SPACE CHARGE BEHAVIOR IN POLYMERIC INSULATING MATERIALS UNDER DC STRESS AND PRE-BREAKDOWN PHENOMENA

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Abstract

Space charge has been recognized as an agent of insulation degradation for power transmission cable especially under high voltage dc application. It penetrates into bulk of insulating material, namely cross-linked polyethylene (XLPE), by time during dc voltage application. The increase in space charge density enhances local electric field in the insulating material. In other words, the local electric field is distorted. When the field distortion reaches a critical value, dc breakdown of the insulating material will occurs, causing a catastrophic damage to the cable. Therefore, it is important to understand space charge inside an insulating material in order to improve the reliability of next generation power cables.

In this thesis, pulsed-electroacoustic (PEA) method is utilized in order to probe space charge behavior inside an insulating material under high voltage dc voltage application. This method is a highly proven method in probing space charge behavior in a dielectric material. Furthermore, it is also applicable to on-site cable assessment. In order to improve the reliability of the data, space charge behavior is simultaneously measured with external circuit current, which is partially corresponded to the space charges dynamic. The objectives of this thesis is to investigate space charge behavior in an insulating material (namely low-density polyethylene (LDPE), which is used as insulating material for medium voltage power cables), and to clarify how dc breakdown occurs after space charges accumulate in insulating material.

It has been postulated by several researchers that space charges travel inside the bulk of insulating material through the free-volume. Free-volume is free space between polymer chain of LDPE material. It is believed that, penetration and accumulation of space charge in LDPE will suppress the free-volume size. This is considered by the fact that Maxwell stress is enhanced with the increase of space

charge accumulation in the bulk of LDPE sample. However, free-volumes are too tiny to be observed. Therefore, density of a LDPE material, which is related to the amount of free-volumes, is altered. A 3-dimensionally branching chain material, namely polyisobutylene (PIB), is added into LDPE in order to reduce its density, whereas, a low molecular weight material, namely paraffin wax (Pr), is added into LDPE in order to increase its density. From results, it has been found that under dc positive high voltage application, the penetration of positive space charges are suppressed under Pr-added LDPE, whereas the penetration of positive space charges are increased in PIB-added LDPE sample. It implies that the alteration that is made to micro-structure of LDPE affect the penetration behavior of space charge.

Matsui et al. postulated that penetration of space charges are prevented when it reaches a certain penetration depth inside the bulk of LDPE material. The penetration depth decreases with the increase of high voltage dc stress value. They explain that there are 2 regions exist when the penetration space charges is prevented; high conductivity zone, which is behind space charge closer to anode, and low conductivity zone, which is in front of space charge and closer to cathode. Similar phenomenon was also observed in LDPE with no additive sample and Pr-added LDPE sample. It is unusual to observe such phenomenon. It is like there are another layer of dielectric in front of space charge, which work as barrier, causing the charges to stop penetrating further. From this point of view, observation to clarify where breakdown is initiated, are carried out. A sample composed of 2 layer of different dielectric material (liquid and solid (this film)) is prepared, so that the injected charges may accumulate at interface of the 2 materials. It is important from there to clarify where breakdown will occurs. From the obtained results. breakdown does not necessarily occurs at the film. Sometimes it was initiated at other places.

Declaration

The contents of this thesis are the results of original research and have not been submitted for higher degree to any other university or institution. Much of the work presented in this thesis has been published as journal or conference papers. The following is a list of these publications:

Journals

- A. I. Mohamed, M. Morimoto, T. Akagi, K. Kadowaki and I. Kitani, "Simultaneous Measurements of Space Charge and External Current for LDPE Films with Various Densities," *IEEJ Transaction on Fundamentals and Materials* , Vol. 131, No. 12, pp. 1-6, 2011.

- A. I. Mohamed and K. Kadowaki, "Streak Observation System for DC Pre-breakdown Using an Image Guide Scope," *Japanese Journal of Applied Physics* (JJAP), Vol. 51, Issue 24, pp. 028003-028003-2, 2012.

International Conferences

- A. I. Mohamed, Y. Miyamoto, M. Mori and K. Kadowaki, "Streak Observation of DC Pre-breakdown Light in Silicone Oil / Low-density Polyethylene (LDPE) Film Composites Using a Long Image Guide Scope," In *Proc. of the International Conference on Dielectric Liquid* (ICDL 2011), Trondheim, Norway, June, pp. 57-62, 2011.

- A. I. Mohamed, M. Mori and K. Kadowaki, "DC Pre-breakdown Light in Hexane/low-density Polyethylene(LDPE) Insulation Composites using a long Image Guide Scope and a Streak Camera," In *Proc. of the 12th International Symposium on Electrical Insulating Material* (ISEIM 2011), Kyoto, Japan, September 13-15, pp. 406-409, 2011.

- T. Akagi, Amir I. Mohamed, and K. Kadowaki, "Long Time-Range Breakdown Caused by Penetration of Positive Charges Packet in Low Density Polyethylene Sheets," In *Proc. of the Asian Conference on Electrical Discharges* (ACED 2012), Johor Bahru, Malaysia, December 10-12, 2012.

- Amir I. Mohamed, Y. Miyamoto, and K. Kadowaki, "Streak Observation of Dc Pre-breakdown Phenomenon in Silicone Oil / Low Density Polyethylene (LDPE) with Different Viscosities," In *Proc. of the Asian Conference on Electrical Discharges* (ACED 2012), Johor Bahru, Malaysia, December 10-12, 2012.

Local Conferences

- Amir Izzani Mohamed and Kazunori Kadowaki, "Simultaneous Measurement of Space Charge and External Current in LDPE Sheets with Different Densities," In *Proc. of the 41^{th} Symposium on Electrical Insulation Material System*, Akita, Japan, 2010.

- Yuuji Miyamoto, Amir Izzani Mohamed and Kazunori Kadowaki, "Observation of DC Prebreakdown in Silicone-Oil /Low Density Polyethylene Composite under Homogeneous Field," In *Proc. of the 41^{th} Symposium on Electrical Insulation Material System*, Akita, Japan, 2010.

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- Y. Miyamoto, Amir Izzani Mohamed and K. Kadowaki, "Observation of DC Pre-breakdown in Liquid dielectric Using Long Image Guide Scope," In *National Conference of the Institutes of Electrical and Engineers Japan*, Japan, 2011.

- Y. Miyamoto, Amir Izzani Mohamed and K. Kadowaki, "Observation of DC Pre-breakdown in Silicone-Oil/Low Density Polyethylene Composite Using Long Image Guide Scope and Streak Camera," In *Electrical Discharges Workshop*, Ehime, Japan, 2011.

- Y. Miyamoto, Amir Izzani Mohamed and K. Kadowaki, "Streak Observation of DC Pre-breakdown in Composite Dielectrics," In *Shikoku-section Joint Convention of the Institutes of Electrical and related Engineers*, Tokushima, Japan, 2011.

- T. Akagi, Amir Izzani Mohamed and K. Kadowaki, "Dependence of space charge behavior of low density Polyethylene on applied DC electric field at the long term," In *Shikoku-section Joint Convention of the Institutes of Electrical and related Engineers*, Tokushima, Japan, 2011.

- T. Akagi, Amir Izzani Mohamed and K. Kadowaki, "Dependence of breakdown time of low density Polyethylene on applied DC electric field at the long term," In *National Conference of the Institutes of Electrical and Engineers Japan*, Hiroshima, Japan, 2012.

- T. Akagi, Amir Izzani Mohamed and K. Kadowaki, "Dependence of Breakdown Time Lag of Low Density Polyethylene on Applied DC Electric Field," In *Shikoku-section Joint Convention of the Institutes of Electrical and related Engineers*, Kagawa, Japan, 2012.

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Chapter 1

Introduction

1.1 Overview

There are 2 phenomena occurring in these days related to solid dielectric insulation material that motivated studies in this thesis :

1. the increase in demand on renewable energy power generation and
2. the replacement of old cables

Firstly, the increase in renewable energy (RE) based power generator as a replacement of the conventional fossil fuel generator, is one of the efforts to fulfill the Kyoto protocol [6]. In European countries and Japan, the number of RE-based power generation facilities that are built far away from the load centre, is growing (e.g. the coastal area (wind farm and wave power generator)) [7]. High voltage direct current (HVDC) transmission is more favorable in order to transfer the generated power to the load area. This is due to the low transmission loss of HVDC transmission, which is 30 % - 50 % than comparable HVAC transmission line. Furthermore, transmission loss can be reduced if operated at higher voltage. In HVDC transmission, polyethylene (PE)-based power cables, such as cross-linked polyethylene (XLPE) based cables offer outstanding electrical and mechanical properties are utilized in order to run HVDC power transmission efficiently. Therefore, nowadays, XLPE has become a very crucial part of a power cable for HVDC transmission. Basically, the power cable for HVDC transmission is a huge-sized coaxial cable that is used in residence as antenna cable. To know the limit and later to increase the limit especially of breakdown strength are important for long term run and under harsh condition utilization.

Secondly, most of old underground cables (e.g. those installed during 1950s - 1960s) reach end of its lifetime mainly due to insulation degradation [8]. These important apparatuses need to be exchanged with a far better system in responding to the higher demand on electricity and also for the continuity of electricity supply to customers. The increase in demand for energy requires power companies to deliver higher energy capacity per cubic meter of the power cables. The cable should ideally possess long-term endurance, high temperature and mechanical resistant, no impurities and voids content and strong water-tree resistant.

Factors such as localization of contaminants and micro-voids during production have significant role in initiating breakdown. A lot of experiments and theories related to breakdown of dielectric insulating material have been proposed. One of those, is a theory on the influence of space charge on the breakdown of PE material. However, how space charge affects breakdown of dielectric insulating material is not clearly answered. Therefore, continuous efforts are needed in understanding space charge phenomena in dielectric insulation material and how to suppress its influence on breakdown of PE. In this thesis, space charge behavior that leads to breakdown process is discussed. Space charge behavior in insulation material was observed by Pulsed Electroacoustic method. Optical observation of the material during dc breakdown test was also carried out. In optical observation of breakdown, the material was composed of liquid and solid insulating material for easy observation.

1.2 Polyethylene

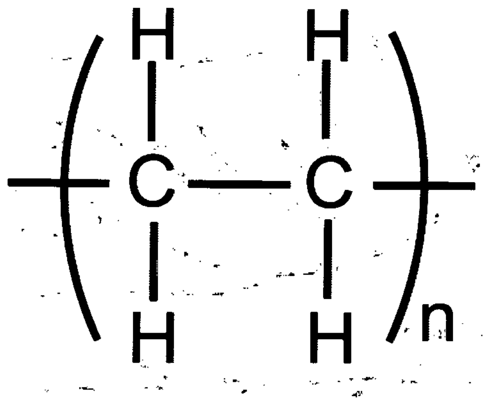

Figure 1.1: Polyethylene chemical structure.

Polyethylene (PE) chemical structure is shown in figure 1. It is a polymer of ethylene monomer through radical polymerization process. In a highly purified method, ethylene is directly transfer from petroleum refinery to polymerization plant. Under appropriate condition (temperature, pressure catalyst), the double-bond of ethylene monomer break to give way to form long chain. It was first accidentally discovered by Hans von Pechmann, a German scientist, while heating diazomethane, in 1899. His colleagues Eugen Bamberger and Friedrich Tschirner characterized the white solid as containing methylene units and called it polymethylene. The discoveries did not make it very useful for some 30 years. PE from ethylene monomer was probably initially synthesized by M.E.P. Friedrich when it was an unwanted by-product from reaction of ethylene and lithium alkyl compound. That occasion took place while he was a student of Carl S. Marvel in 1930 [9].

The PE we know nowadays is attributed to the work of British ICI's Eric Fawcett and Reginald Gibson in 1930's[1] when they were experimenting on what kind of product can be made with ethylene under extremely high pressure. On march 1933, formation of a white solid was found when they combined ethylene and benzaldehyde under high pressure, at approximately 1400 atmospheres. Next time, they attempted the same process but with ethylene alone but it did not produce

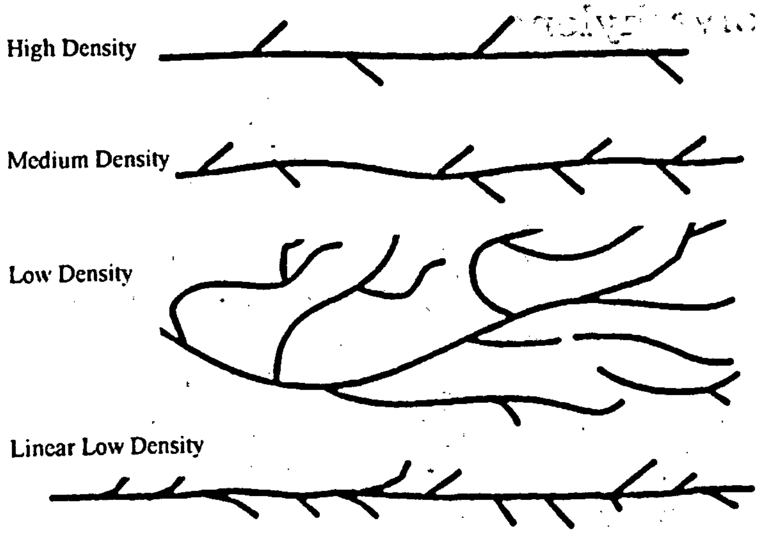

Figure 1.2: Polyethylene chain type [1].

PE. It was barely a decomposition process of ethylene. Their work delayed again until december 1935 when they got new and better high pressure equipment. While experimenting with the new vessel at 180 °C, pressure inside the vessel consistently decreased with the formation of solid. They increased the pressure in the vessel again with pumping more ethylene into the vessel. They found out that the pressure drop was not totally due to formation of the solid but there were small leakage found. The leakage allow small amount of oxygen in which accidentally a right amount needed to catalyze reaction of the additional ethylene. As the ICI scientists found the new material, J. N Dean of British Telegraph Construction heard about the new material. He believed that PE would be a suitable material to insulate and enclose their new undersea cable [9]. In july 1939, sufficient amount of PE was made to coat 1 nautical miles of cable. Polyethylene is classified by its density. The density reflects the level of crystallinity of each type. Usually, PE is divided as low density polyethylene (LDPE), medium density polyethylene (MDPE), high density polyethylene (HDPE) and linear low density polyethylene (LLDPE). Figure 1.1 shows chain type of different PE [1].

- **Low density polyethylene (LDPE)** - Has between 40 - 150 short alkyl branches for every 1,000 ethylene units. It can be produced under high pressure between 15,000 psi to 50,000 psi and at 350°C. LDPE has density between 0.912 g/mL - 0.935 g/mL. The branching in LDPE matrix making it an amorphous with 50 % of crystallinity and allowing gas to permeate through. Melting point of LDPE is 100 °C [9].

- **Medium density polyethylene (MDPE)** - It can be produced by ion-coordination polymerization. MDPE has density between 0.93 g/mL - 0.94 g/mL. It has good shock and drop resistance [9].

- **High density polyethylene (HDPE)**- HDPE chain is packed due to its low level of branching. This is attributed to the regularity in structure of its chain. It is a 90% crystalline with density of approximately 0.96 g/ml. HDPE can be produced under low pressure with the presence of Ziegler-Natta or Phillips catalyst. It possess less than 15 short-alkyl branches for 1,000 ethylene units. Physically, it is stiffer than LDPE with higher melting point which is at 130 °C [9].

- **Linear low density polyethylene (LLDPE)** - This material can be produced under 300 psi and at 1000 °C. Its density lies between 0.915 to 0.925 g/mL. It is a copolymer of ethylene with approximately 8 % - 10 % of an alpha olefin (chemical formula C_nH_{2n} having a double bond at the the most front position) e.g. 1-butene, 1-penetene, 1-hexene and 1-octene. It does not contain long branch as such in LDPE. It has higher tensile, impact and puncture strength than the LDPE. Usual application can be seen as packaging for cables, toys, pipes and containers [9].

- **Crosslinked polyethylene (XLPE)** - Cross-linking method is introduced to PE in order to improve its chemical, mechanical and electrical characteristics. Chain of PE is 'binded' to each other making the XLPE cables resilient to elevating temperature during voltage application. As a results, a thermoset material is borne. Other than as electrical insulating material, XLPE also used as tubing material [10].

In the early 1970s, PE cables failure were reported. It occurred earlier than the estimated lifetime and at rapidly increasing rate [1]. However XLPE cables failure rate not so much as that of PE cables [10]. Since then, extensive approaches were undertaken either in cables production process or the polymeric material itself to solve the problem.

1.3 Space Charge in Solid Dielectric Material

Mason clarified polarity effect on breakdown of solid dielectric material [11]. 25 % greater stress was required to trigger breakdown under negative polarity voltage than under positive polarity voltage, regardless of the solid dielectric thickness. Artbauer and Griac, Coopers et al. and Fava found that impulse breakdown strength is approximately 80 % of that under dc [12, 13, 14]. Bradwell et al. discovered that impulse strength of polyethylene (PE) significantly increased, by applying same polarity of dc voltage prior to impulse voltage application [15]. In contrast, application of opposite polarity voltage, reduced impulse strength. This phenomenon is known as pre-stress effect. During pre-stressing, space charges accumulate in bulk of solid dielectric. Pre-stress with same polarity voltage, preventing instant increase of electric field in the insulation thus reducing the impulse breakdown voltage. On the other hand, opposite polarity of residual charge by opposite polarity pre-stress resulting in the increase of electric field near the electrode almost instantly, which causing breakdown to occur at lower voltage. Watson studies shows that breakdown strength increase with lower voltage rise time [16].

Kitani studied several types of polymers under nanosecond (ns) impulses voltage [17]. His studies was further extended to a study in the influence of pre-stress to impulse breakdown strength [17]. The yielded result was in agreement with that by Bradwell et al. [15]. In his other study on PMMA subjected to ns impulse voltage, he found that tree channel (electrical tree) was longer and there discharge light was more intense under positive rather than the negative one. In addition, the tree grew faster under application of alternating positive and negative impulse than under repetitive positive impulses application. Under repetitive negative impulses, the tree channel did not grow instead, it got broadened [18, 19].

Ieda et al. studied the influence of oxidation on space charge in PE [20]. Oxidation increased the conduction current by a few orders of magnitude and negative homo space charges were formed near the cathode. The introduction of an oxidized PE layer on the surface of unoxidized PE enhances both electron and hole injection. The enhanced electron injection predominates over the hole injection and plays an important role in the increase of conduction with oxidation. The negative homo space charge is released a $40\,^0$C and give rise to a Thermally Stimulated Current (TSC) peak. They also quantitatively observed space charge by LIPP method [21]. The amount of negative space charge increased with applied field, and this suggests that the electron injection was enhanced by the applied field.

These previous studies indicate that understanding role of space charge in breakdown of solid dielectric is important particularly in designing high voltage apparatus.

1.4 Space Charge observation method

There are 2 types of method that can be utilized in order to observe the space charge in a dielectric material; 1) destructive and 2) non-destructive methods.

1. Destructive method

   - **Lichtenbeg Figure/Dust Figure method-** Lichtenberg figure is named after the person who found it, a german physicist Georg Christoph Lichtenberg. In his experiment in 1700s, he applied high voltage to various dielectric material such as resin, glass and ebonite (hard rubber). After that, he sprinkled a mixture of sulphur and minium (red lead/ lead tetroxide) on the surface. He found that sulphur is attracted to negatively charged region while minium is attracted to the positively charges region [22, 23, 24].
   - **Capacitive Probe method-** This method allows rough estimation of charge pattern and residual charge distribution. However, the resolution is very poor, which is in the order of milimeter.
   - **Thermal Simulated Current method-** Allows one to obtain activation energies of the different kind of charges and their relaxation time. This method does not allow continuos measurement of space charge. It's very sensitive to polarization particularly in dipolar polarization but less sensitive to space charge [25, 26].

2. Non-destructive methods

   - **Thermal Step (TS) method-** It is proposed by Toureille during external current measurement in PE film. 2 piece of film is electrified by corona discharge on 1 side. Then both electrified-die film were brought together so that the surfaces face each other. The sample was placed in a thermostatic container under -10 °C for 90 minutes and then the temperature was raised to to 20 °C. This process creates the thermal step and the corresponding current is recorded. Temporal behavior of space charge during voltage application in not observable by this method [26].

- **Laser Induced Pressure Pulse (LIPP) method-** Spatial resolution of this method is said of 1.5 m in Teflon FEP [27]. Laser pulse (width from 70 s to 1200 s) is directed to a graphite electrode on the target sample, pressure wave is generated.The pressure wave travel through the sample. Current generated during this time is proportional to charge density. In spite of having very high resolution, this method is prone to noise. Plasma generated during near the electrode can the noise source thus disturbing the desirable space charge distribution.

- **Pulsed Electroacoustic (PEA) method-** Figure 1.3 shows principal diagram of PEA method. This method is the most popular because of its comparatively easy setup and low noise. Pulse generator is coupled to high voltage source. During voltage application, space charge is polarized. The application of a very short pulse voltage (5 ns) the the

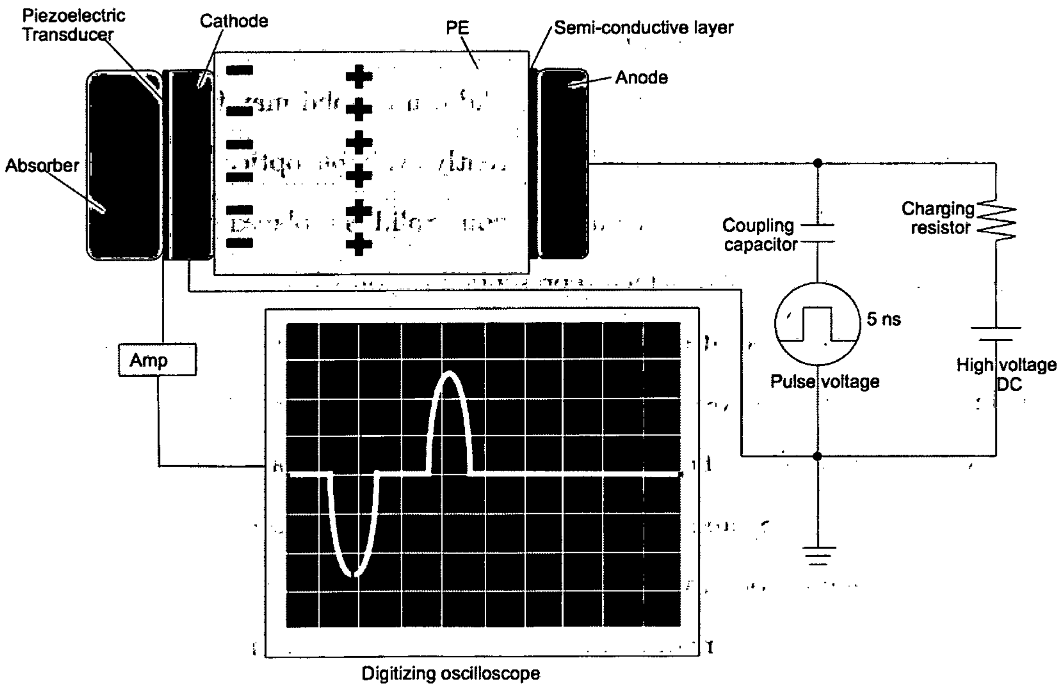

Figure 1.3: PEA method.

polarized sample creates a sudden distortion in the local electric field in bulk of the sample thus generating pressure wave (acoustic wave). The pressure wave is detected by a piezoelectric transducer (in some cases polyvinylidene difluoride (PVDF) is utilized) and then amplified so that it can be viewed on oscilloscope [28].

1.5 Contribution

In this thesis, space charge behavior and pre-breakdown phenomena in solid insulation is discussed. Space charge behavior measurement is carried out by PEA method. In some cases, external current measurement is carried out simultaneously with space charge measurement. External current here is partially corresponding to space charge behavior. It is assumed to consist conduction current and displacement current.

On the other hand, it is extremely difficult to observe a breakdown phenomenon process, in solid under dc stress, for example, from initiating process (pre-breakdown process, such as partial discharge etc.) and its development (how the breakdown light propagate) until the breakdown complete. It can occur almost instantly and at an uncertain time. Dc breakdown in solid may finish in only 1 ns after initiation. The operating speed of currently available optical observation system can not match this ultra-fast phenomenon. Solid is replaced with a composite insulation of liquid and solid, which represents 2 region with different conductivity in 1 sample (will be elaborated further in chapter 2), in making optical observation of dc breakdown process possible. By doing this, pre-breakdown light image can be observed by the optical method. In this thesis, optical observation method refers to image guide scope (IGS) and streak camera system. The processing delay time in streak camera requires one to have an image delay path (sometimes noted as light delay path) so that the camera can record a desired image. In this thesis, image delay path refers to the IGS. It is a fiber optic cable for translating image from