CHAPTER 1

INTRODUCTION

1.1 INTRODUCTION TO DYE LASERS

The invention of dye laser has made a tremendous impact on science and technology. It has fulfilled the dream of laser experimenters: to have a laser that is easily tunable over a wide range of frequencies or wavelengths [1]. Dye laser-based research has grown rapidly, due to the considerable interest in the quantum electronics and the unique applications in various fields of physics, chemistry or biology. Laser dyes can be used in the solid [2-9], liquid [10-16] or gas phase [17, 18], where their concentration are easily controlled, and thus allowing their absorption and gain to be manipulated. These fluorescent organic compounds absorb in the near ultraviolet to the visible and they have broad emission bandwidths ranging from 340 nm – 1200 nm, and as such their attention as tunable lasers. Thus, dye lasers have developed and become important in spectroscopic diagnosis [19-22], holography [23, 24], imaging [25], communication [26, 27], environmental [28, 29], photo-dynamic therapy (PDT) [30, 31] and atomic physics [32].

1.2 HISTORICAL REVIEW OF DYE LASER

The first experimental study led to the realization of an organic laser was carried out by Stockman et al. in 1964 [42]. Stockman found an indication of a small net gain in his system when he used a high-power flashlamp to excite a solution of perylene in benzene between two resonator mirrors. Unfortunately, Stockman was unable to obtain laser action
from this aromatic molecule, which has high losses due to triplet-triplet absorption and absorption from the first excited singlet into higher excited singlet levels.

In 1966, Sorokin and Lankard \cite{43} reported the first stimulated emission from an organic compound at IBM's Thomas J. Watson Research Center. This stimulated emission was observed from an alcoholic solution of phthalocyanine dye (chloro-aluminium phthalocyanine) pumped by a Q-switched ruby laser ($\lambda = 694.3$ nm). Stimulated emission was also observed from polymethine dyes (1, 1'-diethyl-2, 2'-dicarbocyanine iodine, and 1, 1'-diethyl-4, 4'-carbocyanine iodine) by Spaeth and Bortfeld \cite{44}. In the same year, independently, Schäffer, Schmidt and Volze \cite{45} obtained the first evidence that the laser wavelength of the dyes can be shifted over more than 60 nm by varying the dye concentration or the resonator mirror reflectivity. Since then, stimulated laser emissions were observed for many classes of organic compound \cite{11, 46-49} and the dye lasers have covered over a wide spectral range from the near ultraviolet to the infrared.

In 1967, Soffer and McFarland \cite{3} used a diffraction grating to substitute one of the resonator mirrors to introduce wavelength-dependent feedback. They obtained a continuous tuning range of 45 nm and the effective spectral narrowing from 6 to 0.06 nm. However, the peak power of the dye laser was reduced due to low efficiencies of grating. Furthermore, high power and energy pulses may damage the grating. An improvement in this aspect was introduced by Kogelnik et al. \cite{50} by using holographically produced bleached transmission gratings. This problem can also be overcome by the use of prism beam expanders (Fig. 1.1). It can be a single prism \cite{12, 51}; or more complicated designs using several prisms in series \cite{52-53}. 
Fig. 1.1 (a) Single prism as beam expander; (b) double prism as beam expander.

Another important improvement was developed by Shoshan et al. \cite{54}, Littman \cite{55} and Saikan \cite{56} at the end of 1970s, where a grating in grazing incidence was used in combination with a high reflectivity mirror (Fig. 1.2(a)), instead of using intracavity beam expanders. This arrangement was improved by Littman \cite{57} when he used a second grating to replace the high reflectivity mirror (Fig. 1.2(b)). In such an arrangement, single-mode operation could easily be achieved.
Fig. 1.2 (a) Grazing incident laser-pumped tunable dye laser; (b) grazing incident laser-pumped tunable dye laser with a second grating.

Due to the extremely broad spectral bandwidth of dye lasers, it is capable of producing ultrashort pulses with smaller half-width compared any other laser. The first picosecond dye laser pulses was achieved in 1968 \cite{58-60}, where a mode-locked pulse train from a solid-state laser was used to pump a dye solution. In 1974, optical pulses shorter than 1 ps were achieved by Shank and Ippen \cite{61} with a passively mode-locked dye laser. Another important technique for the production of ultrashort pulses is by distributed feedback (DFB) dye laser \cite{62-64}. Femtosecond pulses became reality in 1980s with the introduction of pulse compression technique \cite{65-68}. This nonlinear optical method brought the achievement of ultrashort pulsewidth to 6 fs \cite{69}.
Up to date, liquid dye lasers remain the most widely used dye laser systems. This is due to its high laser efficiency, continuity and wide range tunability. However, these liquid dye laser systems are bulky in size in order to accommodate the flow of dye solutions through the laser cavity. The flow of dye solutions through the laser cavity helps to maintain the laser performance. This brings about high operation costs, in addition to produce large quantities of toxic and flammable waste. Thus, solid-state dye lasers are attractive alternative to conventional liquid dye laser systems. The advantages of solid-state dye lasers over the conventional liquid dye lasers will be discussed in Chapter 2.

1.3 OBJECTIVES OF THIS PROJECT

In this work, we studied the laser properties of Coumarin 460 (C460) and Rhodamine 6G perchlorate (R6G(ClO₄)) doped in poly(methyl methacrylate) (PMMA) and poly(vinyl alcohol) (PVA) respectively. These dye-doped polymers were excited by a high power Transversely Excited (TE) nitrogen laser. The performance of both organic dyes in these two host materials had been examined by measuring the laser wavelength shift, laser efficiency and operating lifetime. At the same time, we also studied the optical properties of these dye-doped polymers and hence optimized the performance of the dye cells by varying the dye cells configuration and parameters during preparation process.

1.4 STRUCTURE OF THE DISSERTATION

This dissertation consists of six chapters. In chapter 2, the principles of dye lasers and discussion on laser dyes are presented. The operation of dye lasers by different pumping methods will be discussed. Besides, a review of solid-state dye lasers is also presented in this chapter. In chapter 3, a brief discussion on host materials of solid-state dye lasers is given. Following this, the fabrications of dye-doped polymers are described.
These include the optimization of the parameters of the preparation process and the configuration of dye cells.

Chapter 4 presents the experimental setup and measurement devices of this research. Besides, the optical methods were in the determination of the refractive index and the thickness of these dye-doped polymers are described. All the experimental results and discussions are given in chapter 5. These include the measurements of laser wavelength shifts, laser efficiencies and operating lifetimes of the dye-doped polymers. The dependence of laser performance of the dye-doped polymer film on film thickness, aluminium-coating on glass substrate and glass substrate thickness are also described in this chapter. Finally, in chapter 6, conclusions and suggestions of future work are presented.