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Susumu Hoshinouchi
Mitsubishi Electric Corporation

Akio Yoshida
Mitsubishi Electric Corporation

Akinobu Kawazu
Mitsubishi Electric Corporation

Kohichi Sakurai
Mitsubishi Electric Corporation

Hidenobu Murakami
Mitsubishi Electric Corporation

See next page for additional authors

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Authors

Susumu Hoshinouchi, Akio Yoshida, Akinobu Kawazu, Kohichi Sakurai, Hidenobu Murakami, and Ryuichi Shimizu

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ELECTRON BEAM LITHOGRAPHY FOR LARGE AREA PATTERNING 2: EXPOSURE CHARACTERISTICS OF ELECTRODEPOSITED THICK RESIST

Susumu Hoshinouchi*, Akio Yoshida, Akinobu Kawazu,
Kohichi Sakurai, Hidenobu Murakami and Ryuichi Shimizu(1)

Manufacturing Development Laboratory
Mitsubishi Electric Corporation, Amagasaki, Hyogo, 661 Japan
1)Department of Applied Physics, Faculty of Engineering
Osaka University, Suita, Osaka, 565 Japan

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Abstract

Exposure characteristics of a large field deflection electron beam lithography system have been examined. Thick film mainly composed of unsaturated acrylic resins and prepared by electrodeposition process is proposed as a resist available to large area patterning. It is proved that the 20 μm thick resist is very sensitive to 60 kV electron beam and has the threshold dosage of 2.0×10^{-7} C/cm^2. The spatial contours of equienergy density deposited by electron beam in a 20 μm thick resist on a semi-infinite thick copper substrate are calculated by Monte Carlo simulation for a line source with a lateral beam profile described by Gaussian distribution.

The comparison of Monte Carlo calculations with the experiment has led to conclusions that the line width of the electrodeposited resist is determined by the contour line corresponding to about 1.5×10^{19} eV/cm^3 and that line widths less than 100 μm can be obtained with high stability within a fluctuation of less than ±10 % for the large scanning field of 52 mm x 52 mm.

Key Words: electron beam lithography, large field scanning, electrodeposited thick resist, threshold dosage, resist pattern profile, Monte Carlo, line source electron beam, equienergy density contour, charging, printed wiring board.

*Address for correspondence:
Manufacturing Development Laboratory
Mitsubishi Electric Corporation
1-1, Tsukaguchi-Honmachi 8-Chome
Amagasaki, Hyogo, 661 Japan
Phone No. (06)491-8021
Fax No. (06)499-2813

Introduction

A novel electron beam system has been designed and developed for large area patterning of electronic devices such as printed wiring boards (Hoshinouchi et al.,1990). The prototyped system features the fast and accurate deflection in the quite large field; the deflection field is either 104 mm x 104 mm or 52 mm x 52 mm, the scanning speed can range up to 254 m/s and the beam diameter in full width at half maximum (FWHM) is 35 μm.

The advance of electron beam lithography owes much to the well-balanced progress of equipment, resist material and analysis of exposure phenomena. For practical use of electron beam lithography for the large area electronic devices, therefore, basic studies on resist and its exposure characteristics are needed in parallel with the development of lithography systems.

Resist and its preparation process

In manufacturing of integrated circuit (IC) devices, very thin polymer film of polymethylmethacrylate (PMMA) has been widely utilized as a resist with adequate performance for applying to microfabrication (Hatzakis et al., 1974; Broers,1981). However, this resist film can not be utilized in the large area patterning because the large area electronic devices such as printed wiring boards have far larger area than the IC devices and the circuit patterns of thick Cu are fabricated through severe wet etching process. Hence, another resist, several tens of micrometers thick, should be provided, which is sensitive enough to allow writing of the desired pattern in a reasonable period of time. Furthermore, its coating process should make the resist film strongly adhesive to the Cu layer with excellent uniformity of thickness.

Method to predict resist pattern profile

For analysing exposure phenomena in very thin resist films, Monte Carlo calculations have been used, particularly for predicting exposure profiles (Kyser and Murata,1974). The simulation method was extended to incorporate the time-evolution of the developed pattern by a solvent (Neureuther et al.,1979). These methods have been widely utilized to correct the proximity effects in the manufacturing of IC devices, and can be extended to the manufacturing of large area devices as well, for optimizing the processing.

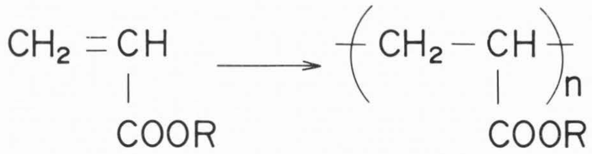
In the present work, we propose a thick negative resist mainly composed of unsaturated acrylic resins as a material and its electrodeposition for the coating process. We will discuss the exposure characteristics of the resist and the direct writing properties of our novel large field deflection electron beam lithography system. Then we describe material and preparation method of the resist and also describe the basic performance of the electron beam system used in this experiments. We present the experimental results on exposure characteristics and spatial resolution, followed by the discussion of the pattern forming phenomena through Monte Carlo simulation, and evaluation of the accuracy of resist patterns fabricated by the newly developed system. Finally, we summarize the present results.

Resist Preparation and Experimental Procedure

Very thin resist films prepared by a spin-coating process have been utilized to fabricate submicron patterns of IC devices by electron beam lithography. In fabricating printed wiring boards, on the other hand, a thick resist with strong adhesion to the substrate is required to etch the thick Cu layer and a preparation process providing uniform coating at the edges and wall surfaces of many small holes to perform the three dimensional wiring. For this, first, novel resist and its preparation process are examined, and, second, the focusing properties of the electron beam system used in the present work is evaluated, because those properties relate strongly to the analyses for direct writing performance.

Composition of The Resist and Its Preparation Process

The main components that contribute to chain cross-linking are unsaturated acrylic resins. Figure 1 shows the monomer and polymer formula of the main components. The electrodeposition process was adopted to form the resist film on Cu clad laminates. An anion type of electrodeposition process was adopted, because the resist suffers attack of acids in the etching process in successive lithography procedure.



$$\mathrm{CH_2=\underset{\displaystyle COOR}{\underset{|}{CH}}} \longrightarrow -\!\left(\mathrm{CH_2-\underset{\displaystyle COOR}{\underset{|}{CH}}}\right)_{\!n}\!-$$

Fig.1 Monomer and polymer formula of the main components in the resist.

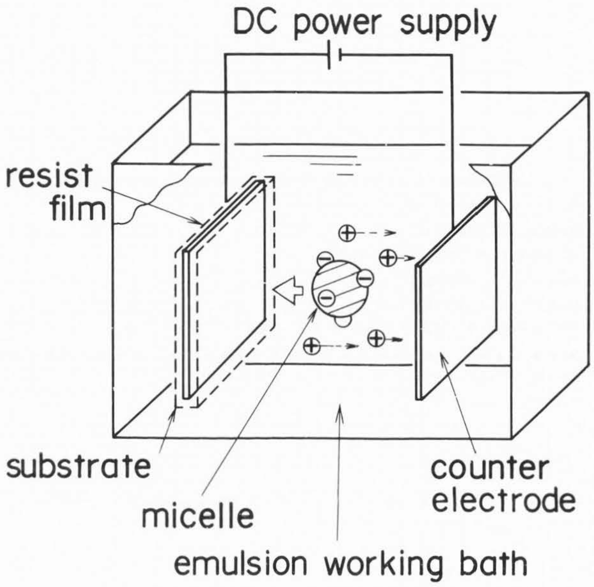


Fig.2 Preparation method of the resist by electrodeposition.

Figure 2 illustrates schematically the principle of electrodeposition. An emulsion working bath is filled with the resist solution. The resist solution contains 15 % solids. The solids are in the form of micelles. Each micelle contains the microenvironment of an organic resist. Upon application of current, the resist micelles begin to migrate within the solution. Reaching the anode, the surface charge on the micelle is neutralized, and the organic material is deposited onto the surface of the exposed copper.

The thickness of the resist film can be controlled by the duration of deposition and the current density. Variation of thickness of the resist fabricated by this process was examined with the Hull cell method. Figure 3 shows the results for various deposition durations, τ. At the first stage of deposition, thickness distribution depending on the distribution of current density was observed. However, the distribution of film thickness becomes uniform as the deposition progresses. In spite of the intentional distribution of current density, thickness of the film is controlled within a fluctuation of less than ±2 μm in the case of a 20 μm thick resist. This property of the electrodeposition can be explained as follows. Deposits in the process are resins with high electrical resistivity, so once fluctuation in film thickness occurs, deposition is localized to the area with relatively thinner thickness. Consequently, the thickness is controlled by itself and sufficient uniformity can be obtained.

Apparatus and Experimental Procedure

The large field deflection electron beam lithography system which has been newly developed (Hoshinouchi et al.,1990) was used in this work. Table 1 shows the basic performance of the system. The maximum beam energy is 60 keV. Two deflection field sizes, 104 mm x 104 mm and 52 mm x 52 mm, can be chosen in this

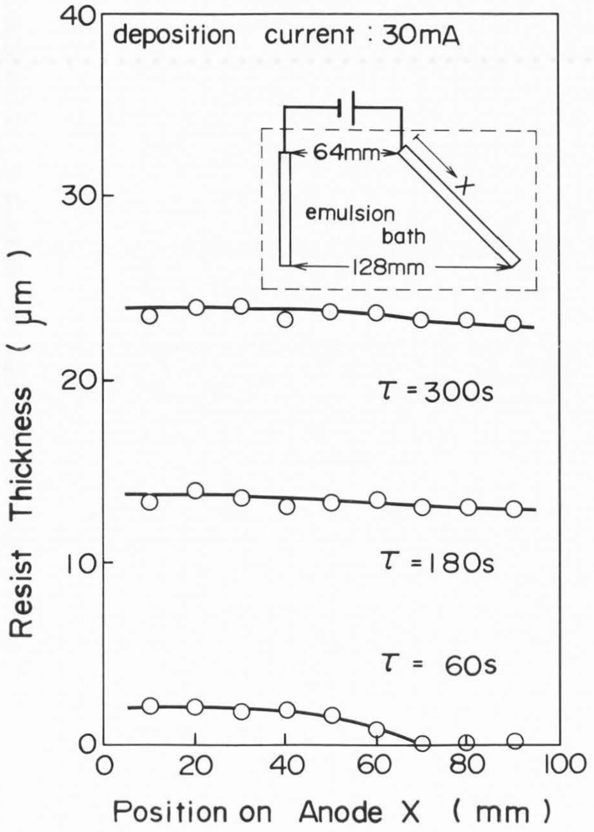


Fig.3 Thickness distribution of the electro-deposited resist prepared by the Hull cell method.

Table 1 Basic performance of the novel e-beam lithography system for large area patterning.

cathode	LaB_6
max. beam energy	60 keV
max. deflection field	104 x 104 mm^2
pixel size	25.4 μm
max. scanning speed	254 m/s

system, corresponding to the required accuracy. In the present work, the latter one was chosen. The scanning speed on the target can range to 254 m/s.

Since the performance of the system has been described in detail elsewhere, only the beam focusing properties are briefly described in this section. The beam intensity profile was obtained by measuring the time transient profile of the reflected electron current from the marker. The time-differentiated profile, therefore, indicates the space-integrated beam intensity profile in the deflection direction. Figure 4(a) shows an example of the results. The beam profile is quite similar to a Gaussian distribution with full width at half maximum of 35 μm. Figure 4(b) shows the measurement of the dependence of the beam diameter, *d*, on both the beam current , *I*, and deflection angle, *α* . The diameter is almost independent on both the beam current and deflection angle. The distribution of beam diameter in the scan area of 52 mm x 52 mm was also measured, to confirm high stability in beam diameter within a fluctuation of less than ±2 μm.

In the experiments of the exposure characteristics and spatial resolution, 20 μm thick resists were prepared on Cu clad laminates by the electrodeposition process. After electron beam exposure by vector scanning under various beam currents and scanning speeds, the resist was developed in a solution of 1 wt% sodium carbonate and remnant water.

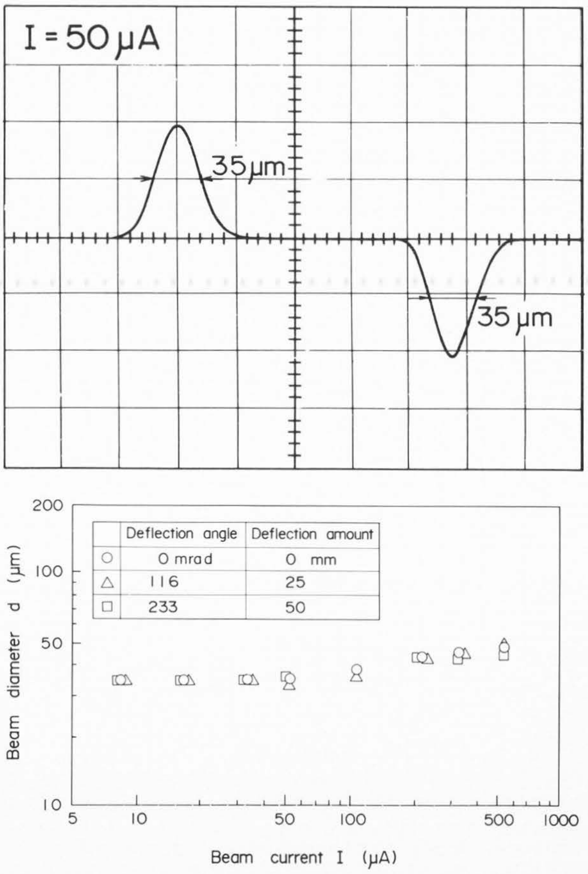


Fig.4 An example of measured beam profile (a) and dependence of beam diameter on beam current and deflection angle (b).

Experimental Results and Discussions

The resist sensitivity to the electron beam exposure was evaluated in terms of electrical charge per unit area of resist film, D (C/cm^2). The resist pattern profiles fabricated under various exposing conditions were measured and compared with the results of Monte Carlo calculations in order to clarify the factor determing the line width. Furthermore, the stability of direct writing was measured to evaluate the utility of both the system and the thick resist. Charging-up of the resist was also examined by charging voltage and accuracy of the beam deflection.

Exposure Characteristics

The electron beam was scanned at an adequate pitch to deposit the electrical charge uniformly on the resist in 5 mm x 20 mm square. The beam current was varied from 1 to 10 μA and the scanning speed, S , from 4 to 254 m/s. Figure 5 shows the sensitivity of 20 μm thick electrodeposited negative resist for 60 kV electrons. Since the resist swells 15 % in volume during the electron exposure and successive development, the measured remaining thickness is normalized by this value. The resist shows high contrast with the threshold dosage of 2.0 x 10^{-7} C/cm^2. This is the lowest exposure, at which no thickness loss is observed after resist development. The threshold dosage of the resist is two or three orders of magnitude lower than those of PMMA resists (Hatzakis et al., 1974). This fact suggests that the 20 μm thick electrodeposited resist is fully sensitive to 60 kV electrons and high speed electron beam lithography is possible with this type of resist.

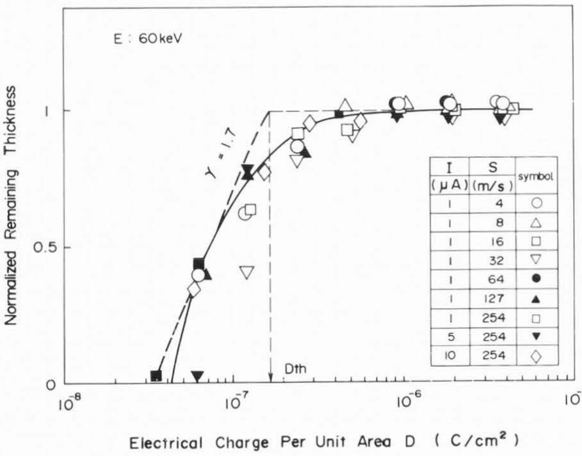


Fig.5 Resist sensitivity.

Spatial Resolution

Measurement of Resist Pattern Profile The line widths of the resist patterns were measured, which were written on the 20 μm thick resist prepared on Cu clad laminates. The line width was defined as edge-to-edge distance at the resist-substrate interface. The resist patterns were written under the following beam parameters; beam current from 1 to 64 μA, beam scanning speed from 4 to 254 m/s. Figure 6 shows an example of a SEM photograph of the resist pattern obtained by this experiment.

Figure 7 shows the results with respect to the influence of the line charge q_l (C/cm) on the line width. This examination has revealed that the line width is determined by q_l, though the dependence is non-linear. The minimum width for lines without thickness loss is 60 μm.

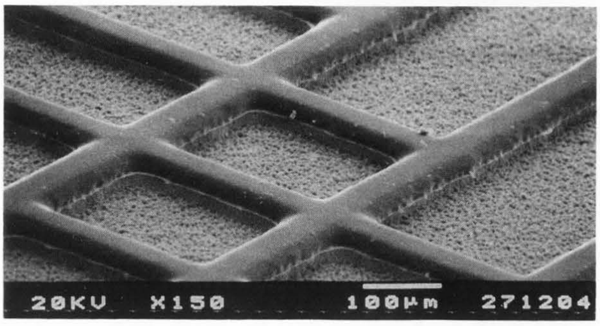


Fig.6 An example of a SEM photograph of the resist pattern.

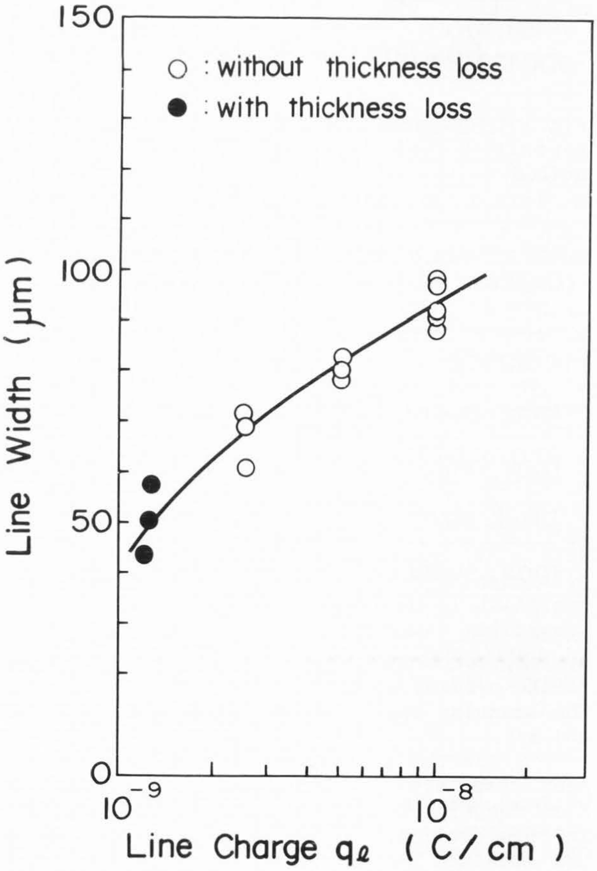


Fig.7 Relation between line width and line charge.

Monte Carlo Simulation Monte Carlo calculations for the scattering and energy dissipation processes of kilovolt electrons penetrating the thick resist have been carried out to understand the phenomena of resist pattern forming. The continuous slowing down approximation (Kyser,1982) and the Rutherford formula for differential cross-section (1) for elastic scattering are also adopted in the present calculations.

$$\left(\frac{d\sigma(E)}{d\Omega}\right)_i = \frac{Zi(Zi+1)e^4}{4E^2(1-\cos\theta+2\beta)^2} \tag{1}$$

where σ : cross-section, Ω: solid angle, Z_i: atomic number, e: elementary electric charge, E: electron energy, θ: conical angle of scattering, β: screening parameter.

The step length between scattering events is given by the mean free path.

Equation (2) which is introduced by F.Rohlich based on the Bethe-Bloch expression is utilized to calculate energy loss by the electron between elastic scattering events along the total trajectory.

$$\frac{dE}{dr} = \sum_{i=1}^{n} \frac{2\pi e^4 Na\rho Zi}{E\cdot Ai}\left\{\ell n\frac{T^2(T+2)}{2(Ji/mc^2)^2} + 1 - \frac{E}{mc^2} + \frac{T^2/8-(2T+1)\ell n2}{(T+1)^2}\right\} \tag{2}$$

where Na: Avogadro's number, ρ: density, A_i: mass number, T: normalized electron energy, J_i: energy of ionization, c: light velosity.

The calculation of energy loss was done until the electron energy decreased to cutoff energy E_{min} = 100 eV. The density, ρ , of the electrodeposited resist is 1.10 g/cm^3. The values for J_i used in the present calcuration are listed in Table 2. Monte Carlo calculations have been carried out for an electron beam source with biaxial summetry around the y-z plane (Kyser and Murata,1974; Kyser and Pyle, 1980). For this calculation, 1,000 electron trajectories were simulated.

Table 2 Constants used in the Monte Carlo calculations.

Atom	Z	J/Z (eV)
H	1	18.7
C	6	13.0
O	8	11.1
Cu	29	10.8

In the calculations, the incident beam profile was described by a Gaussian distribution which corresponds to the real beam profile shown in Fig.4(b). In order to examine the validity of this method, Monte Carlo results for 50 μm thick resist were compared with the experimental results with regard to the dependence of maximum penetrating depth on accelerating voltage. Both the simulations and experiments were carried out for q_l = 5 x 10^{-9} C/cm. The theoretical penetration depths were obtained assuming the threshold energy E_c = 1.5 x 10^{19} eV/cm^3. Experimental data were obtained by peeling off the exposed resist from the Cu substrate and then developing the resist from the surface opposite to the electron impingement. Good agreement between the measured and calculated values was obtained, as shown in Fig.8.

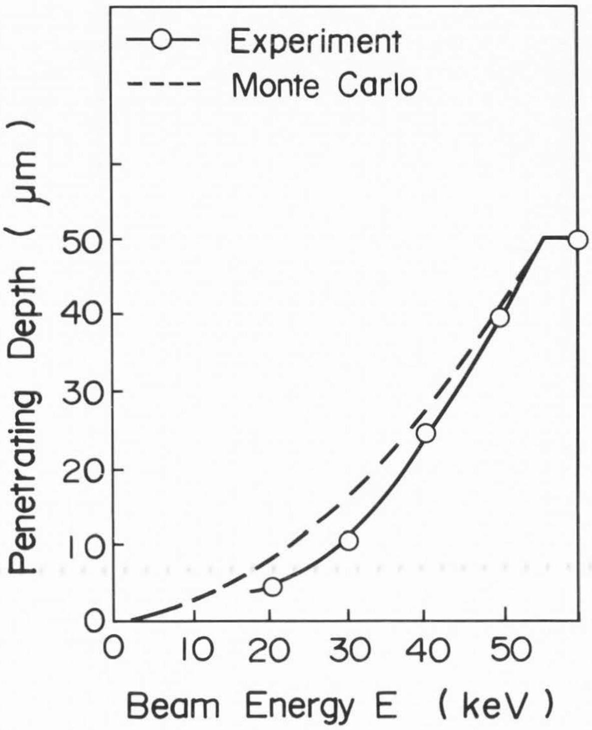


Fig.8 Comparison between Monte Carlo results and experimental results with regard to maximum penetration depth.

Figure 9 shows the Monte Carlo results for equienergy density contours of 1.5 x 10^{19} eV/cm^3 in a 20 μm resist film on a Cu substrate, generated by the line source beam of 60 keV with q_l = 1 x 10^{-9} C/cm. This particular value of energy density was chosen through the matching with experimental value to be discussed. The contours labeled 0.6, 1.3, 2.5 and 5q_l show the movement of equienergy density contours with different values of the line charge. Figure 9 also shows a comparison of the Monte Carlo results with the experimental data. The experimental contours are observed with scanning electron microscopy after developing and cutting the sample in cross-section. The agreement between theory and experiment shown in Fig.9 is generally good.

Figure 10 shows the calculated results for equienergy density contours in both fully thick electrodeposited resist and a 20 μm resist film on a Cu substrate. There is no wide difference between them for contours reaching the surface of the resist, suggesting that the effect of electrons backscattered from the substrate is not dominant and that the lateral profile is determined primarily by the forward scattered electrons.

Figure 11 shows the line width predicted by Monte Carlo simulations. In the present work we were interested in the lateral profiles at the boundary between resist and substrate. We have defined the line width to be the one of the contour with E_c = 1.5 x 10^{19} eV/cm^3 at the depth z = 20 µm. Figure 11 shows also a comparison of the Monte Carlo results with the simulated results.

These results led to the conclusion that the present simulation model predicts the line width of patterns on thick resists with considerable accuracy, well worthy for application to our large field deflection electron beam lithography.

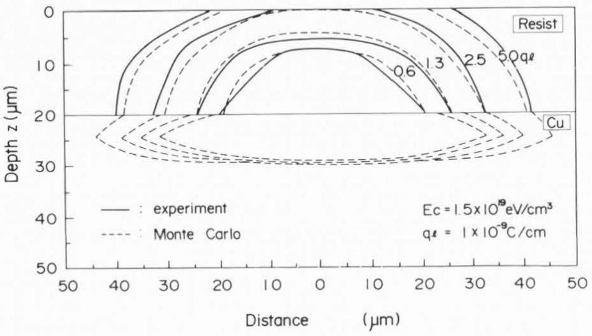


Fig.9 Monte Carlo simulation of equienergy density contours and comparison with experiments.

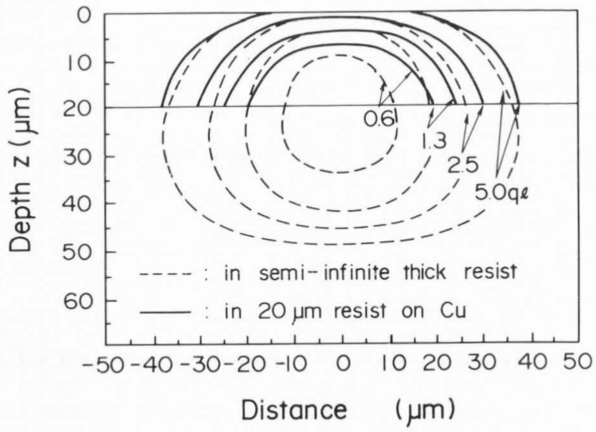


Fig.10 Calculated equienergy density contours in a semi-infinite thick resist and a 20 µm thick resist film on Cu substrate.

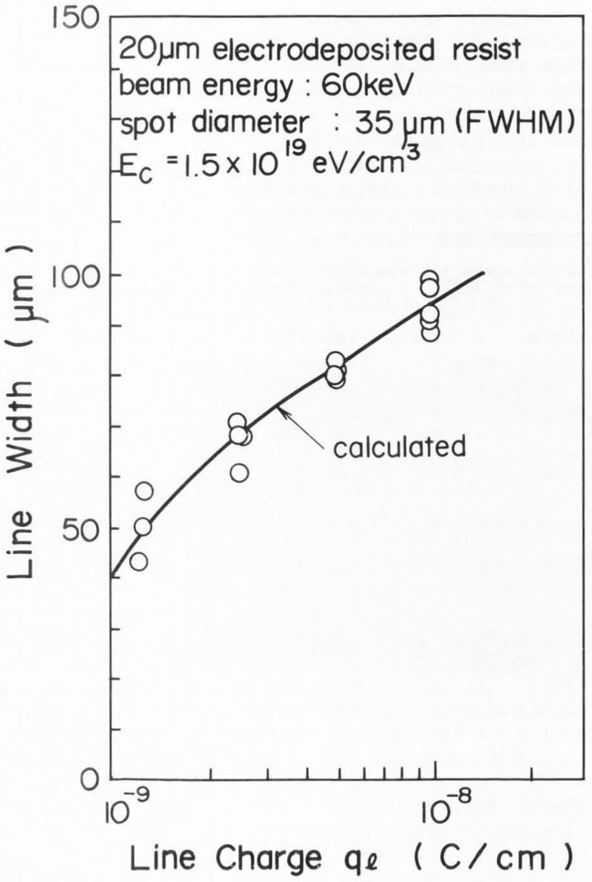


Fig.11 Monte Carlo simulation of the line width.

Accuracy of Direct Writing The novel lithography system developed for large area electronic devices was evaluated with regard to the stability of line width and accuracy of positioning in large field of 52 mm x 52 mm. Figure 12 shows the distribution of line width in the field prepared under q_l = 2 x 10^{-9} C/cm. The distribution can be expressed by a Gaussian distribution with a standard deviation (σ) of 2 µm. It is confirmed that the prototyped system can provide a control for maintaining developed line width within the +10 % (3σ) tolerance.

Usually, error of line positioning from the required position, which originates from charging-up of the resist, is a popular important problem to evaluate the utility of a resist. Several mesh patterns shifted horizontally and vertically by 300 µm were written and positioning accuracy was assessed, leading to the conclusion that every line was written at the desired position within experimental errors. This fact suggests that the charging-up does not influence the positioning in the case of electrodeposited resist. The charging voltage was measured for the resists exposed by sufficient incident electrons, but no charging-up effects were observed in the experiment with the measuring accuracy of 3 V. This fact indicates that the present electrodeposited resist has a slight electrical conductivity. The measured net resistivity of the resist film was 5 x 10^9 Ω-cm. This value is about five orders of magnitude smaller than for conventional resists. This conductivity of the resist is probably due to the ions and water contained in the resist.

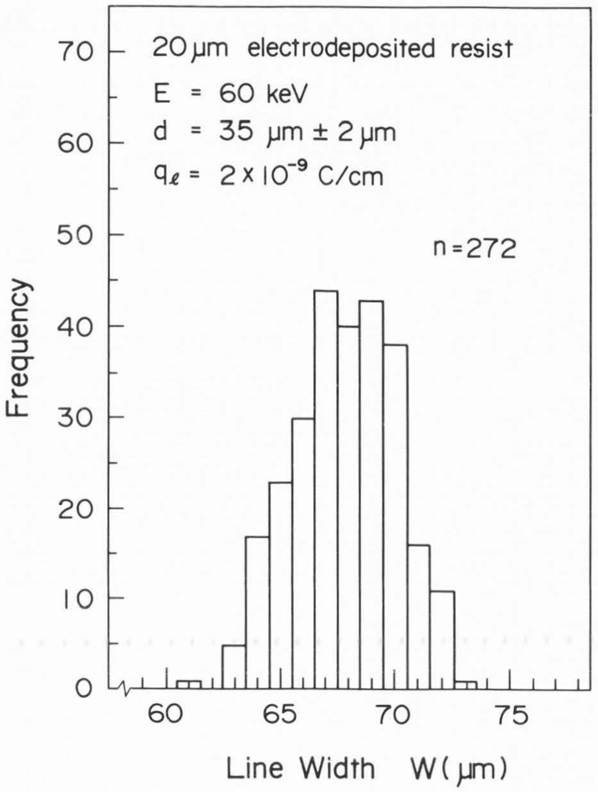


Fig.12 Stability of line width in a scanning field of 52 mm square.

Conclusions

The exposure characteristics of electrodeposited negative resist were examined as a candidate resist for electron beam lithography of large area electronic devices. The results have confirmed the excellent performance of the resist, giving a sensitivity of 2.0 x 10^{-7} C/cm^2, without any charging-up problems.

Monte Carlo simulations were also carried out to compare with experimental data. The results have indicated that the pattern profile is characterized by a critical energy density of 1.5 x 10^{19} eV/cm^3 and the line width can be predicted with considerable accuracy by using this value.

Direct writing properties on the resist of the novel electron beam system were also assessed. Line width less than 100 μm can be obtained with high accuracy.

It is concluded that the present electron beam lithography system and the electrodeposited thick resist are of practical use for large area patterning, meeting the constantly growing demands for greater density and shorter turnaround.

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Discussion with Reviewers

M.G.R. Thomson: After development, the resist wall profile appears to be not very steep. Does this cause variations in linewidth, or other etching problems?

Authors: We found no etching problems in our examinations.

E. Munro: Could you comment on how the performance parameters of your new column and new resist would be expected to vary as a function of the incident beam energy, and what factors led to your choice of 60 keV as the optimum?

Authors: The incident beam energy influences the range and the cross-sectional profile of the resist. The optimum beam energy varies depending on the density and the thickness of the resist and is about 40 keV for the proposed 20 μm thick electrodeposited resist. However, manufacturers of printed wiring boards use 50 μm thick dry film resists in some cases. So, we selected the incident beam energy of 60 keV to stand up under this type of service.

K. Murata: Do you have an idea why the data points in Fig.5 fluctuate so much at low doses?
Authors: The remaining film thickness at low dose depends strongly on the development conditions. Perhaps the fluctuation originates from the fluctuation of the development parameters.

K. Murata: Have you studied the charge-up effect to see if it is a possible reason for the descrepancies between theory and experiment in Fig.8?
Authors: We have not fully studied from this point of view. We consider that the charge-up effect is little because the resist has some electrical conductivity.

K. Murata: Could you comment on what causes the fluctuations of the line widths in Fig.12?
Authors: The measured electron probe size has a fluctuation of ±2 μm in the scan area of 52 mm x 52 mm. Therefore, we think that the greater part of the fluctuations of the widths occurs from the fluctuations of the electron probe size.

K. Murata: The threshold model might be the best one for negative resist pattern analysis because remaining thickness depends on the gel fraction of the negative resist. Is the threshold model appropriate for your negative resist?
Authors: We agree with the reviewer's comment. But we think that the threshold model can apply approximately to the analysis of resist pattern with widths of several tens of micrometers because the g-value of the present resist is about 2 and is much smaller than that of conventional negative resists.