

Surface tension, adhesion and wetting of materials for photolithographic process

J. Bauer and G. Drescher

Institute for Semiconductor Physics, D-15230 Frankfurt (Oder), Germany

M. Illig

OEG G.m.b.H., Ringstr. 1003, D-15236 Frankfurt (Oder), Germany

(Received 18 July 1995; accepted 13 May 1996)

Surface tension of substrates and layers used in semiconductor technology (e.g., Si, SiO₂, Si₃N₄, AlCu, and WTi) as well as surface tension of materials of photolithography as resists and developers were determined by contact angle measurements. Resists and developers could be divided into two groups with polar and nonpolar behavior, respectively. The pretreatment of the substrates (cleaning, dehydration, and exposure to humidity) influences the surface tension. It was found that surface modification by primers, for example, hexamethyldisilazane and trimethylsilyldiethylamine provides stable and reproducible surface tension. To reduce defect density of resist structures, a general optimization approach of surface tension was derived and successfully applied, using Si and AlCu as substrates in a typical photolithographic process. The results show a good adhesion of resist will be achieved if the work of adhesion is greater than 5 dyn/cm. A suggestion is given of process window of the contact angle is given for certain combinations of types of resist and developer.

© 1996 American Vacuum Society.

I. INTRODUCTION

Surface tension of photolithographic materials like resist and developer has a great potential for the photolithographic process itself. For instance, homogeneity of spin coatings, planarization, and adhesion depend directly on surface tension of the materials involved. In this article, the influence of surface tension of photoresists and developers on adhesion and wetting is investigated and a method to optimize the process is suggested. In particular, for reducing feature sizes <1 μm the optimization of adhesion of these small resist structures is a factor of importance to decrease defect density.

Generally the lithographic process requires a modification of substrate surfaces before resist coating to prevent small features from ablation during developing, water rinsing and following wet etch processes. Primers like hexamethyldisilazane (HMDS), trimethylsilyldiethylamine (TMSDEA) and others¹⁻⁴ reduce surface tension of materials used in semiconductor technology like Si, SiO₂, Si₃N₄, AlCu and WTi, etc (Fig. 1). The primer reacts chemically with water deposits held by high-energy substrate surfaces or more closely regarded with bounded OH-groups. Thus surface tension is reduced and so is the possibility of penetration of developer, water, or etchants between resist and the modified substrate surface.

II. THEORETICAL BASICS AND EXPERIMENTAL METHODS

A. Surface tension measurement of solids and fluids

Applying the drop method (Figs. 2 and 3), the nonpolar part of surface tension σ^d (caused by dispersion energy between molecules) and the nonpolar part of surface tension σ^p (caused by dipole interaction, induced dipole moments and

hydrogen bonds) are measured at once (measurement device: SURFTENS from OEG GmbH). The basic additive approach

$$\sigma_{ij} = \sigma_{ij}^p + \sigma_{ij}^d \quad (1)$$

is based on publications of Fowkes,⁵⁻⁸ Gardon,⁹ and Hansen; Beerbower¹⁰ and Meyer *et al.*,^{11,12} and Wu.^{13,14} The model of harmonic mean is used to approximate interaction of low energetic phases (symbols: see Fig. 2).^{13,14}

$$W_{\text{asl}} = (1 + \cos \Theta) \sigma_{\text{gl}} = 4 \left[\frac{\sigma_{\text{gl}}^d \sigma_{\text{gs}}^d}{\sigma_{\text{gl}}^d + \sigma_{\text{gs}}^d} + \frac{\sigma_{\text{gl}}^p \sigma_{\text{gs}}^p}{\sigma_{\text{gl}}^p + \sigma_{\text{gs}}^p} \right] \quad (2)$$

Work of adhesion W_{asl} is defined as reversible work necessary to create an interface area of 1 cm² between two different chemical materials with the phases solid-fluid or to separate them. This work of adhesion is defined by Dupré equation as follows:

$$W_{\text{asl}} = \sigma_{\text{gl}} + \sigma_{\text{gs}} - \sigma_{\text{ls}} \quad (3)$$

In the following, all surfaces are modified by organic compounds. These compounds strongly reduce the spreading pressure. That's why the latter is further neglected ($\pi_s = 0$).^{5,14,15} Using two testing liquids of different polarity, it is possible to determine the surface tension of the substrate by an analytical solution of Eq. (2).¹⁴ In our experiments, water (16–17 MΩ cm): $\sigma_{\text{gl}}^p = 50.7$ dyn/cm, $\sigma_{\text{gl}}^d = 22.1$ dyn/cm and methylene iodide (CH₂I₂): $\sigma_{\text{gl}}^p = 6.7$ dyn/cm, $\sigma_{\text{gl}}^d = 44.1$ dyn/cm were used. If the surface tension of the used substrates is known (σ_{s1} , σ_{s2}), the surface tension of liquids can be determined by solving Eq. (2) using the Newton–Raphson method. For support, a start value calculation was carried out successfully [see Eq. (4a)]:

TABLE I. Overview of kinds of resists and developers in respect to their surface tension based on experimental results (Tables II and III).

	Polar type (type A)		Nonpolar type (type B)	
	σ^p (dyn/cm)	σ^d (dyn/cm)	σ^p (dyn/cm)	σ^d (dyn/cm)
Resist (R)	20	30	10	30
Developer (D)	20	40	10	40

these papers, it was proven that wetting and adhesion are optimal if the polarity of the substrate and the wetting material are equal (polarity $x_i^p = \sigma_i^p / \sigma_i$). In practice the materials are more or less fixed, but modifications of the surface are a possibility to improve adhesion and wetting characteristics. For evaluation purposes, the wetting factor f_w due to Wu¹⁴ is used and is reciprocal to defect density (unwetted area):

$$f_w = (1 - \lambda_{ls} / \sigma_{gs})^{-2}. \quad (9)$$

In the case of spontaneous wetting (contact angle $\Theta=0$) $\lambda_{ls} \geq 0$ and $f_w \geq 1$ are valid. At contact angles $\Theta > 0$ defect density raises up because of $\lambda_{ls} < 0$ and $f_w < 1$. The wetting coefficient λ_{ls} is defined as the difference between Work of adhesion at substrate/liquid and work of cohesion of the liquid:^{14,17}

$$\lambda_{ls} = W_{asl} - W_{cl} = W_{asl} - 2\sigma_{gl}, \quad (10)$$

where W_{cl} is the work of cohesion of the liquid. A further valuation of the wetting may be carried out regarding the contact angle itself. We assume that contact angle $< 62^\circ$ are suited for sufficient wetting.¹⁸

III. RESULTS

A. Surface tension of photochemical materials

The results of measurement of the surface tension of photochemical materials are shown in Tables I–III. I-line resists from HOECHST, SHIPLEY and ALLRESIST were investigated. The finding of surface tension of the liquids is based on several combinations of substrates with the investigated

TABLE III. Surface tension of liquid photoresists and developers.

Liquids	Surface tension	
	σ_{gs}^p (dyn/cm)	σ_{gs}^d (dyn/cm)
Photoresists	1.5–2	33–34
MF501 (type B)	8	39
MFCD (type A)	20	42
AZ351 (1:5) (type A)	23	43
AZ726MIF (type A)	20	36
E6 (type B)	9	40
E6/without wetting agent (type A)	16	42

materials including their modification with adhesion promoter and other polymer layers, for example, polymethylmethacrylate (PMMA). The measurement precision of the surface tension of liquids is about 10%. In the resulting data the materials we used can be divided into two groups (Table I). We think the division can be generalized for all resists and developers. A decrease of surface tension of the developer (type B) could be better managed by adding special wetting agents (Table III, for instance, developer E6). Surface tension of the resist layers depends on softbake parameters and on material composition (resins, photoactive compounds, solutions and wetting agents). Spincoating was used to achieve a resist thickness of 1.5 μm and the softbake parameters were kept stable for all resists (hotplate: 60 s, 90 $^\circ\text{C}$).

Many resists and developers can be divided in this sense. The measurements of Fadda *et al.*¹⁷ show the obvious division of materials into two groups as in Table I also.

B. Surface tension of materials in semiconductor technology

Modified and pure substrate materials were used: (1) Si (2) SiO₂(TEOS)/phosphor doped SiO₂ (PSG)/Si₃N₄ layers, and (3) AlCu0.5(0.5%Cu) and WTi10(10%Ti) layers. An overview of the measured contact angles and the calculated surface tensions according to (2) is given in Tables IV–VI and Fig. 5. The measurement precision of the surface tension

TABLE II. Surface tension of photoresist layers (hotplate 90 $^\circ\text{C}$, 60 s).

Resist type	Resist	Contact angle		Surface tension	
		H ₂ O	CH ₂ I ₂	σ_{gs}^p (dyn/cm)	σ_{gs}^d (dyn/cm)
A	SPR511 ^a	60 $^\circ$	38 $^\circ$	21	30
	SPR511 ^b	63 $^\circ$	36 $^\circ$	19	30
B	AZ6612 ^a	78 $^\circ$	39 $^\circ$	11	32
	AZ6612 ^b	78 $^\circ$	43 $^\circ$	11	30
	AZ711B ^a	73 $^\circ$	32 $^\circ$	12	33
	AZ711B ^b	77 $^\circ$	31 $^\circ$	13	33
	AZ7510 ^a	81 $^\circ$	43 $^\circ$	11	29
	AZ7510 ^b	80 $^\circ$	38 $^\circ$	10	32
	ARP374 ^a	79 $^\circ$	43 $^\circ$	11	30
	ARP374 ^b	75 $^\circ$	48 $^\circ$	13	29

^aExposed.

^bNonexposed.

TABLE IV. Examples for priming with HMDS/5% TMDS.

Material	Contact angle		Surface tension	
	H ₂ O	CH ₂ I ₂	σ_{gs}^p (dyn/cm)	σ_{gs}^d (dyn/cm)
Si	2°	34°	45	29
Si HMDS ^a	65°	52.5°	18	24
SiO ₂	2°	33°	45	29
SiO ₂ HMDS ^a	65°	59°	22	21
Si ₃ N ₄	3.2°	33°	45	29
Si ₃ N ₄ /HMDS ^a	56°	39°	23	29

^aPriming procedure: hotplate 20 s, 120 °C.

calculated by the measured contact angle is estimated to be about 5%. The measured values for Al (Fig. 5) show good correlation with those of Kaelble and Dynes.¹⁹ They determined the surface tension of Al with $\sigma^p=38$ dyn/cm/ $\sigma^d=28$ dyn/cm. Also, the decay of surface tension with increased time of exposure to humidity is documented in this article. Further values of surface tension of Al were published by Moreau¹ ($\sigma^p=38$ dyn/cm, $\sigma^d=25$ dyn/cm), Zissmann²⁰ (critical surface tension $\sigma_c=45$ dyn/cm) and Eley²¹ ($\sigma=93$ dyn/cm).

Comparative values were found in publications concerning Si by Fowkes⁵ ($\sigma=78$ dyn/cm) and Moreau¹ ($\sigma_c=78$ dyn/cm), concerning SiO₂ by Moreau¹ ($\sigma^p=35$ dyn/cm, $\sigma^d=15$ dyn/cm) and concerning Si₃N₄ by Yanazawa⁴ ($\sigma=46.5$ dyn/cm).

Modification of surface properties were done with primer (HMDS/5% TMSDEA) in a gaseous atmosphere at a hotplate temperature of $T_p=120$ °C to reduce surface tension of high hydrophile materials. This was done with Si, SiO₂, and Si₃N₄ successfully, but one problem was a sufficient reduction of the surface tension of metals. For this it is necessary to use a different primer or to change the priming procedure. The differences between Si, SiO₂, and Si₃N₄ are small, for this reason, the investigations for the optimization of adhesion were carried out with Si as a representative material.

The pretreatment of the substrates (cleaning, dehydration and exposure to humidity) will greatly influence their surface tension (see Table V and VI). Contamination with water (dependent on relative ambient humidity) and organic materials on high-energy surfaces will occur during the time of expo-

TABLE V. Contact angle and surface tension of Si for different treatments.

Pretreatment	Contact angle		Surface tension		
	H ₂ O	CH ₂ I ₂	σ^p (dyn/cm)	σ^d (dyn/cm)	σ (dyn/cm)
No pretreatment					
delivery quality	31°	41°	38	26,5	64,5
SC 1 cleaning	2°	32°	45	30	75
SC 1,2 cleaning	2°	33°	45	29	74
SC 1,2 cleaning exposure time=5 days	16°	38°	44	27	71
SC 1,2 cleaning	6°	35°	45	28	73
O ₂ -plasma treatment					

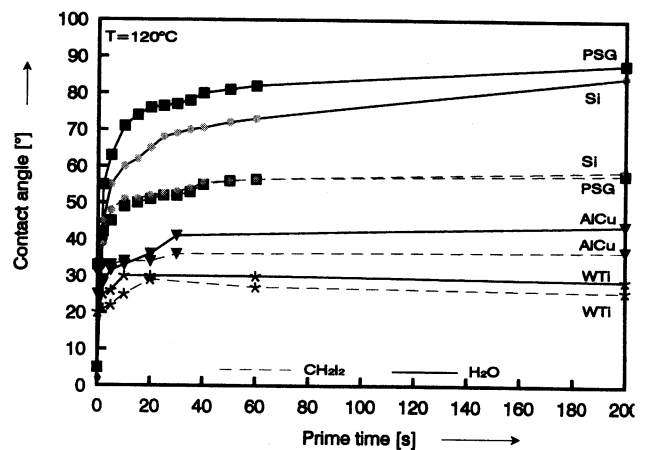


FIG. 5. Contact angle obtained with HMDS/5% TMSDEA for different priming time (hotplate 120 °C).

sure to air, which strongly reduces the surface tension of those substrates ($\pi_e \approx 316$ dyn/cm at SiO₂^{8,14,15}).

Ambient oxygen and cleaning processes with oxidizing effect lead to thin oxide layers on substrate surfaces like on Si and on Al and thus to high-energy surfaces. We used several cleaning processes: SC1 cleaning (ammoniumhydroxide hydrogenperoxide mixture), SC2 cleaning (hydrochloricacid hydrogenperoxide mixture), HNO₃ cleaning and O₂-plasma treatment for Si (Table V), and for AlCu (Table VI).

AlCu surfaces were investigated by SIMS/AES after cleaning processes were completed. It could be shown that the change of contact angle and surface tension are influenced by the thickness of the oxide layer on the surface (Table VI).

By means of these spectra, a distinct lower thickness of the oxide layer on untreated AlCu (contact angle $\Theta_w=29^\circ$, surface tension $\sigma^p=37$ dyn/cm, $\sigma^d=30$ dyn/cm) in respect to AlCu treated with strongly oxidizing medium HNO₃ (contact angled $\Theta_w=4^\circ$, surface tension $\sigma^p=42$ dyn/cm, $\sigma^d=34$ dyn/cm) could be proved. The thickness of the Al-oxide layer d_{ox} of untreated AlCu was determined on the basis of the depth of escape of the Al-KLL Auger electrons (about 2.5 nm) and the existence of elementary Al-KLL signals to be about $1 \text{ nm} < d_{ox} < 2.5 \text{ nm}$. The thickness of the oxide layer of AlCu treated with HNO₃ is about 5–8 times thicker.

TABLE VI. Contact angle and surface tension of Al/Cu for different treatments.

Pretreatment	Contact angle		Surface tension		
	H ₂ O	CH ₂ I ₂	σ^p (dyn/cm)	σ^d (dyn/cm)	σ (dyn/cm)
No pretreatment					
exposure time					
<10 min	29°	31°	37	30	67
1 day	32°	40°	31	30	61
5 days	73°	51°	15	25	40
HNO ₃ cleaning	4°	20°	42	34	76
DI-water					
HNO ₃ cleaning	8°	24°	43	32	75
DI-water					
temp. 200 °C, 30 min					
HNO ₃ cleaning	28°	30°	37	30	68
DI-water					
exposure time=5 days					
HNO ₃ -cleaning	2.5°	34°	45	29	74
DI-water					
O ₂ -plasma treatment					

In accordance to the literature cited, it can be generalized that all high-energy surfaces (glass, quartz, metals, etc.) show, in practice, almost identical surface tension after short time of exposure to ambient humidity (i.e., some hours).^{14,20} The polar part of those high-energy surfaces is about $\sigma^p \approx 40$ dyn/cm and the nonpolar component is about $\sigma^d \approx 30$ dyn/cm. But after increased time of exposure to ambient humidity, the surface tension is more and more reduced depending on cleaning processes used and ambient conditions.

The aim of special surface modification is to provide stable surface conditions for reproducible and nearly defect free process steps. To determine the surface tension the contact angle of water is well suited as shown in the following section.

C. Adhesion of resist in semiconductor technology

In Figs. 6 and 10, the measured values of the contact angle of water on modified surfaces of AlCu and Si are traced. The work of adhesion W_{a12} obviously reduces by increasing the process time of the HMDS coating. The real effect of improved adhesion—that depends on HMDS process time—can be estimated by means of the behavior of the resist layer during developing and rinsing with water [Figs. 6(b) and 6(c)].

The work of adhesion of resist/developer-combination RB/DA at $\Theta_w < 47^\circ$ is negative and spontaneous separation occurs. In that experiment, a lifting of large resist areas was observed. For RB/DA combinations such surface conditions must be brought about by primer treatment to achieve water contact angles of $\Theta_w > 47^\circ$.

Even slight HMDS treatment is sufficient for uncritical water rinsing after development [Fig. 6(b)]. That is caused by the high difference of polarity between water ($x_p = 2.3$) and resist ($x_p = 0.25, \dots, 0.5$).

For experimental verification of adhesion of small resist structures the defect density of a test structure consisting of $1 \mu\text{m} \times 1 \mu\text{m}$ and $0.8 \mu\text{m} \times 0.8 \mu\text{m}$ dies was monitored (Figs. 7 and 8). Exposure of these structures was carried out by GCA 8000 *i*-line stepper (NA=0.35, coherent parameter $S=0.5$). Evaluation was done in the middle of the image area at an exposure dose of $1.8 H_0$ (H_0 is the amount of exposure necessary to promote the total development of large areas of resist during the developing process). In our case, dip developing was used. Evaluation of the defect density was done by the light/dark field optical technique. Figure 9 shows examples of defect densities using SPR511 and AZ6612 resists. It can be generally concluded that for defect free structures in the submicrometer range work of adhesion of $W_{12} > 5$ dyn/cm is necessary (for developer and water treatment).

In Fig. 6, it shows the critical process of adhesion is most affected in the development process. For the resist/developer combinations RB/DA, RA/DA, and RB/DB, a surface modification for adjustment of a contact angle of water of $\Theta_w \geq 65^\circ$ assures good adhesion quality of the resist structures even during development process step.

For combination of RA/DB, there were not observed any defects even at smaller contact angles but, in this case, the contact angle of water should not exceed 85° according to Fig. 6(c).

Generally, in cases combining resist with high polarity ($\sigma^p > 15$ dyn/cm) with developer of low polarity ($\sigma^p < 15$ dyn/cm), only a short primer process should be applied; whereas, the usage of resist with low polarity ($\sigma^p < 15$ dyn/cm) requires a longer primer process to adjust a contact angle of water of more than 65° .

These results are in accordance to the publication of Deckert and Peters²² where they determined the best adhesion is obtained if substrate and resist have low values of σ^p .

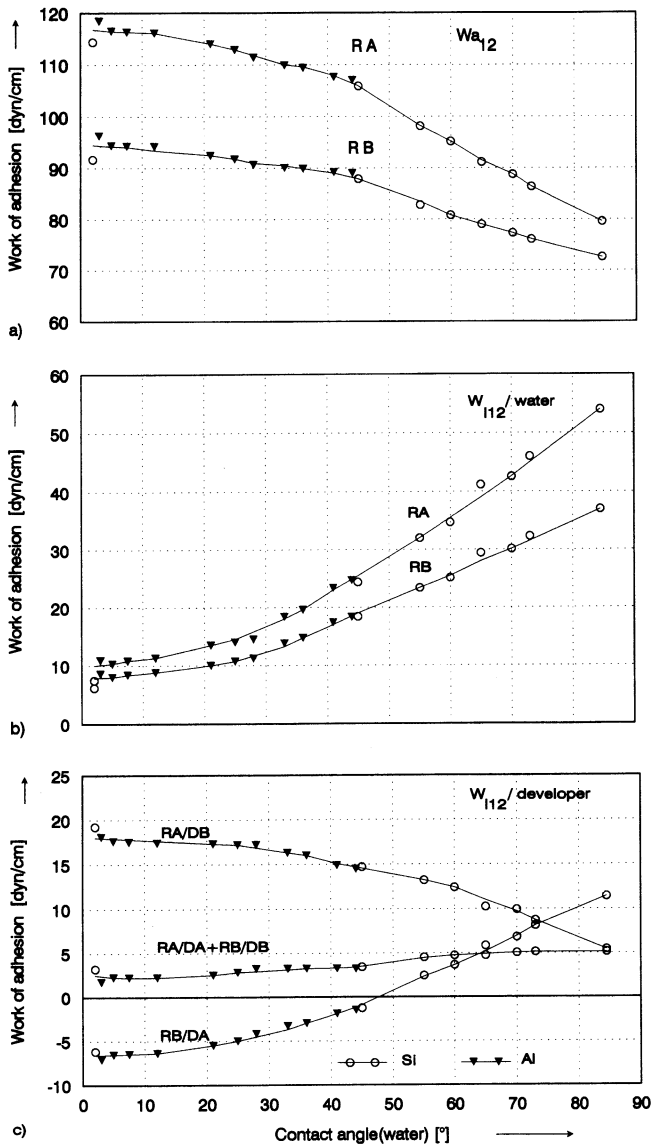


FIG. 6. Work of adhesion for resist as function of contact angle of modified Si and AlCu substrates (a) under air, (b) under water, and (c) under developer environment.

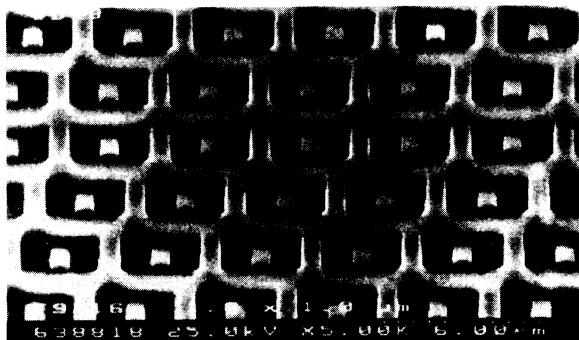
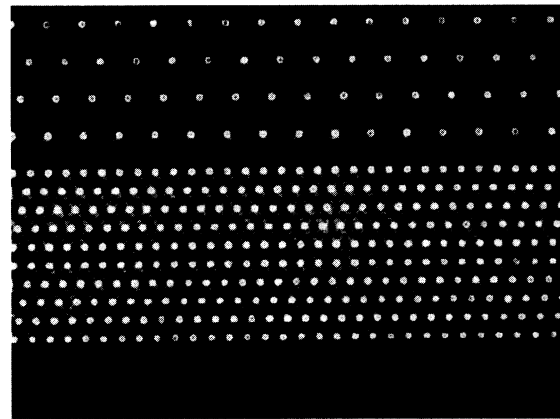
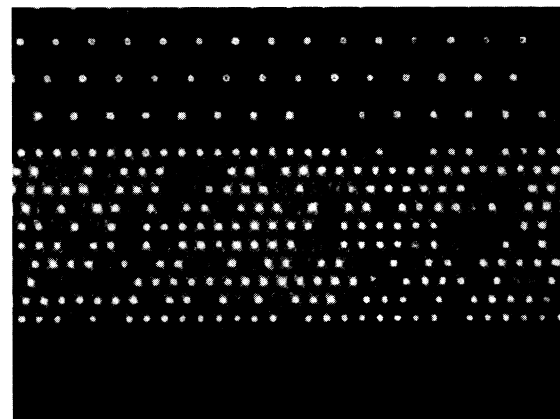


FIG. 7. Test structure for investigation of adhesion (SPR511 on AlCu, $\theta_w = 25^\circ$).



(a)



(b)

FIG. 8. Examples for measurement of defects of photo resist dies ($0.8 \mu\text{m} \times 0.8 \text{mm}$) on AlCu with dark-field image: (a) photoresist/developer: SPR511/MFCD, substrate $\theta_w = 25^\circ$, defect density=0%, (b) photoresist/developer: SPR511/MFCD, substrate $\theta_w = 5^\circ$, defect density=30%.

In contrast to Deckert and Peters²² recommending to combine substrates with high σ^p only with resist of low values of σ^p , we found, applying developer with low σ^p , substrates and resists with high σ^p could be combined successfully (combination RA/DB, in Figs. 6 and 10).

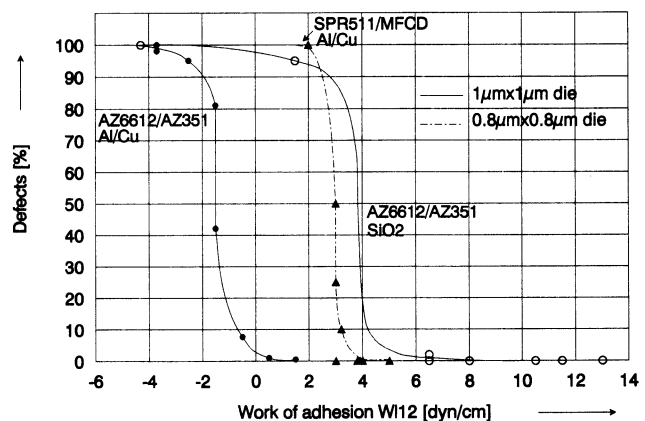


FIG. 9. Density of die defects depending on work of adhesion of resist on AlCu and on SiO₂ in developer solution (surface primer: HMDS/5% TMS-DEA).

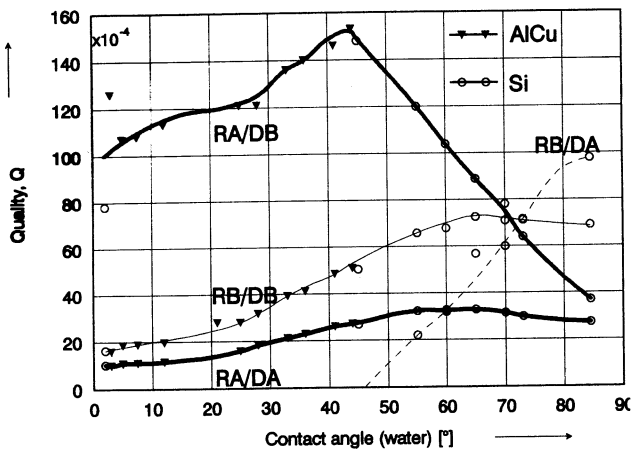


FIG. 10. Quality function [Eq. (11)] vs contact angle of water.

To apply resist, with low polarity at metal surfaces, it is necessary to use other primers (e.g., TMSDEA concentrated) for coating in a gaseous atmosphere or to coat those surfaces by spinning solutions of primer to adjust a contact angle of water of $\Theta_w > 50^\circ$.

D. Wetting characteristics of resist and development

In Table VII, the measured contact angles and wetting factors for developer at Si, AlCu, and resist layers as well as for liquid resist on Si and AlCu are shown. To guarantee good wetting for any liquid a contact angle of that liquid (for example of developer and liquid resist) should be $\Theta < 62^\circ$.¹⁸ That corresponds to a wetting factor of $f_w > 0.30$.

Thus, problems concerning wetting should be only expected to occur using developer of type A (high surface ten-

TABLE VIII. Limits for contact angle of water on materials of semiconductor technology modified with primer.

Resist/developer	(Example)	Contact angle (water)		
		Θ_{min}	Θ_{max}	$\Theta_{optimal}$
RA/DB	(SPR511/MF501)	10°	75°	45°
RA/DA	(SPR511/MFCD)	60°	85°	70°
RB/DB	(ARP374/E6)	60°	85°	70°
RB/DA	(AZ6612/AZ351)	65°	85°	80°

sion $\sigma > 60$ dyn/cm) with substrates of very low surface tension ($\sigma < 35$ dyn/cm), e.g., after a long primer process (Table VII).

E. Optimizing adhesion and wetting

For evaluation of all processed components the following quality function was defined:

$$Q = f_{ad} f_{aw} f_{wD1} f_{wD2} f_{wR}, \tag{11}$$

where f_{ad} is the adhesion factor of interface resist/substrate in developer, f_{aw} the adhesion factor of interface resist/substrate in water, f_{wD1} the wetting factor of developer on resist, f_{wD2} the wetting factor of developer on substrate, and f_{wR} the wetting factor of resist on substrate.

The results of this evaluation are traced in Fig. 10 (only positive values are depicted). On the basis of these results, the following limits of the contact angle of water on the substrates can be fixed and used for control purposes (Table VIII).

TABLE VII. Wetting parameters of developers (type A, B) and liquid photoresist on Si, AlCu and resist layers.

Substrate	H ₂ O Θ_w	Developer type A			Developer type B			Photoresist (liquid)		
		Θ_D	λ_{32}	f_{WD}	Θ_D	λ_{32}	f_{WD}	Θ_R	λ_{R2}	f_{WR}
Si (HMDS)	2°	0°	2.6	1.07	3.6°	-0.1	1.0	19°	-2	0.95
	45°	26°	-6.1	0.82	23°	-4.1	0.87	24°	-3	0.89
	55°	41°	-15	0.59	37°	-10	0.68	37°	-7	0.76
	60°	46°	-18	0.51	41°	-12	0.62	40°	-8	0.72
	65°	51°	-22	0.43	44°	-14	0.56	42°	-8.5	0.69
	70°	54°	-25	0.38	48°	-16	0.50	43°	-9.5	0.65
	73°	57°	-27	0.33	50°	-18	0.47	44°	-10	0.62
	84.5°	64°	-43	0.24	55°	-21	0.38	46°	-11	0.58
AlCu (HMDS)	3°	0°	8.1	1.25	0°	6	1.18	0°	3.3	1.09
	7.5°	0°	5.5	1.16	0°	3.5	1.10	0°	1.3	1.04
	12°	0°	5.3	1.16	0°	3.4	1.10	0°	1.1	1.03
	25°	0°	1.5	1.04	0°	0.4	1.01	13°	-0.9	0.97
	33°	14°	-1.9	0.94	15°	-1.7	0.95	20°	-2.1	0.94
	41°	22°	-4.2	0.87	19°	-2.6	0.92	20°	-2.1	0.93
	44°	23°	-4.8	0.86	19°	-2.8	0.91	20°	-2.1	0.93
	Resist type A	61°	36°	-11.4	0.66	25°	-4.7	0.83
Resist type B	81°	54°	-24.7	0.38	40°	-11.4	0.60

IV. SUMMARY

In accordance with experimental results (Figs. 8 and 9), it is possible to adjust optimal conditions for adhesion of resist on different materials by means of measurement of contact angle of water if surface tension of the applied resist and developer is known. It could be shown that a combination of resist with high σ^p and developer with low σ^p is a good prerequisite for adhesion and wetting. Whereas, using resist with high σ^p combined with developer with high σ^p , good adhesion can only be guaranteed if surface tension of the substrate is strongly reduced. We think that similar results can be achieved in etching processes combining resists with high σ^p with etchants with high σ^p , respectively. According to Fig. 9 for resist layers in liquids (developer, water, etchants), the work of adhesion should be >5 dyn/cm.

ACKNOWLEDGMENTS

The authors gratefully acknowledge Dieter Krüger and Ralf Kurps, Institute of Semiconductor Physics, Frankfurt (Oder), for the SIMS/AES investigations to analyze deposits before and after HMDS treatment. We also would like to thank Janett Guhr and John Dirks for their helpful discussions.

- ¹W. M. Moreau, *Semiconductor Lithography* (Plenum, New York, 1988).
- ²R. Dammel, *Diazonaphthoquinone-based Resist* (SPIE Optical Engineering, Bellingham, WA, 1993), Vol. TT 11.
- ³Michielsen, V. B. Marriott, J. J. Ponjee, H. van der Wel, F. J. Touwslager, and J. A. H. M. Moonen, *Microelectron.* **11**, 475 (1990).
- ⁴H. Yanazawa, *Colloids Surf.* **9**, 133 (1984).
- ⁵F. W. Fowkes, in *Chemistry and Physics of Interfaces* (American Chemical Society, Washington, DC, 1971), pp. 153-167.
- ⁶F. W. Fowkes, *Ind. Eng. Chem.* **56**, 12-40 (1964).
- ⁷F. W. Fowkes, in *Wetting* (Society of Chemical Industry, London, 1967).
- ⁸F. W. Fowkes, *J. Phys. Chem.* **66**, 382 (1962); **67**, 2538 (1963).
- ⁹J. L. Gardon, *J. Paint Technology* **38**, 43 (1966).
- ¹⁰C. M. Hansen and A. Beerbower, in *Kirk-Othmer's Encyclopedia of Chemical Technology*, Suppl. 2nd ed. (Wiley, New York, 1971), pp. 889.
- ¹¹E. F. Meyer and R. E. Wagner, *J. Phys. Chem.* **70**, 3162 (1966).
- ¹²E. F. Meyer, T. A. Renner, and K. S. Stec, *Phys. Chem.* **75**, 642 (1971).
- ¹³S. Wu, *Polymer Interface and Adhesion* (Marcel Dekker, New York, 1982).
- ¹⁴S. Wu, in *Recent Advances in Adhesion*, Proc. Am. Chem. Soc. Symp. (American Chemical Society, Washington, DC, 1971), pp. 45-64.
- ¹⁵N. Hackmann and A. C. Hall, *J. Phys. Chem.* **62**, 12 (1958).
- ¹⁶This factor is comparable to the factor introduced by Yanazawa (Ref. 4) derived from wetting tension, $f_{wet} = 1 - f_a$.
- ¹⁷E. Fadda, G. R. Amblard, A. P. Weill, and A. Prola, *Advances in Resist Technology and Processing XI*, Vol. 2195 (SPIE, Bellingham, WA, 1994), pp. 676-683.
- ¹⁸J. F. Padday, *Wetting, Spreading and Adhesion* (Academic, New York, 1978).
- ¹⁹E. G. Shafrin and W. A. Zisman, *Am. Ceram. Soc.* **50**, 478 (1967).
- ²⁰W. A. Zisman, in *Adhesion Science and Technology*, edited by L. E. Lee (Plenum, New York, 1975), Vol. 9A, pp. 55-91.
- ²¹D. D. Eley, *Adhesion* (Oxford University Press, Oxford, 1961).
- ²²Ch. A. Deckert and D. A. Peters, *Thin Solid Films* **68**, 417 (1980).