



ESR Dosimetry and Dating toward 21st Century

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Received August 22, 2002

Abstract: Dating and dosimetry using electron spin resonance (ESR) in 20th century developed at both Yamaguchi University and Osaka University have been reviewed with emphasis on new prospects and strategies in 21st century. Natural radiation have been generating radicals that accumulated in archaeological and geological materials. ESR detects these radicals and the ESR signal intensity is proportional to the radiation dose and therefore the age. The assessment of the total dose of natural radiation and the annual dose rate give their ESR ages. The ESR dating of stalactites and stalagmites at Akiyoshi cave in Yamaguchi prefecture in 1975 was extended to anthropological dating using bones and tooth enamel excavated in Greek Petralona cave. Fossils of shells and corals gave the ages of marine terraces and sea-level changes. Quartz grains gave the ages of geothermal alteration and fault movements. Future ESR dating of ices at outer planets and their satellite are also investigated as basic studies for ices of H₂O, CO₂, SO₂ as well as terrestrial hydrates in laboratory. Atomic bomb radiation dosimetry at Hiroshima and Nagasaki using ESR lead to the dosimetry of personnel, Chernobyl and JCO criticality accidents. Monitoring of radiation dose with sensitive materials with tissue equivalence are being developed. Finally a new scanning ESR imaging apparatus (a near field microwave microscope) developed in our laboratory gave ESR images of radicals from fossils to Si-CVD and diamond films as summarized in my book in 2002.

Key words: ESR, dating, radiation, dosimetry, accident, fossil, carbonate, apatite, silica, tooth, bone, MRI, CVD, scanning, microscope

Applications of ESR to Interdisciplinary Fields in 20 century

ESR is now a unique technique of dating and environmental assessment in geosciences as well as radiation dosimetry. In ESR dosimetry, the history of ESR is less than 50 years, while that of ESR dating, 30 years. The early stage of ESR dating and dosimetry were reviewed in various reviews, and book chapters, Proceedings (Ikeya and Mki, 1985) as well

as in a monograph (Ikeya, 1993). In every field, the progress in 20 century is so enormous that one becomes pessimistic and liable to think that the best innovative age was over. I have heard that a young postdoc lamented that most of ESR works had been done in 1970's. If we look back the history, scientists at the early 20th century also said so, but faced explosive expansion and progress of science and technology thanks to new discoveries brought about by new techniques.

Progress in semiconductor technology and computer science will completely change ESR spectrometers and created new fields. ESR dating and dosimetry become practical since the JEOL first used the Gun diode instead of a vacuum tube klystron as a microwave oscillator. JEOL's use of TE100 cylindrical cavity with a high Q-value and a large sample volume as a standard attachment gave advantages over others in applications to geosciences

When I started ESR dating of stalactite and stalagmite at Akiyoshi cave, I was said that ESR spectroscopy is to analyze radical species by separating signals by magnetic field (x-axis) and not to take the amplitude of the derivative line. Certainly, the stability of the spectrometer did not allow the derivation of the equivalent and hence I used the standard signal of Mn²⁺ for intensity calibration which was patented in 1980. The intensity became a reliable quantity contrary to the old spectrometers.

I organizes a small symposium specified as *The First International Symposium on ESR Dating*, including dosimetry, at Ube-Akiyoshi, Yamaguchi University, Japan, in 1985. The fields of ESR dating, radiation dosimetry and imaging have progressed remarkably in the last two decades. An *International Symposium on ESR Dosimetry and Dating (2001-ESRDD-Osaka)* was organized to summarize the quarter century of ESR dating and half a century of ESR dosimetry and show new prospects at the start of 21st century.

History of ESR Dating and Dosimetry Symposia

The applicability of ESR techniques was first recognized as a potential method of dating similar to TL for paleo-anthropology in French CNRS Congress at Tautavel in 1981. The *Specialist Seminar (SS) on TL Dating* included ESR from its 3rd symposium. The proceeding volumes are listed below for references.

1982	<i>3rd SS on TL & ESR Dating</i>	Helsingor, Denmark	<i>PACT</i> 9 (1983)
1984	<i>4th SS on TL & ESR Dating</i>	Worm, Germany	<i>Nuclear Tracks</i> 14 (1985)
1987	<i>5th SS on TL & ESR Dating</i>	Cambridge, UK	<i>Quatern.Sci. Rev.</i> 7 (1988)
1990	<i>6th SS on TL & ESR Dating</i>	Clermont, France	<i>Radiat. Meas.</i> 18 (1991)
1993	<i>7th SS on TL & ESR Dating</i>	Krems, Austria	<i>Quatn. Geochr.</i> 13 (1994)
			<i>Radiat. Meas.</i> 23 (1994)
1996	<i>8th Intrn. Conf. Luminescence and ESR Dating</i>	Canberra, Australia	<i>Quatn. Geochr.</i> 16 (1997)
			<i>Radiat. Meas.</i> 27 (1997)
1999	<i>9th I.C. ESR and Optical Dating</i>	Rome, Italy	<i>Quatn.Geochr.</i> 19 (2000).

Radiat. Meas. **32** (2000)

The First International Symposium on ESR Dating, including dosimetry, generated the Proceeding Book, ESR Dating and Dosimetry (Ikeya and Miki ed. 1985) with a complete bibliography up to 1985. Dosimetry people joined and continued International. Symp. on ESR Dosimetry and Applications. Researchers worked hard and the fields were stimulated by two Intern. Symposia, which developed the method rapidly at the early stage of this new field. Followings are Symposia proceedings with the volume and number and publication years for references.

1985	1st <i>ESR Dating (including dosimetry)</i> Akiyoshi-Yamaguchi, Japan	<i>ESR Dating and Dosimetry</i> (1985)
1988	2nd <i>ESR Dosimetry and Applications</i> GFS, Munchen Germany	<i>Appl. Radiat. Isot.</i> 40-10/12 (1989)
1991	3rd <i>ESRDA</i> , NIST, Maryland, USA	<i>Appl. Radiat. Isot.</i> 44-1/2 (1993)
1995	4th <i>ESRDA</i> , GFS Munchen Germany	<i>Appl. Radiat. Isot.</i> 47-11/12 (1997)
1998	5th <i>ESRDA</i> Obninsk, Russia	<i>Appl. Radiat. Isot.</i> 52-5 (2000)
2001	Satellite A of the 3 rd Asia-Pacific (ESR Dosimetry & Dating, Osaka, Japan)	<i>EPR/ESR Symposium</i> <i>ESR Appl. Metrol.</i> 17 (2001)

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and can be downloaded in the autumn of 2002. The 6th ESRDA will be held by Dr. S. Watanabe at the Univ. Sao Paulo, Brazil presumably at the Igas Waterfall.

A brief review and new prospects will be given according to the main topics.¹⁾

- 1) Tokai JCO, Chernobyl accidents and A-bomb radiation
Special lecture on JCO accident and its dosimetry, Chernobyl, Semipalachinsk, etc.
- 2) Radiation effects of minerals and basic studies on waste depository
What ESR and optical methods can do to assess the safety of radioactive waste.
- 3) ESR and optical dating in geosciences toward 21st century
Summary and new prospects of ESR & Optical dating in geosciences.
- 4) ESR and optical dating and dosimetry in planetary sciences
Dosimetry in space missions and noble methods in planetary material survey
- 5) ESR imaging with applications from semiconductors to fossils
Review of ESR imaging and their applications for interdisciplinary studies
- 6) New materials ESR dosimeters and for food irradiation monitoring
Tissue equivalent alkali-organic acids compounds for ESR dosimeter.

Prospects at the Breakthroughs in 21st century

I should not look back the past in 20th century which had been described in my book (Ikeya, 2001) but look forward for future development in 21st century and give a lecture

summarizing past and forecasting future prospects at the breakthroughs in 21st century.

(a) Applications to Planetary Material Survey

Remote sensing methods using TL and OSL have been proposed for future remote dating from a long distance in a planetary survey. New techniques of R-TL and R-OSL were investigated with infrared CO₂ laser beam as a heat source at a distance of 1 m and He-Ne laser to stimulate the material at a distance of 0.1 m, respectively. The upper limits of the distance in an asteroid survey were estimated using the laboratory data of TL and OSL of the Allende meteorite. The estimates are that 4×10^4 photons s⁻¹ for the R-TL and 6×10^3 photons s⁻¹ for the R-OSL could be detected at the distance of 100 m by collecting luminescence using a mirror with an area of 1 m². The upper limit of the age range in the R-TL and R-OSL dating in most silicate minerals may be 10⁵ ~ 10⁶ years considering the saturation properties due to artificial irradiation in those of terrestrial. It would be possible to take R-TL and R-OSL apparatus on board a rover for surveying lunar or planet surface dating.

(b) ESR dosimetry of water and applications to dating and environmental study

Norizawa et al/ (2001) studied irradiated water by freezing them. No ESR signal is observed since radicals react quickly. However, the dimmer of OH can be reproduced by low temperature UV-illumination from H₂O₂. One can estimate the irradiated water. ESR dating of water might be made in future after extensive studies.

(C) Monitoring Irradiated Foods by D-P Conversion

Food irradiation is a method not to oppose emotionally but to compare the risk and benefit. We have to compare the relative toxic effect of preservative drugs with irradiation. Radicals generate cancers and therefore a method of monitoring radiation dose to check illegal overdosed foods must be developed. Our ESR dosimetry is an ideal especially when we use less expensive spectromewter with a permanent magnet Ne-B-Fe alloy (Neomax) (Ikeya et al, 19xx).

Practical application to monitoring irradiated food has just been done in this laboratory for several spices by irradiating them by UV light. The slight change of the ESR spectra of some foods are due to unstable radical species in irradiated food. Low temperature UV-irradiation generated some specific signals only in irradiated foods. This may be used as a simple method of checking irradiated foods at custom (Hirai et al. 2001).

CONCLUSION

Our prospects based on the past knowledge in our fields are generally very pessimistic. If we look our fields from other fields, new prospects may appear as cross disciplinary subjects in the interdisciplinary fields. However, strategically conceived breakthroughs are

limited in a scale to a decade at the longest. Real breakthrough based on wisdoms would suddenly come as lightning after a long time effort and dedication accompanying pains, as though presented by God or gods for your efforts. We must wait for such breakthroughs by dedicating ourselves and believing that someday we can get an insight leading to a big discovery. Many of us gave up research by facing difficulties, becoming too busy in education and/or administration. I hope to urge young scientists to wait in difficulties under pains till the bright sunshine shed light on our work.

REFERENCES

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- 2) M. Hirai, N. Whitehead, A. Tani, C. Yamanaka and M. Ikeya: *New prospects of ESR Dosimetry and dating in 21st Century*. Proceedings of 2001-ESRDD-Osaka (2002): Free download from our web site <http://quarz.ess.sci.osaka-u.ac.jp/>