DNA Nano-circuit for Electronics

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Abstract

This paper describes preparations of nano-scale patterned electric circuit based on high purity DNA molecules which are obtained from Salmon roe. The patterning on silicon substrate was carried out by an ink-jet method of aqueous solution of DNA. However, the Salmon-based DNA has a so huge molecular weight of over billions that even only 1wt% aqueous solution of DNA becomes gel without any fluidity. So, it is necessary to reduce molecular weights of DNA to increase fluidity of the aqueous solution of DNA, keeping the characteristic feature of double helical structures of DNA molecules to form metal-chlating complexes with various metal cations such as silver or copper cations.

Several methods to reduce the molecular weight of DNA, including hydrolytic , enzymatic degradations and ultra-sonification. It was found that the best method to reduce the molecular weights (MW) of DNA was an enzymatic degradation of DNA to increase fluidity, thus being able to apply an ink-jet method for nano-scale patterning on a silicon wafer to form DNA circuit, followed by ion-crosslinking of DNA by dipping the patterned DNA circuit into aqueous solution of copper chloride. The DNA-$CuCl_2$ patterned circuit was reduced to copper nano-lines for electric circuit, by using hydrazine as a reducing agent. Thus, all DNA devices can be prepared by the combination of DNA transistor and circuit.

1. Introduction

Deoxyribonucleic acid (DNA) is the source of all livening things in the earth which carries all of gene information. DNA has a huge molecular weight of over billion and is possible to form films. The characteristic feature of DNA is an intercalation of aromatic dyes into the double helical structure of DNA, resulting in a large enhancement of fluorescence light and electronic activities, and applications of DNA as photonics and electronics have been expanding during recent 15 years.[1-9)]

DNA is water-soluble with sodium counter ions, which are not appropriate for applying DNA to material sciences such as electronic devices. However, DNA molecules become insoluble in water, yet become soluble in polar organic solvents such as ethanol, when

Nanobiosystems: Processing, Characterization, and Applications V, edited by
Norihisa Kobayashi, Fahima Ouchen, Ileana Rau, Proc. of SPIE Vol. 8464, 846403

sodium cations are replaced with quarternized ammonium salts, lipids which contain long alkyl chains to form DNA-lipid complexes, and clear and tough films are easily obtained by solvent casting of ethanol solutions1[7)]. Recent research results on DNA-lipid complexes have shown various attractive applications such as E/O or O/E devices, optical memories, switches and sensors1[-9)]. Thus, applications of DNA photonic devices have been widely studied in the world.

However, problems of DNA optical devices are related to moisture absorption of DNA molecules which are very much hydrophilic, and adsorbed water influences the dye-intercalated structures of DNA molecules. Therefore, it is necessary to protect the dye-intercalated state of DNA molecules by sealing off water penetration. It was reported by us[9)] that a novel hybridization method of the dye intercalated DNA molecules by means of so-called so-gel process was effective to increase stabilities and durability of DNA photonic devices under environmental changes. Also, hybridizations of DNA-lipid complexes which were intercalated with optical dyes were successfully carried out by blending synthetic polymers such as poly(methylmethacrylate or polycarbonate) by a solution blending method.

This paper describes further applications of DNA for material sciences such as photonics and electronics and bio-medicals, so that nano-circuit patterns were printed on a silicon wafer by an ink-jet process, followed by chelation with copper ions to prepare nano-scale copper circuit by reduction of copper ions.

2. Experiments

2.1 DNA

Pure DNA was obtained by an enzymatic separation and purification of DNA from Salmon roe, as described before[1)].

2.2 Molecular weight (MW) control of DNA

Following methods were applied to reduce molecular weights of pure DNA, as one wishes.

(1) Sonification: to reduce MW.

Sonification of aqueous solutions of pure DNA was carried out by using various sonic energy by HITACHI sonification instrument as follows

(2) Hydrolytic degradation of DNA in alkaline solution.

Pure DNA was dissolved in pure water in an amount of 1wt%, followed by adding various amounts of NaOH to adjust concentration of NaOH from 1N TO 5N. The DNA alkaline solutions were heated at 80^{o}C for one day, followed by adding excess amount of ethanol to precipitate DNA which was separated by filtration.

(3) Enzymatic degradation of DNA in aqueous solutions.

The enzymatic degradation of DNA was carried out as described as follows:

Preparation of aqueous solution of DNA

① 2%DNA/H_20、② 3％DNA/H_2O、③ 2％DNA/PBS、④ 2％DNA/DMEM (Ｎｏ FBS)、

⑤ 4％DNA/DMEM(FBS)、⑥ 2％DNA/10%FBS/H2O

＊Apparent solution viscosity；②＞①＝⑤＝⑥＞③＝④

Degradation by Fetal Bovine Serum

Dulbecco Modified Eagle Medium (DMEM) 500ml

Fetal Bovine Serum Qualified (FBS) 55ml

Penicillin-Streptomycin (AB) 6ml

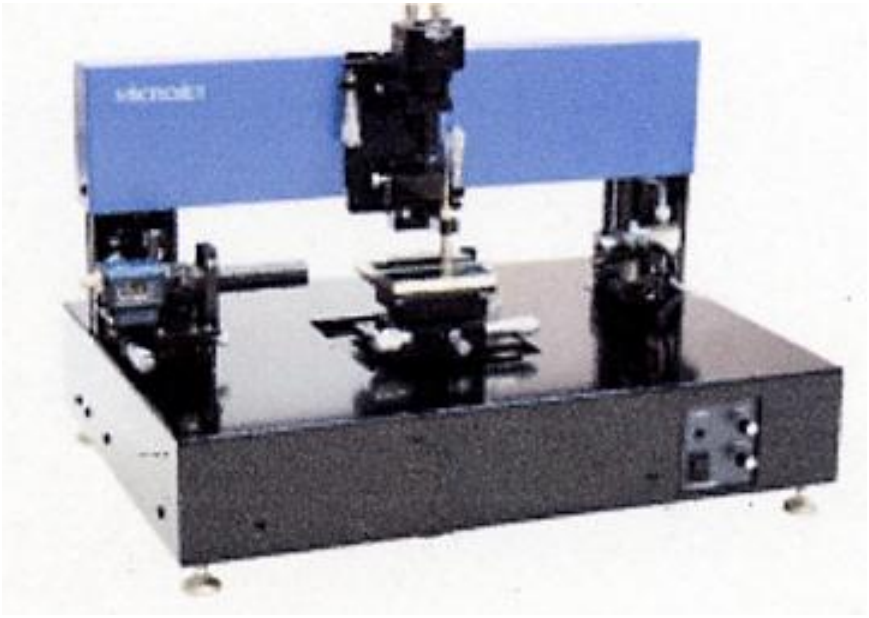
DMEM 500ml 10% FBS、1% AB

Precipitation of DNA

Enzymatic degraded DNA was recovered by pouring excess amount of ethanol to the aqueous solution to precipitate DNA, followed by drying.

2.3 Patterning by Ink JET method

MW-degraded DNA was dissolved in pure water in an amount of 1wt% to use as an ink for the Jet printing. A Silicon wafer was used to print circuits by the Ink Jet instrument Labo-jet 3000 made by the Microjet Co. was used for circuit pattering. The DNA ink was jetted on a Silicon wafer through a nozzle of the ink-jet machine to draw a circuit pattern, followed by drying at 50^0C. The circuit=pattered wafer was dipped into 1N aqueous solution of copper chloride in order to introduce chelation of copper ions with DNA. Reduction of the chelated copper ions was carried out by dipping the Cu ion-chelated circuit wafer into 1N hydrazine aqueous solution at room temperature for one day, followed by washing with ethanol and drying.



Labo-jet 3000 instrument

３． Results

3-1 Degradation of DNA by sonification

Results of the decrease of MW of DNA by various sonification energy are shown in Figure 1, where it is clearly seen that the MW of DNA decreased with increasing sonification energy.

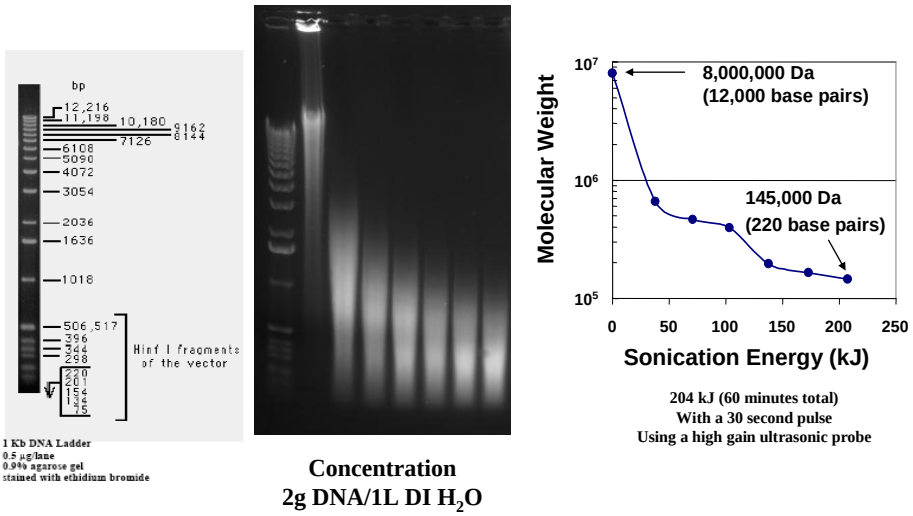


Figure 1 DNA MW decrease with different sonification energy

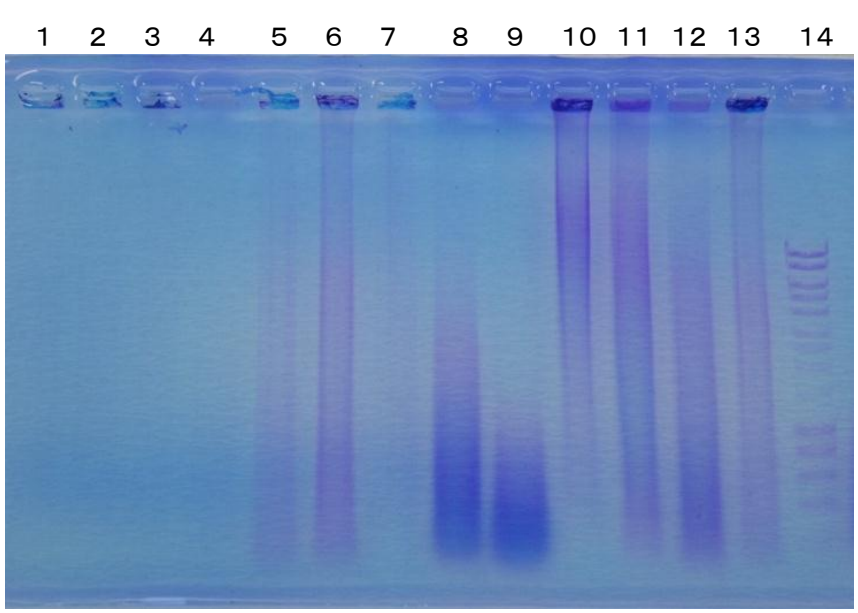
However, circular dichromic (CD) spectra of the MW-decreased DNA did not show any positive and negative Cotton effects, so the double helical structure of DNA would have been cleaved by sonification which would cut off hydrogen-bonding between Ademin-Thymin (A=T) and Guanin-Cytocine (G-C) in the double helix of DNA, which is essential for photonic and electronic amplification of dyes. Therefore, the MW decrease of DNA by sonification needs more precise controls of sonic energy.

3.2 Hydrolytic degradation of DNA

Hydrolytic degradation of DNA molecules was carried out in various concentrations of sodium hydroxide aqueous solutions, followed by precipitation of DNA by excess amount of ethanol and drying. Apparently, MW of DNA decreased by the hydrolytic method, while CD spectra of the degraded DNA did not exhibit any positive and negative Cotton effects, indicating the double helical structure of DNA was not kept. So, this hydrolytic degradation of DNA is not appropriate for DNA devices.

3.3. Enzymatic degradation of DNA

Results of MW control of DNA by enzyme, FBS are shown in Figure 2 as results of electrophoresis, It is clearly seen in Figure 2 that MW of DNA decreased as functions of applied days.



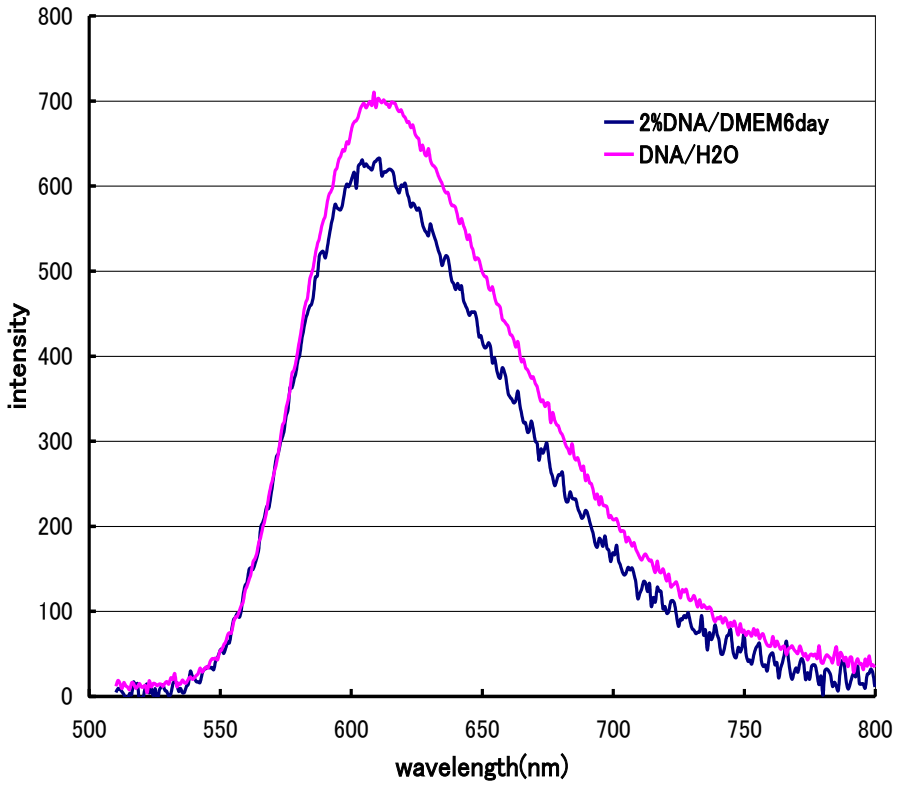
. Figure 2 Electrophoresis diagram of DNA

Apparent solution viscosities of the 1wt% DNA aqueous solution decreased as summarized in Table 1. After 7 days of the FBA treatment, the DNA aqueous solution viscosity decreased to 55 poise, which would be enough to be applied to the ink-jet patterning process. Fluidity of the aqueous solution of WM control DNA is dependent on the days applied by FBS at room temperature and 5~7 days would be good enough to apply as a DNA ink for the ink-jet method.

Table 1 Viscosity changes of the FBA-treated DNA aqueous solution

Time (day)	Viscosity (poise)
0	Gel, no fluidity
3	500
5	300
6	100
7	55

CD spectra of the FBS-treated DNA are shown in Figure 3, which indicates almost no changes of the CD spectra, indicating the preservation of the double helical structure of DNA after the cutting of DNA by FBS.



Figured 3 CD spectra of DNA after enzymatic treatment

Figure 4 shows an example of DNA circuit by the ink-jet method drawn n a Silicon wafer. Electrical conductivity of the circuit was equivalent to a Cu line so that the DNA nano-circuit was obtained. Thus, all DNA-based bio-electronics open the door to a new world for applications.

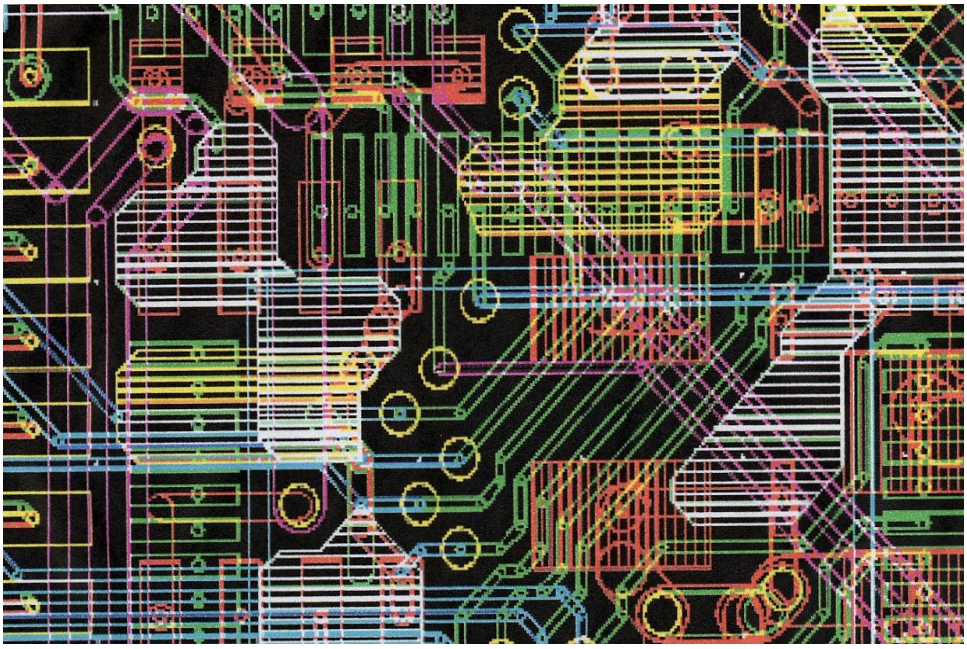


Figure 4 Nano-circuit pattern drawn by DNA ink

Summary

Nano-circuit by an ink-jet method by using MW-controlled DNA was successfully prepared. The best method to reduce the MW of DNA is that the addition of Fetal Bovine Serum (FBS) to 3wt% DNA aqueous solution at 37^{o}C for 5 days without stirring, followed by adding ethanol to precipitate the MW-reduced DNA which was 1/3. The MW-reduced DNA was applied as an ink for an ink-jet method to draw a circuit pattern on a Silicon wafer, followed by dipping into Cu ion chelation and reducing the Cu ions to Cu.

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