Guest Forum

Lecture by a Jury Member for the 2006 Masao Horiba Awards

What Do We Need Other than Luck?
— XGT and Its Subsequent Research —

Hiromoto Nakazawa
National Institute for Materials Science
Fellow
Doctor of Science

XGT (X-ray Guide Tube) has history as already 25 years passed since its first presentation. Although the idea to install a long XGT in the second generation photon factory (PF) was rejected, the technology of XGT won a degree of success in the form of HORIBA's X-ray analytical microscope XGT series partly thanks to a number of luck. Luck is indispensable to win Nobel Prize or victory in high school baseball at Koshien, but what else do we need to get a success? In this report, I am writing about the circumstances, basic ideas, the process of development and how I shifted my focus to research on the theory on the origin of life after XGT so as to provide you some food for thought.

Introduction

I just heard from the chair of the lecture for the first time that 1,000 units of XGT series (HORIBA's product name for X-ray analytical microscope) have been already sold. I thought machines in this series are very lucky and I, the inventor, am lucky, too. But in the initial phases of research, people said XGT would not be useful and that made me feel so sad. Now my invention has been used by many people and I feel very proud of it as the inventor. I greatly thank the people in HORIBA. From now, I am telling you about the history of XGT. I hope each of you think about what we need other than luck while reading my report.

The Basic Idea for XGT

Three patents are involved in the X-ray analytical microscope: microscopic X-ray analytical equipment (Japanese Patent No. 1699838) in 1982, X-ray divergent angle control unit (XGT) (Japanese Patent No. 1806535) in 1984, and scanning X-ray microscope (Japanese Patent No. 1828290) in 1985. The research outcome on the effect of XGT was orally announced in 1981, and before those patents were applied, many things happened that gave birth to the basic idea of XGT. One of them was the atmosphere in the days of the 1970s when X-ray was very frequently used. In those days, one of the active fields of research was the dynamical theory of X-ray diffraction as crystal physics. There were also many people working on X-ray topography, and research on semiconductors, ceramics and other materials was conducted at various universities in this country. Crystal structural analysis was also actively studied, and the focal point was shifted from inorganic crystals to organic crystals such as protein. The work on ordinary, inorganic crystal had already been completed then. For crystal structural analysis, activities of sharing or exchanging of analytical programs were carried out mainly among the Crystallographic Society of Japan. Under these circumstances, the Congress of the International Union of Crystallography was held in Kyoto in 1972. Then, there was a growing demand for "much stronger X-ray and more light." That leaded to synchrotron radiation.

Background to Synchrotron Radiation

The first thing that shed light on the trail to "more light" was the Physical Society of Japan in 1971. A symposium

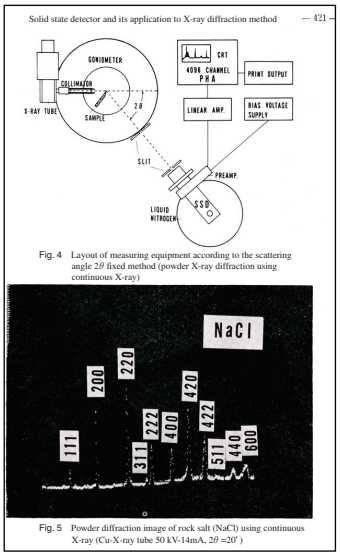
was held on how to produce super powerful X-rays and how we should use them better. There were two main subjects to discuss. One was upsizing the rotating target X-ray generator, which enjoyed great development later. Meanwhile, there were people who preferred synchrotron radiation and they proposed the calculated predictions on synchrotron radiation. Prof. Kikuta and Prof. Kohra disclosed their calculations that even 2 GeV and 500 mA synchrotron radiation bent by 1 T bending magnet can emit stronger X-ray by two digits than that by 50 kV and 1 A rotating target. This brought about the impetus that later realized set-up for synchrotron radiation. On the other hand, in terms of national policies on science and technology, the idea to construct an elemental particle research facility was already suggested by the Science Council of Japan in 1962. The proposed facility was a combination of a large accelerator department and a cosmic ray observation department and the suggested budget was 30 billion yen. Considering the total spending on scientific research in those days was 4 billion yen, the figure was astronomical. The proposal was gradually shrunk in size, and only the accelerator department was established as the National Laboratory for High Energy Physics in 1971. Under these circumstances, demands for stronger X-ray were integrated into the idea of Photon Factory (PF) in 1973. It was adopted by the Science Council, that issued a recommendation in 1974. But the Council's recommendation was modified, which was to attach Photon Factory as an auxiliary facility, not an independent one, to the National Laboratory for High Energy Physics nearing its completion then. Toward the end of the 1970s, construction of a 2.5 GeV synchrotron radiation facility was initiated. The decade of the 1970s was an era when laboratory X-ray was being shifted to synchrotron radiation.

My Research at That Time

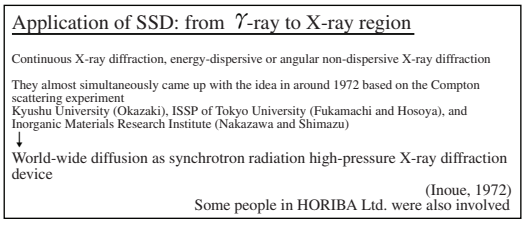
At that time, I was engaged in research on long-period superstructure (modulated structure). This structure is a large lattice structure with waves lying over the rows of unit cells, and I found a non-integer type superstructure in it. The relationship between the satellite reflection position and the main reflection position is a non-integral multiple. It means the superstructure was against the concept of crystal, or "infinite repetition of unit cells." Since reflections resulted from the superstructure were weak, I used X-rays produced by the rotating target. This was the reason I got into the group of "more light." This superstructure of a non-integral multiple was clarified by the HRTEM (high-resolution transmission electron microscopy) image (which was then called lattice image), and the answer later obtained was that it was a multidimensional structure. On the other hand, my official work at the National Institute for Research in Inorganic Materials (NIRIM) was development of a new material called Fe_3S_4. We synthesized sulfides by vacuum deposition (close to today's ion beam epitaxy) and found some new compounds.

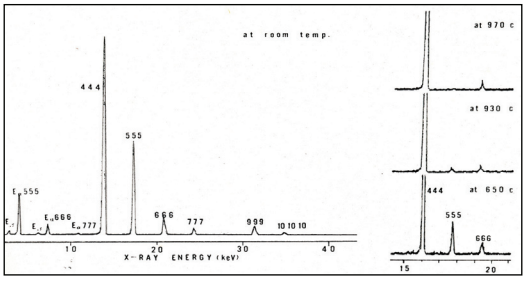
Eve of Synchrotron Radiation

While I was involved in those things, the eve of synchrotron radiation came. I conducted research on energy dispersive X-ray diffraction or angular non-dispersive X-ray diffraction, that is, X-ray diffraction, or continuous X-ray diffraction, using a solid state detector (SSD) (Figure 1). This research was not carried out by only me but also at the Institute for Solid State Physics



Commentary thesis (Shimazu and Nakazawa, 1974)





Observation of regular and irregular transition of V_5S_8 with SSD (Nakazawa et al. 1975)

Figure 1 Eve of synchrotron radiation: application of solid state detector to X-ray diffraction method

(ISSP) of Tokyo University or at Kyushu University, and Prof. Hosoya or Dr. Fukamachi of ISSP were a step ahead of me. I used this method for research on order and disorder transition. Since synchrotron radiation is continuous X-rays, that research provided the spring board for the dawn of Japan's synchrotron radiation application. Dr. Inoue of IPPS, who was working on high pressure, published a technique that run X-rays in the high-pressure equipment and saw them with SSD in 1972. In this way, in early 1972, the stage was set ready for continuous X-ray diffraction.

From Synchrotron Radiation to XGT

Now the Photon Factory was finally set up. Most of the Japanese people related to X-rays were offering voluntary cooperation. I also went to the construction site of the Photon Factory and did some work including positioning of the machines using the transit. I was in charge of the 4-circle diffractometer for synchrotron radiation. I was a member of the group to make it and start it up.

There was an atmosphere that we should create a new thing together and the idea of X-ray guide tube sprang up. First synchrotron radiation was not so strong and was instable; it faded away only in about 30 minutes after its emission started. For the synchrotron radiation facility, since the distance from the bending magnet to the laboratory station was 10 m minimum and 20 m maximum, even highly directional synchrotron radiation would spread. We tried to concentrate the radiation into a small region using the convergent optical system. One of the ways to concentrate the radiation was to diffract the radiation with the bent single-crystal. And the second was to diffract it with artificial laminated films. Moreover, the third was a total reflection mirror using total reflection. The methods that used the diffraction phenomena had some shortcomings, which were the limitation of X-ray wavelength and poor efficiency. The total reflection mirror was most widely used, but it also had some shortcomings, which were that the size was big and that since it was a mirror, it could not collect radiation of entire solid angle but only collects radiation from the reflecting plane. To collect radiation from all solid angle, the system had to have multiple mirrors. But the Western countries, which were ahead of Japan in this respect, already had very advanced mirror systems, and Japan just imitated theirs.

Then the idea I thought up was X-ray guide tubes using molten glass. It is very hard to polish the glass surface so perfectly that it totally reflects X-ray. But the molten glass is considerably flat because of the air-liquid interface. The surface has some undulation, but waves are gentle. It causes some loss but is still more advantageous than polishing the ordinary glass surface. In the first place, I believed the idea would become Japan's original optical system.

It was in 1980 that I proposed this idea. It was just when the synchrotron facility was established and people were discussing how to use it better. I proposed installation of X-ray guide tubes, but unfortunately the idea failed to have the approval. The principal I thought up was below. Radiation emitted from the bending magnet of the synchrotron diverged at about 10^{-3} rad (3 mrad) in the horizontal direction at that time. The level was one digit smaller in the vertical direction. It was 10^{-4} rad in the vertical direction as it did not almost spread out. Then, when it was emitted to a distance of 20 m, it spread out to 6 cm in horizontal at the position of the sample (Figure 2). Now, if we placed a crystal of 30 μm or 40 μm in size there, the crystal ended up using almost no X-rays.

If the radiation was brought to the position of the crystal by using the total reflection of cylindrical XGT, the intensity became very strong as it increases to the square of the ratio of distance. The divergent angle of the synchrotron radiation, the total reflection critical angle of glass, and the inverse calculation of the reciprocal lattice point of crystal, all results turned out to be about 3 mrad. I proposed production of X-ray guide tubes as we would bring the reciprocal lattice with spread and therefore, we did not need any more parallelily.

There was no predecessor and nobody tried it before. It was all in my mind. So I hurriedly conducted an experiment as in Figure 3 to demonstrate its validity. As inclining 2 glass tubes with 200 mm long (both tubes) and with 0.5 mm (the one tube) and 0.2 mm (the other tube) in diameter, I thought we would get such patterns as shown in Figure 3 (a) if total reflection occurred. From this experiment, we got the patterns as in Figure 3 (b) as I just predicted. It was proven that, with ordinary glass tubes, total reflection could be caused easily.

Another experiment was conducted to show the gain of X-ray intensity as in Figure 4. We used a long glass tube and a lead pinhole having the same diameter as the glass tube. And we put the pinhole at the front of the tube. Then, X-ray intensity was measured when the glass tube was removed, and X-ray intensity was also measured when the glass tube was there to get ratio between the

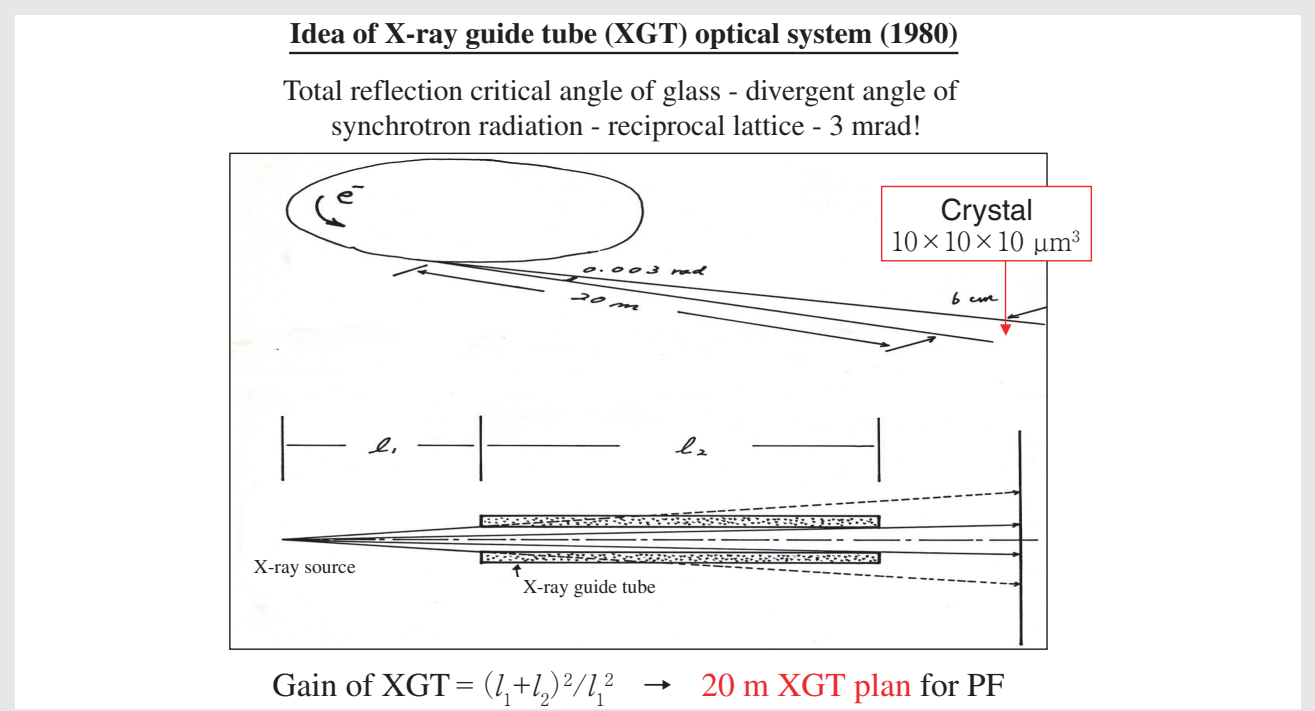


Figure 2 Idea of X-ray guide tube (XGT) optical system (1980)

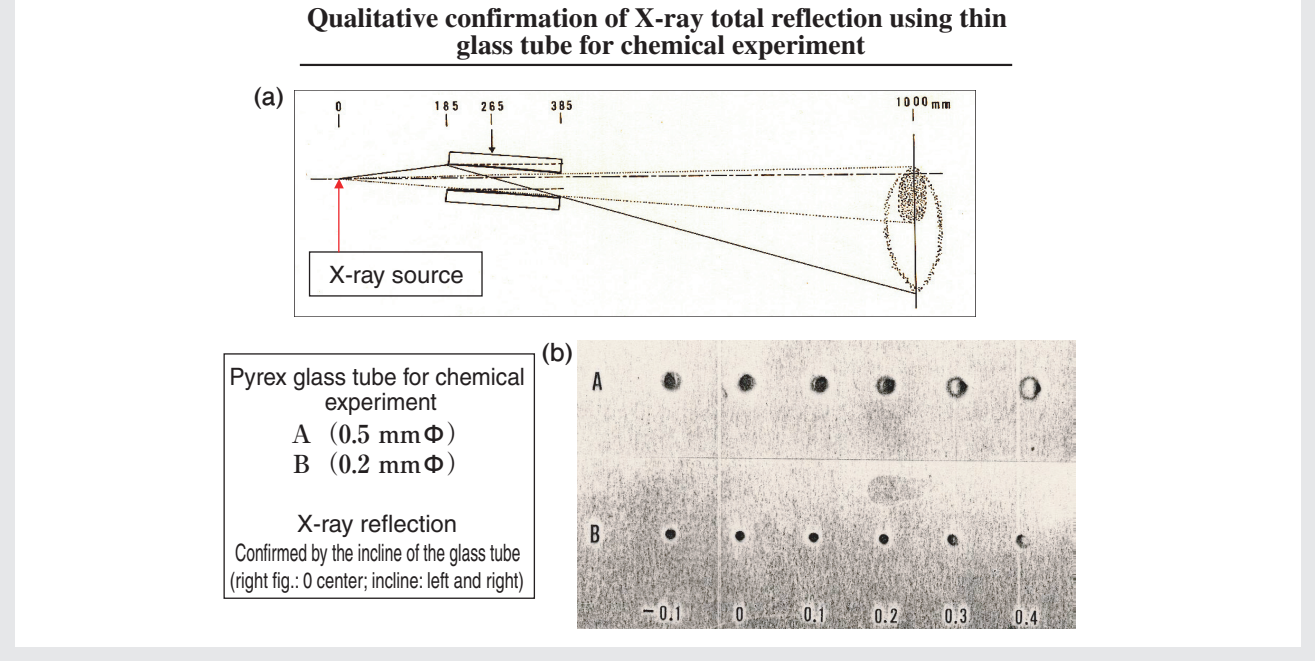


Figure 3 Qualitative confirmation of X-ray total reflection using thin glass tube for chemical experiment
(a) Schematic diagram of the experiment
(b) Direct and total reflection X-ray image
Values in Figure (b) show the movement degree (mm) of the glass tube as the incident end of the tube is fixed and the other end is pushed and represents the degree of incline.

results. Figure 4 (b) shows the result of measurement using a X-ray tube of the copper target and a SSD. The intensity ratio about 35 times was obtained for Cu Kα radiation.

My proposal had these advantages, but, since you had to put a long glass tube into super vacuum, the people who were constructing the PF those days hated it. Finally, the idea of installing a 20 m long X-ray guide tube was rejected.

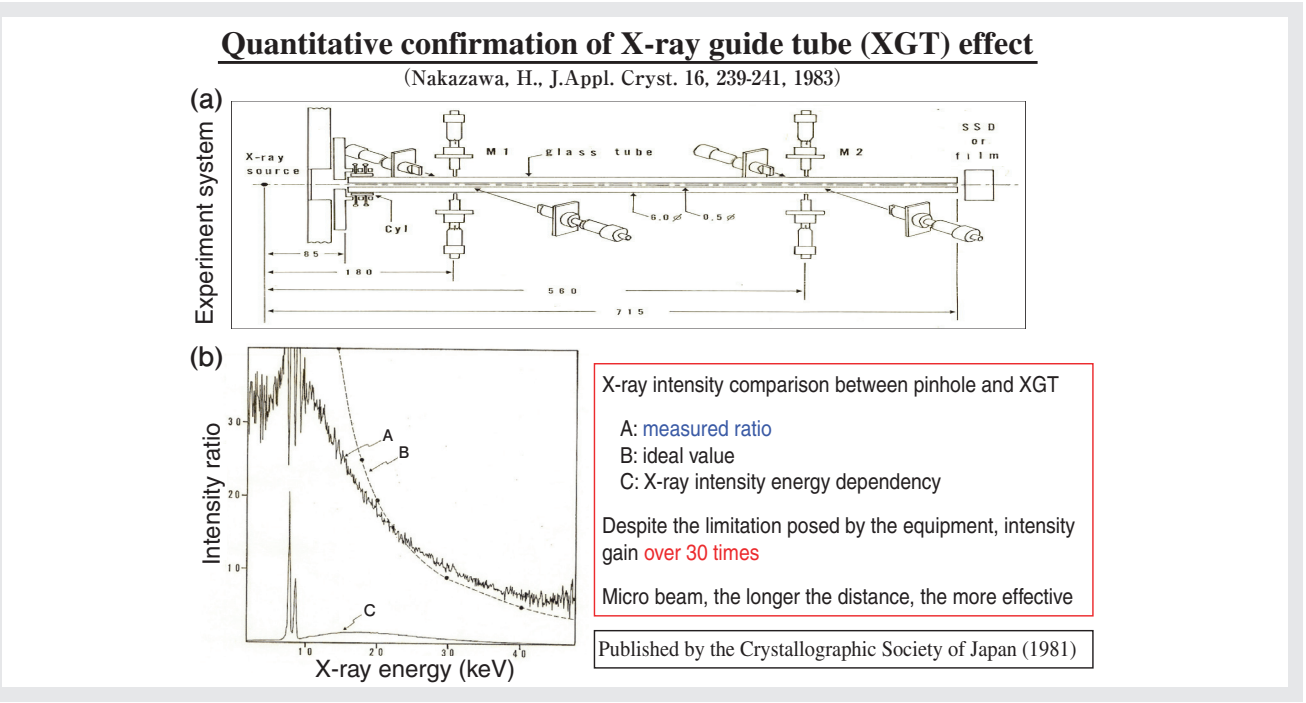


Figure 4 Quantitative confirmation of X-ray guide tube (XGT) effect
(a) Experiment system
(b) X-ray intensity comparison between pinhole and XGT

From 20 m XGT to 0.2 m XGT

The poster presentation on the advantages of the X-ray guide tube in the 1981 issued by The Crystallographic Society of Japan (Figure 5) was so popular that I thought I could not just throw it away. So I put it on the wall of the dark corridor in the laboratory.

In 1985, Mr. Shigeyuki Akiyama, who was in charge of planning a semiconductor project in HORIBA and happened to visit one of my colleagues for something related to his job, saw the poster and told about it to his colleague, Mr. Yoshinori Hosokawa. Later, I came to work with Mr. Hosokawa and this opened up our relationship. Probably the human relationship in HORIBA was so good that an employee knew very well what was troubling another employee and wanted to help him by giving information.

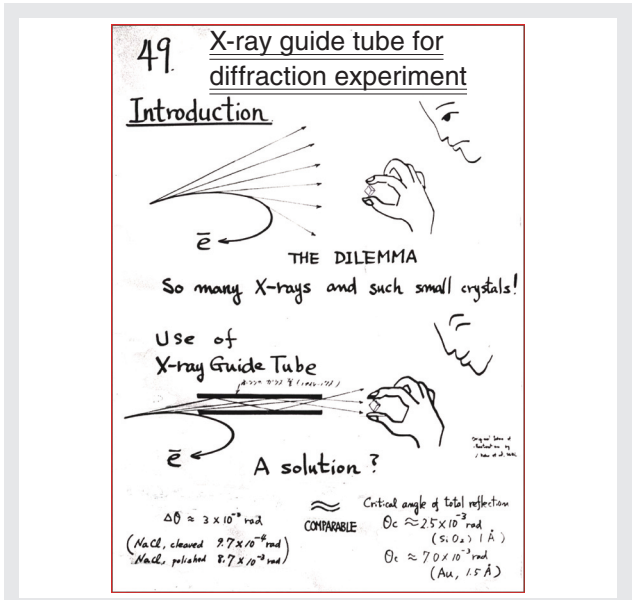


Figure 5 First half of the poster presentation of the crystallographic society of Japan in 1981 (one piece of simili paper, reproduced from the photo)

In a sense, rejection of my idea for synchrotron radiation was another example of my luck. For me, I believed the X-ray guide tube was a good idea and felt sad when it was not accepted for synchrotron radiation. But as I gave it another look, it was not indispensable to extend the tube to 20 m. If we made the length decreased to 200 mm and the diameter to 200 µm, there would have been more things we could do. At the same time, by changing the shape from a long cylinder to a long spheroid, we then could focus X-rays from a point source to another point, and with a shape of paraboloid, we could focus parallel X-rays. To be sure, there is no dimensionless point light source and no complete parallel X-ray at hand, so we had many difficulties on how to do in practice. But technically speaking, it seemed possible to reduce the light source size and enhance parallelism. But I did not consider about it so strictly and just focused on light converging.

What I did actually was that; we could not produce a

spheroid, so we regarded an inverse cone as half of the spheroid and conducted an experiment to see how small the place X-rays could be collected to (Figure 6). As shown in the photo of the beam section of this figure, the inverse conical XGT was proven to converge X-ray better than the cylindrical XGT. I had known that if this experiment went successful, we would be able to measure minute parts. So the successful results made me believe the basic concept of X-ray microscope was almost fully established.

I applied for a patent as an X-ray microscope in 1985 and at the same time offered the proposal to some domestic X-ray specialized manufacturers, but no manufacturers evaluated my idea except HORIBA, the company of Mr. Hosokawa who I was in contact with through Mr. Akiyama. I decided to work with HORIBA and started joint work since then. We started production of a model version of scanning X-ray analytical microscope (Figure 7) in 1987 and disclosed the fluorescent X-ray image and diffraction image of zincblende including iron (Figure 8) at the Tochigi gathering of the 1st International Conference on X-ray Microscopy in 1988. The images were the first obtained by the scanning X-ray analytical microscope using the X-ray guide tube. The person who was in charge of data collection software is Shintaro Komatani, today's Scientific Systems Products R&D

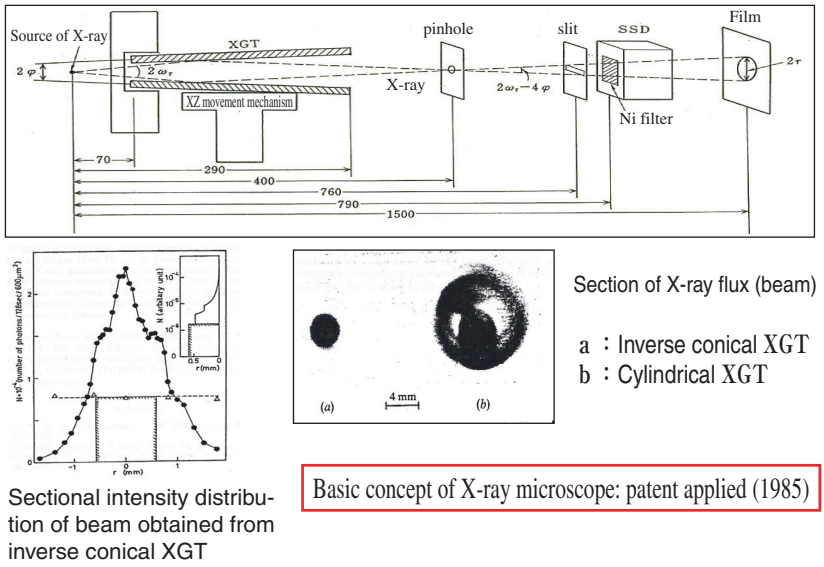


Figure6 Experiment using inverse conical XGT

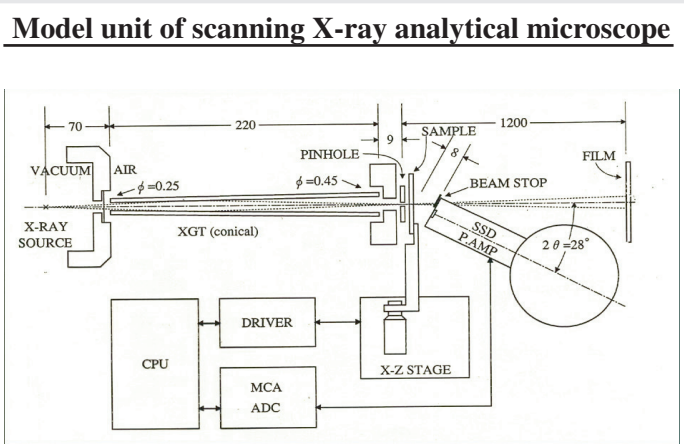


Figure 7 Production of the model unit of scanning X-ray analytical microscope

Guest Forum What Do We Need Other than Luck?

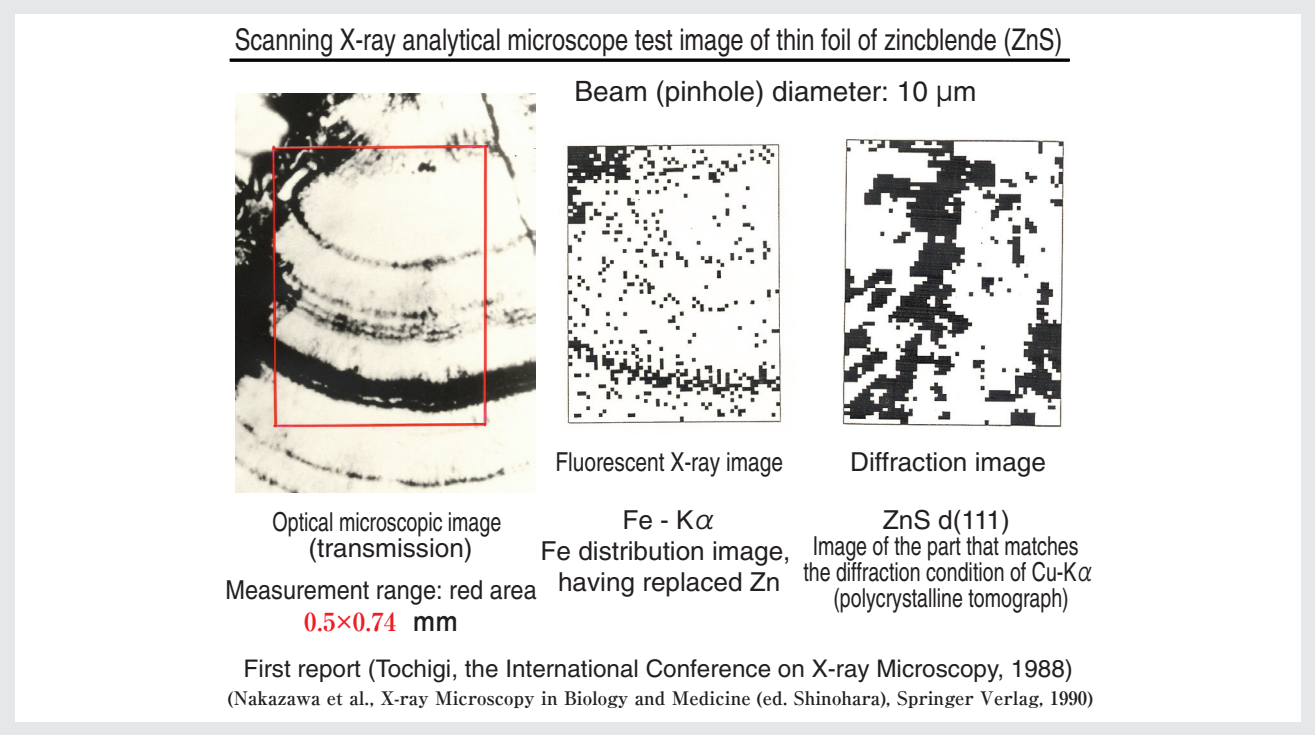


Figure 8 Scanning X-ray analytical microscope test image of thin foil of zinc blende (ZnS)

Department Manager.

Since the X-ray analytical microscope (HORIBA's product name; XGT-2000) is operable in a non-destructive manner, without causing contamination and in non-vacuum, it has many advantages including the capability to measure electronic products and biological bodies and to observe the inside with its permeation property. Compared with SEM or TEM, the product has apparent advantages, but HORIBA had to overcome considerable barrier until their commercial product was accepted in the market. The barrier would be its novelty: it was quite a "novel microscope" with no precursor machine present. Those barriers were torn apart by the combined efforts not only of the engineers, but also of people in sales department and supporting department trying to let the product known to the users. It took a lot of time and labor. It can only be done by an organization of a company. Later, accepting commission development work with the Research Development Corporation of Japan, HORIBA added some new features to the microscope. The major features are downsizing to the desktop type, the capability of measurement of transmission X-rays in simultaneous measurement of fluorescent X-rays and transmission X-rays. Other than these, features including vertical placement of XGT for operation and the capability of switching two types of XGT, each having a different beam size, also enabled HORIBA to tear apart the barriers, I think. In 1993, XGT-2000 made a debut and more improvement was made to create new-generation XGT-5000.

Use of X-ray Analytical Microscope

While X-ray analytical microscope became widely used as HORIBA XGT Series among the front-runners in the field of science and industry, I committed myself to improve the measurement resolution to the maximum. Ultimately, with the current level of our laboratory capacity, we achieved a maximum level of 1 μm in resolution. I resigned the NIRIM and moved to Tohoku University, where I found my colleague Prof. Yoshida used XGT-2000 for his research on rock thin section (Figure 9). That is the kind of moment I strongly feel proud and happy to be the inventor.

When I was assigned as the on-duty docent of the museum belonging to the university, as one of the roles of a professor in the university which was open to the citizens, I used Figure 10. The left photo is the image of a Jurassic fish fossil unearthed in China measured with XGT-5000. Green shows calcium, red shows sulfur and blue shows iron. Sulfur is from barium sulfate, and special bacteria that condense barium ate the cartilage and left barium sulfate. The iron inside is the product of

different bacteria. Since bone is calcium, bone, flesh and fins are seen separately. Other than bone, various parts of the fish are seen different depending on the bacteria that ate them. The right drawing is a trilobite. When analyzed by XGT-5000, calcium is seen in red, while iron in green. Iron is Fe of FeS_2, which is the result of bacteria eating the inside and replacing it with FeS_2. It was shown that while somebody was touching the fossil, shells of thin calcium carbonate layers were peeled, and the green part is shown to be the inside. These observations are exactly the process of fossil anatomy.

What Do You Need Other than Luck? — In the Case of XGT

What do you need other than luck? You must have thought about it while reading my story so far. You may try to think the way only unique to you; you may doubt the "common knowledge," or you may respect whatever comes up to your mind. Each interpretation from the position you are in is probably right. According to the Science and Technology White Paper in 2003, the total

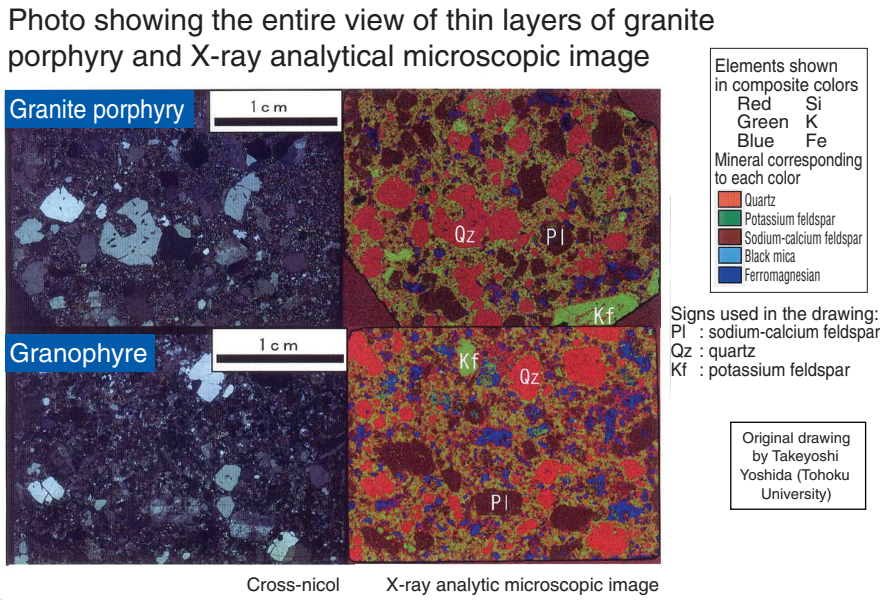


Figure 9 Photo showing the entire view of thin layers of granite porphyry and X-ray analytical microscopic image

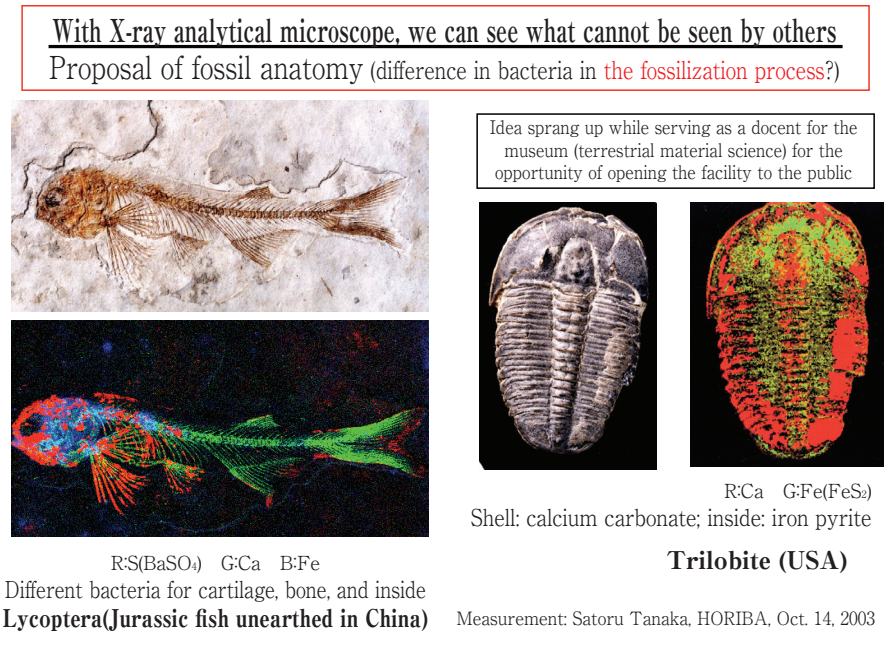


Figure 10 X-ray analytical microscopic image of fossil

number of Japanese researchers is about 730,000, and the proportion of researchers among corporations, universities and research institutes is 40:15:4, indicating corporate researchers are dominant. I hope you, researchers who support Japan, consider what you need other than luck and work for it.

Career After XGT

When my research on XGT was almost over, synchrotron radiation became available for everyone. It was put to open use after 1985, and I was old enough to think about changing the subject of my research. As we all are, when we become experienced, we know what is right and wrong, and can make good judgment. That means we are superior as supervisors. But at the same time, we lose the opportunity to make a big discovery. Great discoveries often come from thoughtless judgment or mistakes. Therefore, aiming at great discoveries will not be suitable for sensible and experienced people.

When we are young and newly-fledged, we often come up with interesting ideas with thought, "It might be…", but they often turn out to be wrong or misunderstanding. We believe we got things without understanding properly by listening to your teachers. But still, we have gained great profits just by doing what you think of. Ignorance is bliss, indeed. If this theory works, then what should a veteran researcher do? Changing to a different subject is one way. If you go to a field you are not familiar with, you would find it so fresh and interesting because there are many things you do not know. You may misunderstand or hold illusions. From this idea, I decided to go into a little unknown field.

When I thought back what interested me most in youth, one book came up to my mind, which was "The Physical Basis of Life" by J.D. Bernal (Iwanami Shinsho).

The author was a famous X-ray crystallographer and the person I admired when I was young. The book explains the system of life very logically and physically, and says that clay mineral is greatly involved in the origin of life. The origin of life is the topic much discussed by other great scientists including Oparin, Miller, and Schrödinger other than Bernal but, in the course of history, we presently know quite a lot that those people did not know. So I thought this genre would be worth a try and decided to get involved in research of the origin of life.

By using the research theme proposal scheme of my institute, I set up a research group on smectite, a clay mineral believed to be involved in the origin of life. It was in 1985, and we were trailblazers of the era when the environment or new materials started to draw attention. One of our achievements in those days was production of a single crystal smectite under super high pressure and high temperature conditions, which are the same as those for diamond synthesis. This single crystal is very similar to an organic polymer molecule although it is an inorganic crystal, as shown in the SEM image of Figure 11.

I continued the research and came up with a proposal that

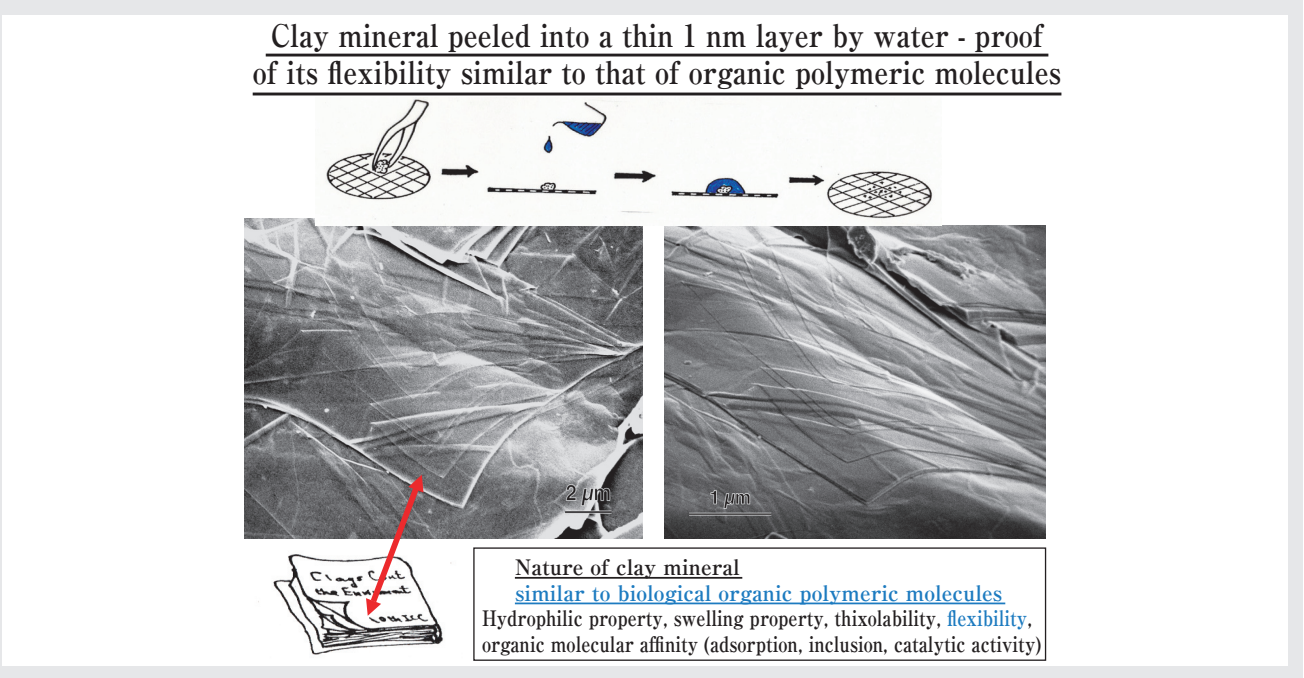


Figure 11 SEM image of water-induced peeling smectite

life was born in the underground, however, this content is beyond the scope of this report. If you are interested, please check my book in Reference[2].

"Common Knowledge" of the Origin of Life

Now back to the main theme, and I would like to ask you three questions.

Q1: Was the ancient sea the mother of life (was life born in the sea)?

Q2: Were organic molecules, or the origin of life, made from ancient air molecules?

Q3: Why do organic molecules or living organisms evolve? (This is not about why each individual life form evolves its own way but is the question asking why evolution occurs in the first place.)

When you are asked to show the evidence to these, "common knowledge," generally taken, many people would not be able to answer with confidence. Surprisingly, a priori "common knowledge" is rarely doubted. Nobody wonders why. This is the dogma of common knowledge. If you doubt it, you think it does not make sense, but, since nobody doubts it, it becomes common knowledge. This kind of thinking is the spring board for ideas for research.

It is conventionally believed that, as the ancient soup (ancient sea) is boiled down, simple organic molecules (amino acids and nucleobases) are polymerized (dehydrated) to become giant molecules (proteins and nucleic acids). But this is contradictory from the viewpoint of thermodynamics. The sea that has plenty of water is the condition of hydrolysis. It also applies to submarine vents, where decomposition easily occurs because of hot temperature. Therefore, it is difficult for life, which is an assembly of giant molecules, to occur in the ancient sea. I omit the detailed explanation here, but considering the history of the Earth, it is logically straightforward to think molecules were polymerized in the underground deposit. I recently announced this view and received good reputation.[1, 2]

According to the famous experiment of Miller, spark discharge was caused in the reducing ancient atmosphere, which was thought to be the ancient atmosphere in those days, (H_2, CH_4, NH_3, H_2O), and amino acids were successfully produced. But in the new theory of the ancient Earth, there was a magma ocean (1200 °C) or the total global melting. There, methane (CH_4) or ammonia (NH_3) was decomposed and could not exist. H_2 was gone out of the atmosphere and the ancient atmosphere became oxidized one (N_2, H_2O, CO, CO_2). Hence no amino acids could be formed. Therefore, we have to give a second thought to the common knowledge that organic molecules were formed in the ancient atmosphere.

Schrödinger said that being alive was seemingly against the second law of thermodynamics (natural phenomena move toward a maximum of entropy [chaos]). He also presented the solution explaining with metabolism of entropy that life takes in food of small entropy and discharges excreta of large entropy. But this logic cannot explain why life evolves. Life is destined to evolve in an a priori manner, and nobody generally doubts it. But evolution from small and simple living creatures such as bacteria to large organized living creatures such as plants and animals seems to be against the second law of thermodynamics. Nobody can explain because thermal transfer is not in their mind. Today, the Earth continues to emit heat over billions of years, and its temperature is going down. One can explain that ordering is promoted as the entropy of the entire Earth is reducing because of thermal emission. If we take that biological evolution is part of global ordering or ordering of light elements (H, C, N, O), the reason that life must evolve can be understood. These are three examples to show that we must always doubt common knowledge.

Conclusion

When we think about "what do we need other than luck," there are probably no common answers and everybody can find a different understanding and focal point depending on their own personality and situation. But the only thing needed is a certain kind of will. I want to do something or I should do something — there are various ways to express it, however, that kind of firm will is definitely necessary. Here again, luck is the common denominator for success. The high-spirited will and luck. Please consider what else you need for yourself and make a good use of it for your research and your work.

Lastly, it is because of the people in HORIBA, Ltd. that I feel so proud as the researcher. I appreciate them from the bottom of my heart.

<Excerpt from the Lecture by a Jury Member for Masao Horiba Awards (on November 7, 2006)>

(Publication members have responsibilities for the translation)

Reference

[1] Nakazawa et al., Impact synthesis of NH_3, *Earth and Planetary Science Letters*, 235, 356-360 (2005).

[2] Nakazawa, H., *Origin of Life - Scenario Written by the Earth*, Tokyo, Shinnihon Publishing, Co., Ltd, (2006).