Invited, Review

45 Years in Monte Carlo Simulation for Microbeam Analysis - A personal retrospective review –

Ryuichi Shimizu
International Institute for Advanced Studies,
9-3 Kizugawa-dai, Kizugawa-City, Kyoto, 619-0225, Japan
shimizu@iias.or.jp

(Received : October 4, 2010 ; Accepted : January 11, 2011)

Monte Carlo (MC) simulations for microbeam analysis, in which the author has been involved for 45 years, are retrospectively reviewed by tracing the development of simulations models for describing complicated scattering processes of incident projectiles (electron, ion, etc.) in matter. The simulation model is based on the uses of theoretical expressions which describe elastic and inelastic scattering, respectively, no matter whether incident projectile be electron or ion.

MC simulation modellings of different types are outlined by presenting applications to microbeam analysis with primary electrons and ions, respectively, drawing attention into the close correlation between a new modeling and a new micoanalytical instrumentation that was marketed at different times.

Finally, the author takes a liberty to propose an international cooperative joint work for database construction of secondary electron yield, by introducing the working group activities which the JSPS-141 committee (microbeam analysis) has supported since 2009.

1. Introduction

Monte Carlo (MC) simulation has been widely used as the most powerful approach for microbeam analysis, shedding an intimate insight into basic mechanism in signal formation. This approach turned out to be very useful to establish quantitative analyses as well as date-base construction for quantification.

This paper outlines retrospectively the development of the simulation models for describing complicated scattering processes of incident projectiles (electron, ion, etc.) and photon (X-rays) in matter by focussing to those works in which the author has been involved for 45 years.

2. Monte Carlo simulations

In Table 1 basic studies on MC simulation modellings for microanalysis, in which the author had been involved in the developments, are listed in the third column together with inventions of microanalytical instruments (1st column) and academic activities in Japan (2nd column). The Table allows us to see very close correlations between appearance of new instrumentations and development of MC modelling of signal generation observed using a given instrument.

Concerning the scattering processes of incident projectiles, no matter whether it be an electron or ion, are basically described by two processes, i.e. elastic scattering and inelastic scattering. Modellings for electrons and ions are described below.

2.1 Electron beam

The MC modelling describing the scattering processes of incident electrons in matter had started by using Rutherford scattering formula and Bethe's stopping power equation for elastic and inelastic scatterings, respectively. Particularly, in the beginning stage of the development (mid 1960's) when the restriction for computing time was very crucial, so-called multiple scattering model had been studied. The modellings developed since then are briefly described in chronological order as follows:

(1) Modelling I: Multiple scattering model (screened Rutherford scattering formula + Bethe's stopping power equation) (a) -- depth distribution functions

of characteristic X-rays and backscattering coefficients used for quantitative corrections in electron probe microanalysis (EPMA).

(2) Modelling II: Single scattering model (screened Rutherford scattering formula + Bethe's stopping power equation) (b, c) -- electron beam lithography (proximity effect) and scanning electron microscopy (ultimate spacial resolution).

(3) Modelling III: Mott scattering formula + Bethe's stopping power (i, j) -- quantitative correction for Auger electron spectroscopy (AES).

(4) Modelling IV: Mott scattering formula + Dielectric function (n) -- surface electron spectroscopy (REELS-spectrum, Energy Loss spectra). It is worthy to note that the use of Mott-scattering formula owed to the Doctrate-Thesis of Y. Yamazaki (1977, Osaka University) (h), in which the source program for calculation of Mott scattering cross-sections was presented in Appendix.

This Modelling III was, first, used in MC calculation to provide backscattering corrections factors which is indispensable for quantitative analysis by AES and published as the database by Shimizu and Ichimura in 1977.

The Modelling IV has also been used to describe more precisely electron energy loss spectroscopy (EELS)-spectra and, in particular, for deriving excitation function from REELS- and X-ray photoelectron spectroscopy (XPS)-spectra by applying the quasi-Landau formulation (p).

2.2 Ion beam

With respect to the MC simulation as applied to ion beam, the modelling based on the single scattering model had, first, been reported in the beginning of 1970's by Ishitani and Shimizu (c), along with rapid expansion of the use of secondary ion mass spectrometly (SIMS) which appeared quite at sudden in market in 1970. Basics of the MC-modelling is as follows:

(5) Modelling V: Rutherford scattering formula + Lindhard's equation (in place of the Bethe's stopping power in Modelling II). This modelling has also done very much to shed an intimate insight into the basic mechanism of ion bombardment phenomena, e.g., depth distribution of implanted ion (c), atomic mixing process (c), altered layer formation on specimen surface under ion bombardment (m), etc. Since then, this modelling V has been widely used in microanalysis by ion beam bombardment (SIMS, Depth profiling, Surface modification, etc.) as yet without any essential changes in the model.

3. Working group activities for database construction of secondary electron yield

Introducing the working group activities for database construction of secondary electron yields established in 2009 by Japan Society for Promotion of Sciences (JSPS)-141 Committee (Microbeam Analysis), JSPS141-WG-SEY)(1), the author would like to take liberty to propose an international cooperative joint work for the data-base construction on secondary electron yield.

This proposal (2) is based on the well known theoretical expression

$$\delta(Ep) = K\int_0^\infty \left[\frac{dE}{dz}\right]_{Ep} e^{-\alpha z} dz \qquad (1)$$

which has been recognized to describe the SEY with considerable accuracy since Broody had proposed in 1950(3). K is the secondary electron emission coefficient and α, the decay constant describing absorption of SEs during escaping process before coming out to vacuum. Since the energy dissipation in depth $[dE/dz]_{\mathrm{Ep}}$ can be calculated from MC simulation with considerable accuracy, the question remained is how to derive K and α from equation (1) for given experimental data δ(Ep). However, it has been long believed that K and α are independent with each other and ,therefore, one needs another experimental data to back up *δ(Ep)* as Bronshtein and Segal(4) proposed by measuring SEY and backscattering coefficient for a specimen, on which the other material is deposited layer by layer. This requires laborious work and sophisticated experimental technique.

The detailed examination based on precise MC calculation of $[dE/dz]_{\mathrm{Ep}}$, however, has revealed that α controls the peak position, $\delta(E_m)$, in which E_m is the primary electron energy providing the maximum SEY, $\delta m=\delta(Em)$. This enables us to derive the best fit value ,α_0, of α from MC simulation of eq.(1), which provides the best fit to the peak position of $\delta(Ep)$ obtained in experiment. Once α_0 is decided, K can easily be derived from

$$\delta(E_m) = K\int_0^\infty \left[\frac{dE}{dz}\right]_{E_m} e^{-\alpha_0 z} dz \qquad (2),$$

leading to the construction of the data base (K, α) as illustrated in Fig.1 and Table 2. More details are described in reference(1). It is noted that the set of (K, α) allows to provide SEY for given experimental condition , various angles of incidence, various primary energies, even for multi-layered specimen, etc.

The JSPS141-WG-SEY aims at measuring $\delta(Ep)$ for insulating materials by using charge amplifier with pulsed primary electron beam to derive the set of material constants, (K, α).

Since details of the activities of the JSPS141-WG-SEY is going to be reported by T.Nagatomi, chairperson of the WG, in this symposium, the author is hoping that the partipants of this symposium find the proposal well worthy for consideration.

4. Acknowledgements

The author acknowledges his heartiest gratitude to those colleagues who had worked with him in their doctorate works in Osaka University(1965～2000) and in Osaka Institute of Technology(2000～2007) over 45 years.

Kenji Murata, Tohru Ishitani, Takanori Koshikawa, Yasunori Yamazaki, Suk Tai Kang, Shingo Ichimura, Hee Jae Kang, Ding Ze-jun, Hideki Yoshikawa, Takaharu Nagatomi, Hyung-IK Lee, Akihiro Yoshioka, Takeshi Iyasu, Yoshikazu Yamaguchi.

Doctrate Theses:

Kenji Murata;「Studies on Behavior of Incident Electrons and Generation of X-rays in EPMA-specimens」(1966, Osaka University, in Japanese)

Tohru Ishitani;「Interaction of Ion Beam with Solid Surfaces」(1972, Osaka University, in English)

Takanori Koshikawa「Studies on Secondary Electron Emission in Surface Analysis」(1973, Osaka University, in Japanese)

Yasunori Yamazaki;「Studies on Electron Scattering by Mercury Atoms and Electron Spin Polarization Detector」(1977, Osaka University, in English)

Suk Tai Kang;「Investigation on Basic Problems in Secondary Ion Mass Spectrometry」(1978, Osaka University, in English)

Shingo Ichimura;「Basic Study of Scanning Auger Electron Microscopy」(1980, Osaka University, in English)

Hee Jea Kang;「Study on Sputtering Processes in Solid Surfaces under Ion Bomberdment」(1984, Osaka University, in English)

Ding Ze-jun;「Fundamental Studies on the Interactions of kV electrons with Solids for Applications to Electrons Spectroscopies」(1990, Osaka University, in English)

Hideki Yoshikawa;「Fundamental Study of Spectrum Analysis in Electron Spectrometry at Solid Surface」(1991, Osaka University, in Japanese)

Takaharu Nagatomi;「Study on Surface Excitation in Surface Electron Spectroscopy」(1998, Osaka University, in Japanese)

Hyung-IK Lee;「Studies on Ion Beam Induced Effects in Sputter Depth Profiling」(1998, Osaka University, in English)

Akihiro Yoshioka;「Object-oriented Monte Carlo simulation programming and its applications to optimum designing of high power X-ray source by the aid of visualization」(2004, Osaka Institute of Technology, in Japanese)

Takeshi Iyasu;「Computer Simulations Programming for Secondary Electron Emission and Data-base Construction」(2007, Osaka Institute of Technology, in Japanese)

Yoshikazu Yamaguchi;「Development of Computer Simulations Program of Optimum Desgning of X-ray Microscopy and It's Application」(2008, Osaka Institute of Technology, in Japanese)

References

[1] T.Nagatomi, K.Goto, R.Shimizu, et al., *Surf. Interface Analysis* **42,** 1541 (2010).

[2] T.Iyasu and R.Shimizu, *Journal of Surf. Anal.* **13**, 200 (2006).

[3] E.M.Baroody, *Phys.Rev.* **78**, 780 (1950).

[4] M.Bronshtein and R.B.Segal, *Sov. Phys. Doklady* **3**, 1184 (1958).

5th International Symposium on Practical Surface Analysis (PSA-10) & 7th Korea-Japan International Conference on Surface Analysis
organized by
Surface Analysis Society of Japan (SASJ) & The Korean Vacuum Society (KVS)

PSA-10

Table 1. Chronological Table of microbeam analytical instruments appeared, Academic activities in Japan, and Monte Carlo simulation modellings.

	Instrumentation	Academic activities	Basic studies
1951	Electron probe microanalyzer [EPMA] Castaing, PhD-Thesis		
1955	Review paper (Ōyobutsuri) Introduction of EPMA G. Shinoda 1st CAMECA EPMA → ONERA		
1956		1st Int. Cong. on X-ray Optics & Microanalysis (UK)	
1957	Development of EPMA (U. Tokyo)		
1961	CAMECA-EPMA (14th) (NIMS)	Grants-in-Aid for Sci. Res. "Study on EPMA" (Y. Sakaki)	
1962	Hitach-EPMA (1st) (NTT, Tohoku. U.) ARL-EPMA (Yahata Iron & Steel)		
1963	JEOL JAX-3 (Osaka U., U. Tokyo) Akashi TRA-25 (Waseda U.)	Grants-in-Aid for Sci. Res. "Study on EPMA" (G. Shinoda)	
1965		4th Int. Cong. on X-ray Optics & Microanalysis (Paris) 1st Monte Carlo calculation of generation of characteristic X-rays (Bishop, Shimizu et. al.)	Monte Carlo simulation of characteristic X-ray generation (Shimizu, Shinoda & Murata) [(a)]
1968		1st US-Japan Joint Seminar on EPMA (Hawaii, November) [Shinoda, Wittry]	
1969		1st US-Japan Joint Seminar on SEM (Berkeley, November) [Maruse, Everhart]	
1970	Secondary Ion Mass Spectrometry [SIMS] ARL-IMMA (H. Liebl) CAMECA-IMA (R. Castaing) Hitachi-IMA (H. Tamura)	1st Int. Conf. on SIMS (Brussel, August) First commercial SIMS's (ARL, CAMECA, Hitachi) Apolo 11 project: EPMA analysis (U. Tokyo) : SIMS analysis (ARL Cris Andersen)	
1971		4th Int. Cong. on X-ray Optics & Microanalysis (Osaka, G. Shinoda)	MC simulation (SEM) (Shimizu & Murata) [(b)] MC simulation of ion scattering (Ishitani, Shimizu & Murata) [(c)]
1972		2nd US-Japan Joint Seminar on SEM (Takarazuka, November) [Sakaki, Everhart]	Monte Carlo simulation (electron beam lithography) (Shimizu & Everhart) [(d)]
1973		2nd US-Japan Joint Seminar on EPMA (Hawaii, September) [Shinoda, Wittry]	
1974		JSPS-141 Committee (Microbeam Analysis) (chair: Y. Sakaki)	MC simulation of atomic mixing (Ishitani et. al.) [(e)] MC simulation (secondary electron) (Koshikawa & Shimizu) [(f)]
1975		1st US-Japan Joint Seminar on SIMS (Hawaii, October) [Someno, Evans]	MC simulation of Dynamical effect in TEM (Kamiya & Shimizu) [(g)]
1976		US-Japan cooporative SIMS-Analysis for Fe-Cr-X Steel samples	
1977			Mott scattering cross section (Yamazaki Ph D Thesis) [(h)] → NIST data base* MC simulation based on use of Mott-scattering cross-section (Shimizu et. al.) [(i)]
1978		2nd US-Japan Joint Seminar on SIMS (Takarazuka, October) [Someno, Wittry]	
1980			MC simulation for Quantification by AES (Shimizu et. al.) [(j)]
1981			Data base for electron backscattering

			effect (Shimizu & Ichimura) (k) → NIST data base*
1983		4th Int. Conf. on SIMS (Minoh, J. Okano)	Direct MC simulation of Inelastic Scattering Processes (Shimizu & Ichimura) (l)
1984			MC simulation of Ion Sputtering in Alloys (Kang, Kawatoh & Shimizu) (m)
1985		VAMAS (TWA- Surface Chemical Analysis) (JSPS 141 Committee)	
1987			Dielectric function for energy loss process (Ding, Shimizu & Ichimura)(n)
1989			MC simulation based on use of Dielectric function for energy loss process (Ding & Shimizu) (o)
1992		ISO-TC201 (Surface Chemical Analysis) Secretariat: Japan ISO-TC201 (Microbeam Analysis) Secretariat: China	MC analysis for quasi-Landau formulation (Excitation fun. → database proposed) (Yoshikawa, Ding & Shimizu) (p)
1993			MC simulation of X-ray spectra from Rh-Cu-targets (Araki, Kimura & Shimizu) (q)
1995	DIMP-EM (Ikuta et. al.)		Dynamic MC simulation of Depth Profiling (H-I. Lee et. al.) (r)
1998	Aberration corrected EM (Rose et. al.)		
2005			MC simulation for Data base of Secondary Electron Emission (Secondary electron emission coefficient → database proposed) (Iyasu, Inoue & Shimizu) (s)

(a) R. Shimizu, K. Murata & G. Shinoda: in R.Castaing, P. Desdamps and J. Philibert (Eds.), X-ray Optics and Microanalysis (Herman, Paris, 1965) p.127.
(b) R. Shimizu and K. Murata: J. Appl. Phys., 42, 387 (1971).
(c) T. Ishitani, R. Shimizu and K. Murata: Japan J. Appl. Phys. 10, 1464 (1971).
(d) R. Shimizu and T. E. Everhart: Optik 36, (1972) 59.
(e) T. Ishitani and R. Shimizu: Phys. Lett., 46A, 487 (1974).
(f) T. Koshikawa and R. Shimizu: J. Phys. D: Appl. Phys. 7, 726 (1974).
(g) Y. Kamiya and R. Shimizu: Japan J. Appl. Phys. 15, 2067(1975).
(h) Y. Yamazaki: Doctorate Thesis (Osaka Univ. Faculty of Engineering, 1977).
(i) R. Shimizu, M. Aratama, S. Ichimura, Y. Yamazaki and T Ikuta: Appl. Phys. Lett. 31, 629 (1977).
(j) S. Ichimura, M. Aratama and R. Shimizu: J. Appl. Phys., 51, 2853 (1980).
(k) R. Shimizu and S. Ichimura: Quantitative Analysis by Auger Electron Spectroscopy – Monte Carlo Calculation of Electron Backscattering Effects – (The Toyota Foundation, 1981).
(l) R. Shimizu and S. Ichimura: Surface Sci., 133, 250 (1983).
(m) H. J. Kang, E. Kawatoh and R. Shimizu: Japan J. Appl. Phys. 144, 1409 (1985).
(n) Z.-J. Ding, S. Ichimura and R. Shimizu, Surf. Interface Anal. 10, 253 (1987).
(o) Z.-J. Ding and R. Shimizu: Surface Sci. 222, 313 (1989).
(p) H. Yoshikawa, Z.-J. Ding and R. Shimizu, Surface Sci. 261, 403 (1992).
(q) K. Araki, Y. Kimura and R. Shimizu: Scanning Microscopy Supplement 7, (SEM Inc. AMF O'Hare, 1993) p81.
(r) H.-I. Lee, R. Mitsuhashi, M. Inoue, R. Shimizu and S. Hofmann: Japan J. Appl. Phys. 34, L1010 (1995).
(s) T. Iyasu and R. Shimizu. J. Surface Analysis 14,312(2006)

* Basis of the present of NIST data base

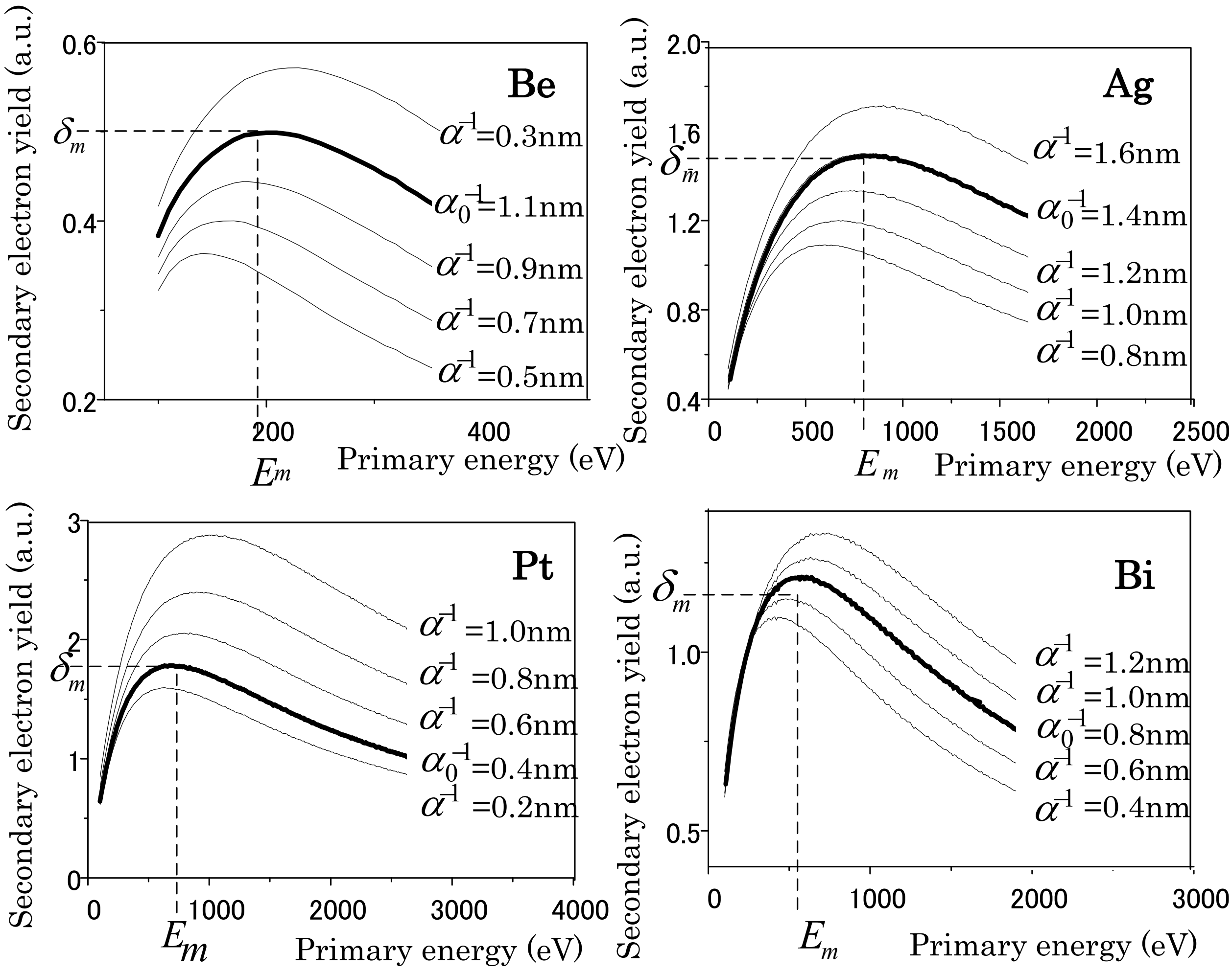

Fig.1. Comparison of Eq.(1) with experimental data (broad line) to find the best fit α.

Table 2. Comparison of the best fit α with experiment.

Material	Bronshtein & Segal α^{-1}(nm)	obtained from the present approach	
		α^{-1}(nm)	κ
Be	0.7	1.1	0.017
Ag	1.0	1.4	0.010
Pt	0.7	0.4	0.086
Bi	0.8	0.8	0.027