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Development of Electron Probe Instrumentation during Those Early Days When Professor Castaing Visited Japan

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Abstract: The development of the electron probe microanalyzer (EPMA) in Japan in the early 1960s, when Prof. R. Castaing visited Japan, is briefly outlined. In 1955, a review article was published by Prof. G. Shinoda in *Oyobutsuri*, the most popular journal in Japan, in which the EPMA was introduced. In 1957, a research group at the University of Tokyo started to develop an EPMA with a Grant-in-Aid for Developmental Research. Their research results led to the funding of a 2-year Grant-in-Aid for Cooperative Research Project (April 1960 to March 1962), which was chaired by Prof. Y. Sakaki. Prof. G. Shinoda who became the chairman of that project in April of 1962 led that group for another year until March of 1963. It was just after the start of the project that Prof. R. Castaing visited Japan in September of 1960 as a representative of the French Mission Culturelle. This visit gave a great push forward for the commercial development of EPMA instruments in Japan. The first three commercial EPMA instruments from Hitachi, JEOL, and Akashi Ltds. were installed in Tohoku, Osaka, and Waseda Universities in 1962, 1963, and 1964, respectively. Photographs of those first commercial EPMA systems, together with a brief description of the activities of the cooperative research projects, are presented.

Key words: Prof. R. Castaing, electron microprobe instruments, cooperative research on electron probe microanalyzer, first commercial electron probe microanalyzers, early 1960s, university–industry cooperation

Introduction

The development of the electron probe microanalyzer (EPMA) in Japan in the early 1960s, when Prof. Raimond Castaing visited Japan, is listed in Table 1. In May 1955, Prof. Shinoda introduced the development of the EPMA in a review article on "Non-Destructive Quantitative Analysis by X-ray Methods" that was published by *Oyobutsuri* (*Journal of Applied Physics*, written in Japanese; Shinoda, 1955). That article included a schematic diagram of the EPMA system. Prof. Shinoda, who referred to the paper of Castaing and Guinier that had been published in 1953 in the journal, *Analytical Chemistry*, drew attention to its construction which included an optical microscope. That article, published in the most popular journal in Japan, was very probably the paper that got the attention of the people involved in the field of material science, in particular, the field of metallurgy, who became interested in EPMA. This landmark article stimulated the development of EPMA in Japan.

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Cooperative Research Projects

In 1957, the funding provided by the Grant-in-Aid for Scientific Developmental Research to the group headed by Profs. T. Akutagawa, I. Gokyuu, and H. Abe was used to

Table 1. Early Research in Japan on the Development of EPMA Instrumentation[a]

Year	Research	Instrumentation
1951	Ph.D. thesis (R. Castaing)	
1955	Review paper (G. Shinoda)	
1957	Grant-in-aid for developmental scientific research (University of Tokyo and Hitachi Group)	
1958		XMA-2 (Hitachi), prototype
1960	Grant-in-aid for cooperative research: "The Study of X-ray Microanalyzer" (April 1960–March 1962, chaired by Y. Sakaki) Visit of R. Castaing to Japan (September 18–October 4, 1960)	
1961		JXA-2 (JEOL), prototype
1962	Grant-in-aid for cooperative research: "The Study of X-ray Microanalyzer" (April 1962–March 1963, chaired by G. Shinoda)	XMA-3 (Hitachi), Tohoku University (Sanbongi and Omori's group)
1963		JXA-3 (JEOL), Osaka University (Shinoda's group)
1964		TRONALYZER-TRA (Akashi), Waseda University (Ichinokawa's group)

[a]EPMA, electron probe microanalyzer.

develop the EPMA in cooperation with the Hitachi group that was headed by Drs. F. Tadano and H. Watanabe. This project led to the first prototype EPMA made in 1958 in Japan, the XMA-2 (Fig. 1), that Watanabe described in detail in an article published in 1959. Their research results led to the funding of a 2-year Grant-in-Aid for Cooperative Research Project from April 1960 to March 1962 called "The Study of X-ray Microanalyzer," that was chaired by Prof. Y. Sakaki and consisted of 37 members from various universities, national institutes, and industries (unpublished report).

It was just after the start of the Cooperative Research Project that Prof. Castaing visited Japan in September of 1960 as a representative of the French Mission Culturelle. He gave several talks on EPMA in different places, including: (1) Hitachi Central Research Laboratory on the 19th of September; (2) Nagoya University for the members of the Cooperative Research Project on the 23rd and 24th of September (Fig. 2), where the program covered: (i) Application of Magnet-Prism for electron and ion optics, (ii) X-ray microanalyzer (1)—accuracy and reliability, (iii) X-ray microanalyzer (2)—detection of light elements; and (3) Osaka University on the 28th of September. During his stay in Japan from September 18th to October 4th, Prof. Castaing talked about his work on EPMA a total of six times which included two informal presentations. This visit of Professor Castaing gave a great push forward for the commercial development of EPMA instruments in Japan.

Development of Commercial EPMAs

One year after the visit of Prof. Castaing, in 1961, JEOL also reported its first prototype EPMA, JXA-2 (Fig. 3). In 1962, Hitachi installed its first commercial EPMA, the XMA-3 (Fig. 4), at Tohoku University. Soon afterwards, the first commercial EPMA made by JEOL, the JXA-3 (Fig. 5), was installed at Osaka University, and it was followed by the first commercial EPMA (Fig. 6) from the Akashi Co. that was installed at Waseda University in 1964.

Research groups at these Universities, where the first commercial EPMAs from different companies had been installed, started different and unique studies with EPMAs. Profs. Sanbongi and Omori's group at the Research Institute of Mineral Dressing and Metallurgy at Tohoku University in cooperation with the steel industry, have, since then, been heavily involved in applying EPMA to metallurgy, in particular, to establishing the identification of various mineral phases containing lime, ferrous oxide, and silica in self-fluxed sinter that is a principal burden of iron blast furnaces. At Osaka University, Prof. Shinoda's group started to develop a Monte Carlo simulation program with the aim of determining $\phi(\rho z)$-functions theoretically, together with the determination of $\phi(\rho z)$-functions for multilayered specimens with EPMA. The installation of the EPMA at Waseda University initiated Prof. Ichinokawa's work on the instrumental development of electron beam instruments, which has gone on for the past 40 years and

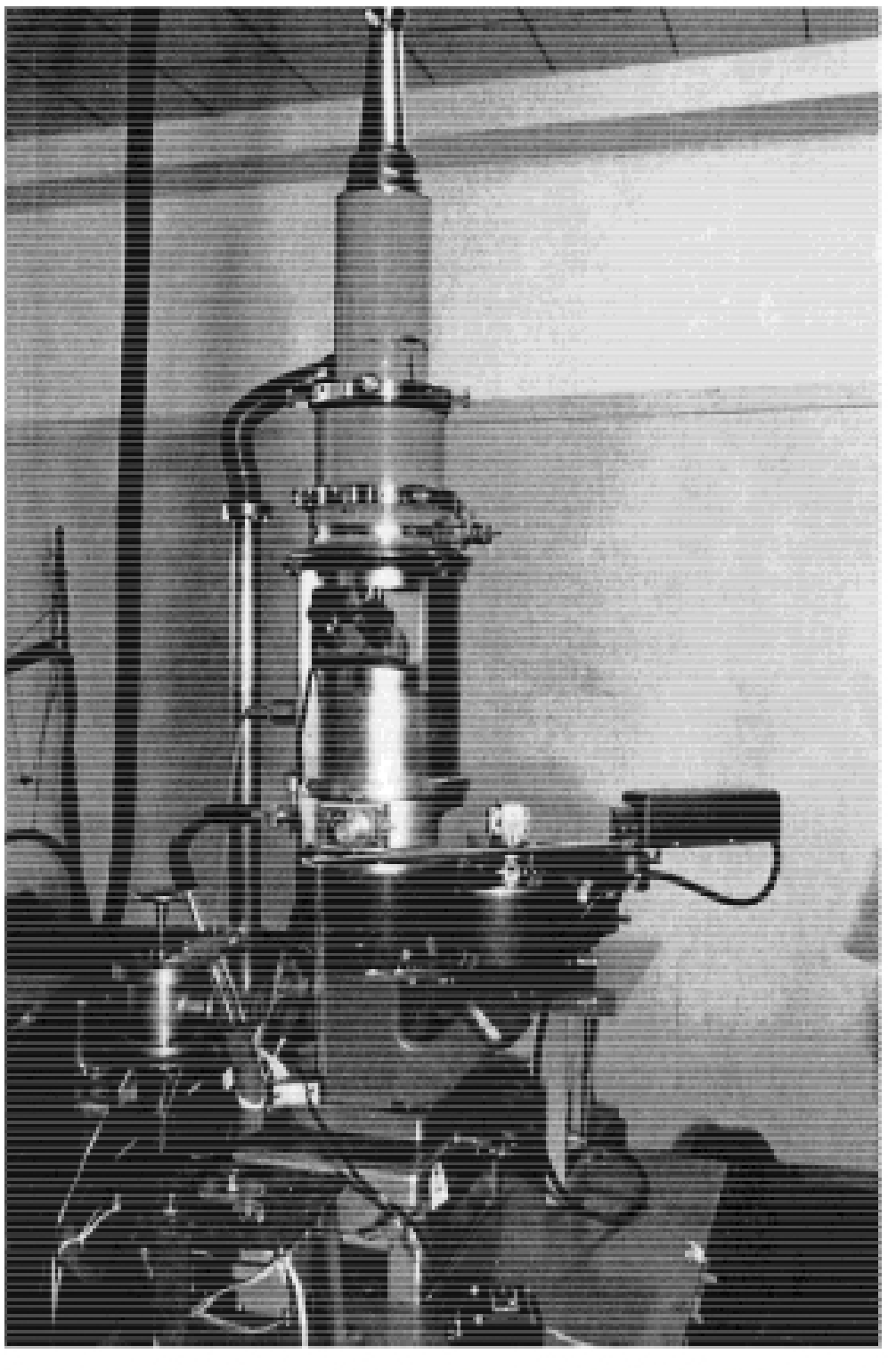

Figure 1. Prototype electron probe microanalyzer (EPMA), XMA-2 (Hitachi), developed in 1958 (Watanabe, 1959).

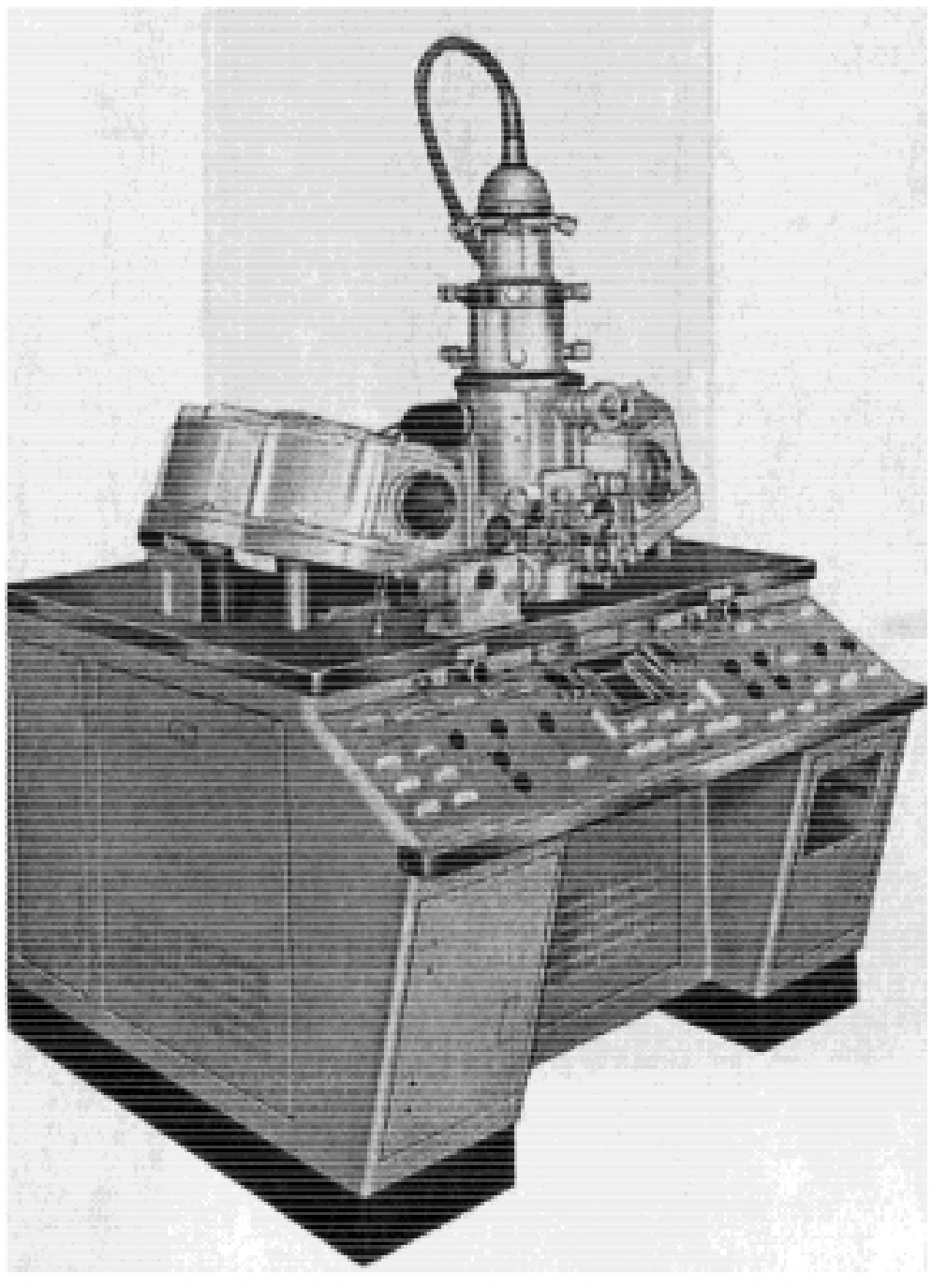

Figure 3. Prototype EPMA-JXA-2 (JEOL) developed in 1961.

Figure 2. Prof. Castaing and members of the Cooperative Research Project in front of the lecture hall, Nagoya University, on September 23, 1960. The board at left announces "Special Lecture by French Mission Culturelle, Professor R. Castaing."

has led to the development of an ultra-high vacuum SEM for surface studies. For that work, the 1999 Distinguished Scientist Award of the Microscopy Society of America was given to Prof. Ichinokawa at the Microscopy and Microanalysis 1999 Meeting.

Very rapid progress in the development of commercial EPMA instruments enabled the Grant-in-Aid for Cooperative Research on "The Study of an X-ray Microanalyzer" to be extended for another year. From April of 1962 to March 1963, this project was chaired by Prof. Shinoda (unpublished report). Although the members were almost the same, the subjects had greatly changed from the development of EPMA instruments to the application of EPMA. This happened at just the time that the steel industry had began to pay keen attention to the use of EPMA for the practical assessment of the quality of steel products. Soon afterwards, the steel industry had started to install EPMA systems as a part of their production lines, leading to the so-called explosive evolution of EPMA in mid-1960s Japan.