A tunable 3D optofluidic waveguide dye laser via two centrifugal Dean flow streams†

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This paper presents a tunable optofluidic waveguide dye laser utilizing two centrifugal Dean flows. The centrifugal Dean flow increases the light confinement of the dye laser by shaping a three-dimensional (3D) liquid waveguide from curved microchannels. The active medium with the laser dye is dissolved in the liquid core and pumped with an external pump laser to produce stimulated emission. The laser’s Fabry–Pérot microcavity is formed with a pair of aligned gold-coated fiber facets to amplify the fluorescent emission. The advantage of the 3D optofluidic waveguide dye laser is its higher efficiency, thus to obtain lasing at a reduced threshold (60%) with higher output energy. The demonstrated slope efficiency is at least 3-fold higher than its traditional two-dimensional equivalent. In addition, the laser output energy can be varied on demand by tuning the flow rates of the two flows. This technique provides a versatile platform for high potential applications microfluidic biosensor and bioanalysis.

Introduction

Widely tunable microfluidic dye lasers are outstanding examples of optofluidics. 1,2 There, the liquid medium of the dye laser provides the optical gain and sets the lasing wavelengths. In contrast to bulk (jet-based) dye lasers, the optofluidic dye lasers are more flexible to select the TEM-modes and control the output energy. 3,4 A large variety of optofluidic dye lasers have been developed, such as Fabry–Pérot (FP) microcavity dye lasers, 5,6 microdroplet dye lasers, 7 distributed feedback dye lasers, 8,9 ring resonator dye laser 10 and waveguide dye lasers. 11 Among these optofluidic dye lasers, the liquid waveguide dye laser has attracted more interest because it may offer more freedom in tuning the liquid medium, as the structure is independent to the solid channel. 12 A dye laser usually consists of three parts. The first part is a dye dissolved solvent as the gain medium, the second part is a pump laser delivering the energy to the lasing medium, and the third part is a resonator providing feedback for the stimulated emission. In a liquid waveguide dye laser, the liquid waveguide is the key part as it retains all the functionalities associated with the waveguiding capability while providing a natural synergy with lab-on-a-chip systems. 13-14 Using liquid waveguide, the emission wavelength, the energy intensity and the modal content of the dye laser can be controlled in real time because the liquid waveguide is a dynamic system and can be reconfigured by changing the flow rates, the dye and the liquid compositions. A two-dimensional (2D) liquid–waveguide using three flow streams was demonstrated. 15-17 The microfluidic chip is made of polydimethylsiloxane (PDMS) with a refractive index of n ≈ 1.410 and the liquid cladding is made of DI water, n = 1.332. Therefore, the cut-off frequency and the gap of the liquid–liquid waveguide depend on the chip materials. 18 Propagation modes that should be confined in the liquid–liquid waveguide leak in the vertical direction, preventing the liquid cladding in the horizontal direction from functioning optimally and subsequently reduce the confinement factor. In addition, the cross-sectional liquid profile of the 2D liquid–liquid waveguide can only be varied in the horizontal direction. New concepts such as liquid cladding/air cladding waveguides 19 also suffer from the 2D limitations.

In a three-dimensional (3D) liquid–liquid waveguide, the core liquid is fully surrounded by the cladding liquid. The coating of the core liquid is realized by the transport of multiple streams through a curved microchannel. Due to the curvature, the liquid becomes accelerated, which leads to a transverse flow field, known as Dean flow. 17,18 Recently, a 3D hydrodynamic focusing has been reported in planar microfluidic devices for flow cytometers. 19

In this paper, we make use of the Dean flow to prepare a 3D liquid waveguide and demonstrate its feasibility with an optofluidic dye laser, which is designed using only two centrifugal Dean flow streams in a curved microchannel. The dye is dissolved in the liquid core acting as the lasing medium with an external pump laser to produce stimulated emission. An FP microcavity is formed in the microchannel with a pair of aligned gold-coated...
Design and theoretical analysis

Fig. 1(a) shows the schematic of the 3D optofluidic waveguide dye laser. It consists of two parts: the microfluidics to form a 3D liquid–liquid waveguide in the microchannel, and the micro-optical part to provide optical feedback for lasing. The microfluidic design can be divided into a curved and a straight microchannel. Two flow streams are injected into the microchannel and flow side-by-side, (inset 1 of Fig. 1). In the curved microchannel, Dean flow acts on the two flow streams with the counter-rotating flow profile (inset 2).

The transverse flow may transport the inner flow stream towards the outer wall while the outer flow stream is pulled inward causing the positions of both flow streams to be exchanged. Under certain conditions, the inner flow stream is fully surrounded by the outer flow stream (inset 3). When the refractive index of the inner flow stream is higher than that of the outer flow stream, a 3D liquid core–liquid cladding waveguide is formed. The 3D liquid waveguide structure is maintained downstream in the straight microchannel (inset 4).

Fig. 1(b) demonstrates lasing from the 3D optofluidic waveguide. The dye laser consists of a dye dissolved in the inner liquid of the liquid waveguide within a FP microcavity that is formed by two reflective mirrors positioned at the two ends of the waveguide. The pump laser is focused into the 3D liquid waveguide to excite the fluorescence dye in the vertical direction. The fluorescence emission is confined within the FP microcavity formed by two optical fiber facets at both ends of the straight microchannel. The fibers are coated with a thin layer of Au to improve the reflectivities, and subsequently the Q-factor or finesse of the dye laser.

Two Dean flow streams

The Dean flow can be characterized in terms of a dimensionless Dean number \( (De) \), in which the relative magnitudes of inertial and centrifugal forces to viscous forces can be expressed as

\[
De = \frac{\delta d}{Re}
\]

Where, \( \delta = d/R \) is a geometrical parameter with \( d \) and \( R \) being the channel hydraulic diameter and the flow path radius of curvature, respectively. \( Re = Vd/\nu \), where \( V \) is the average flow velocity, and \( \nu \) is the kinematic viscosity of the liquid. When the liquids are fixed, the flow rates of the liquid streams and the geometry of the curved microchannel determine the Dean number and the spatial change of the liquid profile in the microchannel. Two criteria are critical to realize a 3D liquid waveguide by the two flow streams with the inner flow stream being completely surrounded by the outer flow stream: (1) a high Dean number, which leads to larger curvature effects, and (2) longer flow path, which provides longer time for the centrifugal force to act on the liquids. These criteria can be fulfilled by having a substantially large \( R \) across \( 180^\circ \) and by adjusting the flow rates of the liquids. With a fixed microchannel length and curvature, the flow rates play an important role in forming the 3D liquid waveguide. When the flow rates are too low, the outer flow stream does not completely surround the inner flow stream. When the flow rates are too high, the two flow streams are exchanged in position with the similar liquid profile shown in the inset 1 of Fig. 1(a). Therefore, suitable flow rates are required so that the inner flow stream can be surrounded completely by the outer flow stream at the end of the curved microchannel. In the straight microchannel, \( R \) is infinite and results in the absence of the transverse flow \( (De = 0) \). Thus, the liquid profile in the straight channel is kept stable with the inner flow stream surrounded by the outer flow stream as shown in the insets 3 and 4 in Fig. 1(a).

In order to reveal the profile reconfiguration of the two flow streams by the Dean flow after the curved microchannel, the laser scanning confocal microscopy (Carl Zeiss, LSM 710) is conducted to monitor the 3D liquid spatial profiles in the straight microchannel. Two pigtail-coupled lasers with polarization preserving single-mode fibers are inserted to plot the fluorescent images of the two flow streams. An argon-ion laser (514 nm) and DPSS-laser (561 nm) are used to obtain the fluorescent images of the inner injected flow stream (Red, Rhodamine 6G) and the outer injected flow stream (Green, Rhodamine B), respectively. The dye molecules are optically pumped using a mercury lamp (Nikon, C-SHG1) and the 3D structure of the two flow streams

![Fig. 1](image-url)
are scanned using a Z-stacked mode at 2 μm interval with an inverted optical microscope and a 10×/0.3 NA objective lens.

The shape of the fluidic interface can be conveniently altered by changing the rates of the two flow streams. Higher flow rates generate a higher Dean number in the curved microchannel, hence causing stronger fluidic interface reconfiguration. The full 3D fluidic profiles are shown in Fig. 2(a–c), which reveal the reconfiguration processes as the Dean number is increased from 1.4 to 11.2. The ethylene glycol mixture (EG, 75% (CH₂OH)₂ and 25% (CH₃OH) in mass) solution and DI water are at a 1 : 2 ratio of flow rates. When the flow rates of EG solution and DI water are at \( Q_1 = 25 \mu l \text{ min}^{-1} \) and \( Q_2 = 50 \mu l \text{ min}^{-1} \) (Fig. 2(a), \( \text{De} = 1.4 \)), respectively, the centrifugal effect is not sufficient to significantly change the laminar flow profile. The EG flow stream is totally surrounded by the DI water to form a circular profile. The full 3D profiles are seen to be static in the straight microchannel, and the distortion caused by diffusion can be ignored. The EG flow stream from the inner side changes to the outer side and vice versa when the flow rates are increased. When the flow rate is higher, the EG flow stream is totally surrounded by the DI water to form a complete 3D liquid waveguide. When the ratio of the flow rate is changed, from \( Q_1 = 200 \mu l \text{ min}^{-1} \) and \( Q_2 = 600 \mu l \text{ min}^{-1} \) to \( Q_1 = 200 \mu l \text{ min}^{-1} \) and \( Q_2 = 800 \mu l \text{ min}^{-1} \), the diameter of the liquid core is changed from 40 μm to 30 μm. This illustrates the tunability of the 3D liquid waveguide and the nature of the flow liquids can be further applied to control the output intensity of the dye laser.

**Fabry–Pérot microcavity**

In Fabry–Pérot microcavity, standing waves can be formed for standing longitudinal waves, thus for wavelengths with \( m \lambda = 2nL \), where \( m = 1, 2, 3, ..., n_1 \) and \( L \) are the refractive index of the core liquid and the length of the FP microcavity. As a result, the enhanced intensity of the light causes lasing by the constructive interference that occurs in the FP microcavity. The FP transmission spectrum can be expressed as:\(^{20,21}\)

\[
T = \frac{(1 - R_1^2)/(1 - R_2^2)}{1 + 4R_1R_2/(1 - R_1^2)\sin^2 n_1L\pi/\lambda}
\]

where \( R_1 \) and \( R_2 \) are the reflectivity of the two FP mirrors. For example, there are more than 20 longitudinal modes in the range from 570 to 580 nm for laser application with the parameters of \( R_1 = 85\%, \ R_2 = 99\%, \ L = 320 \mu m \) and \( n_1 = 1.410 \). The longitudinal modes position is only dependent on the effective cavity length \( n_1L \).

**2D versus 3D optofluidic dye laser**

In a 2D liquid–liquid waveguide, the core flow stream is horizontally sandwiched by two cladding flow streams.\(^6\) However, the core flow stream is in contact with the vertical channel walls. The refractive index of the core flow streams is lower than that of the PDMS and optical leakage is significant from the core flow stream to the PDMS channel wall. The light fields in the 2D liquid–liquid waveguides and the 3D liquid–liquid waveguides are simulated using beam propagation techniques, and the results are depicted in Fig. S1 of the ESI\(^\dagger\). In the simulation, the microchannel has dimensions of 100 μm × 100 μm × 4 cm. The refractive index of the liquid core, liquid cladding and solid channel wall are set as \( n_1 = 1.410, \ n_2 = 1.332 \) and \( n_3 = 1.410 \). The light source has a wavelength of 575 nm in simulation and the absorption of the materials is ignored. The simulation result of the 2D liquid–liquid waveguide suffers strong intrinsic optical leakage (2.8 dB) in the cross-sectional (Fig. S1(a)\(^\dagger\)) and the propagation direction (Fig. S1(c)\(^\dagger\)). Compared to the 2D liquid waveguide, the 3D liquid waveguide overcomes this drawback because the light is well confined in the liquid core as shown in Fig. S1(b) and (d)\(^\dagger\). As a result, the 3D optofluidic dye laser has higher output laser energy and a lower threshold level.

The output lasing energy can be tuned by changing the volume ratio between the two flow streams. The variation of the laser intensity as a function of the central stream volume ratio for both 2D and 3D liquid waveguides can be calculated by considering the gain and losses of the dye lasers.\(^4\) Under lasing conditions, the gain of the system compensates the losses, i.e. \( gL = \rho \) where \( g \) is the gain of the lasing medium, \( L \) is the length of the lasing medium, \( \rho \) is the loss of the lasing cavity and \( \rho \) is the loss of the lasing cavity. Zero absorption of the sample solution is assumed and the cavity loss of the lasing cavity is mainly due to the mirrors’ reflectance \( (R_1 \text{ and } R_2) \) and the loss of the waveguide \( (\rho_\omega) \), i.e. \( \rho = \rho_\omega - In\sqrt{R_1R_2} \). The gain of the waveguide

**Fig. 2** The confocal microscopy images of the fluidic profiles when the flow rates (a) \( Q_1 = 25 \mu l \text{ min}^{-1} \) and \( Q_2 = 50 \mu l \text{ min}^{-1} \), (b) \( Q_1 = 100 \mu l \text{ min}^{-1} \) and \( Q_2 = 200 \mu l \text{ min}^{-1} \), (c) \( Q_1 = 200 \mu l \text{ min}^{-1} \) and \( Q_2 = 400 \mu l \text{ min}^{-1} \), (d) \( \phi_1 = 40 \mu m, Q_1 = 200 \mu l \text{ min}^{-1} \) and \( Q_2 = 600 \mu l \text{ min}^{-1} \), (e) \( \phi_2 = 30 \mu m, Q_1 = 200 \mu l \text{ min}^{-1} \) and \( Q_2 = 800 \mu l \text{ min}^{-1} \).
The equations, the laser emission intensity ratio of 2D and 3D waveguide can be derived as

\[ \frac{I_{3D}(\gamma_1)}{I_{2D}(\gamma_2)} = \frac{\gamma_3D \left( g_0L - \rho_{w,3D} + In(R_1 + R_2) \right) \rho_{w,2D} - In(R_1 + R_2)}{\gamma_2D \left( g_0L - \rho_{w,3D} + In(R_1 + R_2) \right) \rho_{w,2D} - In(R_1 + R_2)} \]  

where \( \rho_{w,2D}, \rho_{w,3D} \) and \( \gamma_2D, \gamma_3D \) are the single pass loss and the central stream volume ratio of the 2D and 3D waveguide dye lasers, respectively. According to eqn (3), the relationship between laser emission intensity for the two configurations is determined only by the waveguide loss with the same central stream volume ratio and under the same optical pump, the optical loss of 2D waveguide is much higher than that of the 3D waveguide when \( \gamma_{2D} = \gamma_{3D} \). Therefore, it is easy to understand that the 3D waveguide configuration can achieve higher laser emission intensity as compared to the 2D waveguide.

Based on the lasing rate equations, which describe the time evolution of the energy level populations and the laser cavity mode, the threshold level can be estimated. The triplet state effects can be neglected as a laser pump with pump pulse duration (6 ns) shorter than the inter-system crossing time (\( \sim 100 \) ns) is used. In addition, the self-absorption at the lasing wavelength is assumed to be low. The threshold population inversion density with the cavity loss is determined as

\[ I_{th} = \frac{(\rho_s - h\nu_p d)\sigma_em}{\tau_{sp}L} \]  

where \( h \) is the Planck constant, \( \tau_{sp} \) is the spontaneous emission lifetime, \( \sigma_em \) is the stimulated transition cross section at the lasing wavelength, and \( d \) is the penetration depth of the pump light into the dye solution, given by \( 1/n_0s\sigma(d) \), \( n_0 \) is the ground state population density and \( \sigma(d) \) is the absorption cross section at pump wavelength. In this case, the laser pump has a focusing geometry in which the penetration depth of the pump light can be considered to be the same for both 2D and 3D waveguide dye lasers. From eqn (4), it is clear that the 3D waveguide configuration has a lower threshold level for laser emission because of its high confinement. For \( \sigma_{em} = 1 \times 10^{-16} \text{ cm}^2 \), \( \sigma(d) = 3.8 \times 10^{-16} \text{ cm}^2 \), \( \tau_{sp} = 5 \text{ ns} \) and \( L = 320 \mu \text{m} \). The calculated threshold is approximately 10 kW cm\(^{-2}\) for the 3D waveguide dye laser while 19 kW cm\(^{-2}\) for the 2D waveguide dye laser. In practical systems, the threshold intensity is usually much higher due to other factors such as self-absorption or thermal distortions.

Besides higher output energy and lower threshold level, the circular 3D liquid waveguide has also a more uniform cross-sectional profile than its 2D equivalent. The uniform cross-sectional profile is another advantage of the 3D liquid waveguide. Fig. S2 of the ESI† shows the calculated relationship between the volume ratio of the liquid core and the parameters \( \phi \). The volume ratios of the liquid core flow stream are calculated as \( \pi \phi^2/(2W)^2 \) for the 3D dye laser and \( \phi/W \) for 2D ones. \( \phi \) represents the liquid core flow stream diameter of the 3D liquid–liquid waveguide and \( \phi \) is the width of the sandwiched core flow streams of 2D liquid–liquid waveguide, \( W \) is the width of the microchannel. For example, when the volume ratio is 0.2, the diameter \( \phi \) of 3D liquid waveguide is approximately 50 \( \mu \text{m} \), while the core width \( \phi \) for 2D liquid waveguide is only 20 \( \mu \text{m} \). Such an ideal circular waveguide is a better design than the 2D waveguide for optofluidic dye laser application because of its isotropic properties.

**Experimental results and discussions**

The optofluidic dye laser chip is fabricated using the standard soft lithography process. First, a 100 \( \mu \text{m} \) layer of SU8 photoresist (Micro-Chem, SU8-50) is spin-coated onto a silicon wafer. After pre-baking, the master is exposed to UV light under a glass mask using a mask aligner (OAI, 506). Then, microchannels are moulded using PDMS (Dow corning, Sylgard 184) and sealed against a flat PDMS sheet after plasma oxidation. The fabricated PDMS chip is stored in an oven at 75 °C for at least 24 hours to regain its hydrophobicity. The advantages of PDMS materials are transparency at visible wavelengths and minimal absorption loss.

Fig. 3 shows the microphoto of the fabricated optofluidic chip. The microchannel has dimensions of 100 \( \mu \text{m} \times 100 \mu \text{m} \).
curved region, the microchannel is bent with $R = 2 \text{ mm}$ across $180^\circ$, while in the straight region, the length of the straight microchannel is $L = 320 \text{ mm}$. A small bend with $R = 200 \text{ mm}$ between the two regions is used to connect them smoothly. The two flow streams are pumped into the microchannel from the inner and the outer inlets using syringe pumps (Genie, Kent Scientific Corporation, CT). In the experiment, the chosen liquids are EG as the inner liquid and DI water as the outer liquid, as they do not swell PDMS. In addition, they are good solvents for fluorescent dyes. For initial characterization, the inner flow streams contained Rhodamine 6G with a concentration of 2 mM as the gain medium. The refractive index of the inner flow streams is $n_1 = 1.410$ (75% (CH$_2$OH)$_2$ and 25% (CH$_3$OH) in mass), which is equal as PDMS. The refractive index of outer flow stream is $n_2 = 1.332$. The refractive indices of the liquids and PDMS are measured by the refractometer (Reichert, AR200 digital hand-held).

For efficient light coupling and easy microfluidic manipulation, an FP cavity with Au-coated mirrors is fabricated on the facets of the two fibers by electron beam evaporator, and then aligned in the microchannel. The mirror $M_1$ is coated with 80 nm Au, and has a reflectance of approximately $R_1 = 85\%$. The mirror $M_2$ is coated with 160 nm Au, and has a reflectance $R_2 = 99\%$.

The pump laser (532 nm, 6 ns, 1 Hz) is a frequency-doubled Nd:YAG laser (Orion, New Wave Research, Fremont, CA). Varying the Q switch delay, the energy of the laser pulse can be controlled.

Fig. 4 shows the output spectrum of the optofluidic dye laser and the emission spectra of the dye Rh 6G (2 mM). The intensity is measured by a miniature fiber optic spectrometer (Ocean, USB 2000). Without the Au mirrors to support the feedback, the

![Fig. 4](image1.png)

**Fig. 4** The output intensity of the waveguide dye laser (Red) and the emission spectrum of the dye Rh 6G with the concentration 2 mM.

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![Fig. 5](image2.png)

**Fig. 5** (a) The laser output intensities at different core diameters from 40 µm to 80 µm. (b) The wavelength shift by changing the core diameter. (c) The output intensity as a function of the volume ratio between 2D and 3D liquid waveguide dye laser. (d) The threshold of 2D and 3D liquid waveguide dye laser at a volume ratio of 0.2.
spontaneous emission of the Rh 6G in EG solution is observed. The full width half maximum (FWHM) is 40 nm. However, when the FP cavity is formed, the laser emission of the 3D liquid waveguide laser is emitted at 575.6 nm (red). The FWHM of the waveguide laser is improved by 10-fold to 3 nm. The dye laser is a multimode laser consisting of many longitudinal modes. The spectra resolution of the spectrometer does not allow us to resolve the longitudinal multimodes of the present laser system. Correspondingly, one can estimate an overlap of longitudinal modes in the observed laser emission peak. The $Q$-factor is approximately 190 by measuring the emission spectrum (peak wavelength/FWHM).

The laser outputs at different core diameters ranging from 40 μm to 80 μm are shown in Fig. 5(a). The output laser intensity can be tuned by changing the diameter of the liquid core to control the gain medium. It can be concluded that the emission intensity is increased by increasing the diameter of the liquid core. Moreover, the water gap between the fiber mirror $M_1$ and the gain medium at the end of the bend is varied by different diameters of the liquid core when the flow streams are flowing into the FP cavity, resulting in the lasing wavelength being tuned by the core diameter. Fig. 5(b) shows the shifting of the lasing wavelength in the 3D liquid dye laser by forming a different core diameter. The laser output is normalized to show the wavelength shifting prominently. It can be seen that the peak wavelength is shifted by 1.7 nm when the core diameter is tuned from 40 μm to 50 μm.

Fig. 5(c) shows the comparison of the output energy as a function of the central stream volume ratio between 2D and 3D liquid waveguides, showing approximately a linear relationship. The 3D liquid waveguide laser has higher output intensity than the 2D equivalent at the same pump energy. The output intensity of the 3D dye laser can be achieved 6-fold higher than the 2D equivalent at the pump intensity of 50 μJ per pulse when the volume ratio is fixed at 0.5. This shows that the 3D liquid waveguide dye laser has a higher efficiency than its 2D equivalent.

Besides higher output intensity, the 3D waveguide dye laser also excels in terms of threshold level. Fig. 5(d) shows the threshold levels of 2D and 3D liquid waveguides at a volume ratio of $0.2 (\phi = 50 \mu m, \phi' = 20 \mu m)$. The threshold level of the 3D liquid waveguide dye laser is measured as 16.5 μJ mm$^{-2}$ while the 2D is measured as 28.5 μJ mm$^{-2}$. It is clear that the threshold level of the 3D dye laser is only 60% of the 2D dye laser under the same conditions. Besides, the slope efficiency is also 3-fold higher than that of 2D ones at this condition. The energy efficiency of the 3D waveguide dye laser is estimated to be 5%.

In fact, the 3D optofluidic waveguide dye laser based on the centrifugal Dean flow is versatile. Some materials, such as ethanol, methanol or some mixtures, are important in laser applications because they are very good solutions for dyes. These solvents, however, cannot be used in traditional 2D waveguides because their refractive indices are lower than the solid cladding. This design solves this problem because with these materials, it is not necessary to be in contact with the solid cladding. Consequently, the confinement is much greater, and the dye laser is more efficient.

**Conclusions**

In conclusion, a tunable 3D optofluidic waveguide dye laser based on centrifugal Dean flow is demonstrated. The tunable optofluidic dye laser consists of two main parts: (1) a 3D liquid waveguide with dye dissolved in the liquid core is constructed by two Dean flow streams as the gain medium, and (2) a FP micro-cavity is formed by two Au-coated mirrors, which is fabricated on the facets of the two fibers and aligned in the microchannel to provide optical feedback. The 3D tunable optofluidic waveguide dye laser is much more efficient in obtaining a much lower lasing threshold (60%) and higher output energy with a slope efficiency (demonstrated 3-fold) than the traditional 2D equivalent. It has a real-time tunability in varying the laser output energy by changing the flow rates of the two flow streams with the $Q$-factor of approximately 190. The developed 3D optofluidic waveguide dye laser provides a versatile platform for microfluidic biosensors and bioanalysis in a liquid environment.

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**Notes and references**