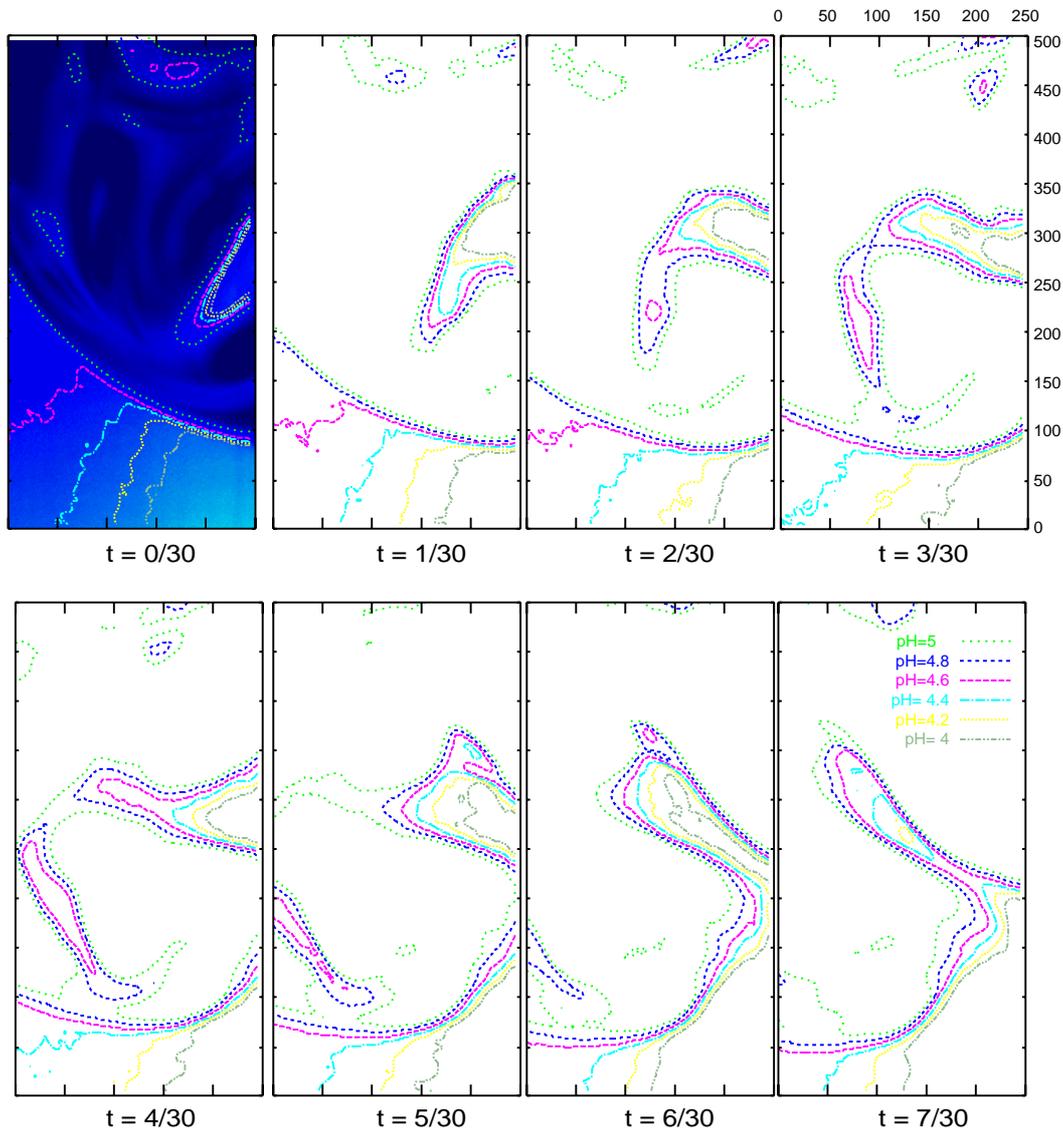


Figure 7: pH Distribution

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