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Contact Adhesion of Thin Gold Films on Elastomeric Supports: Cold Welding Under Ambient Conditions

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Thin gold films placed in contact on compliant elastomeric poly(dimethylsiloxane) supports weld together. This "cold welding" is remarkable both for the low loads required and for the fact that it occurs under ambient laboratory conditions, conditions in which the gold surfaces are covered with films of weakly adsorbed organic impurities. These impurities are probably displaced laterally during the welding. Welding can be prevented by the presence of a self-assembled gold(I) alkylthiolate monolayer on the gold surfaces. The welded contacts have low electrical resistivity and can be made thin enough to transmit light. This system is a promising one with which to study interaction between interfaces.

WELDING OF METALS UNDER AMBient conditions ("cold welding") has been practiced for more than 700 years, but only with high applied pressures (such as under the impact of a smith's hammer) or with frictional work (1–3). The adhesion of metals in ultrahigh vacuum (UHV) under light loads is also known (4) but requires flat, ductile, and atomically clean surfaces. In this report we

describe the self-adhesion of thin gold films on elastomeric supports, under ambient laboratory conditions, with very small applied loads (Fig. 1). Adhesive bonding of metal surfaces under ambient laboratory conditions—that is, in the presence of air, humidity, and volatile organic contaminants—and with very small applied loads (<0.1 to 0.2 g/cm²) (5) is therefore remarkable. For self-adhesion of these "dirty," supported films of gold, an underlying elastomeric support is required. The self-adhesion is inhibited or prevented by monolayer films [self-assembled monolayers (SAMs)] less than 1 nm thick on the gold.

We prepared the systems by the procedure summarized in Fig. 1. Treatment of a

film of poly(dimethylsiloxane) (PDMS) (6) with a radio frequency, oxygen plasma formed a thin [<50 Å by x-ray photoelectron spectroscopy (XPS)] silica-like layer on its surface. We denote this oxidized surface as PDMS/SiO₂; its surface chemistry is similar to that of SiO₂ (7). Chemisorption of 11-trichlorosilylundecyl thioacetate [$\text{Cl}_3\text{Si}(\text{CH}_2)_{11}\text{SCOCH}_3$] from the vapor phase onto PDMS/SiO₂ produced a monolayer of the corresponding alkylsiloxane (8, 9). Thin films of gold (~ 20 nm), thermally evaporated onto the surface of the PDMS-bound SAM (9–11), adhered well to it (9, 12, 13).

When placed in contact, two gold films supported on 1 cm by 1 cm squares of PDMS adhered strongly across the gold-gold interface. Failure occurred by decohesion within the polymer (the tear strength of the PDMS used here is 2.7×10^3 g/cm) (14). We hypothesize that the elasticity and compliance of PDMS allow the gold surfaces to conform to one another, increasing the area of gold-gold contact and tangentially displacing loosely adsorbed contaminants (15). This hypothesis implies the possible formation of "islands" of the contaminants at the gold-gold interface.

We measured the strength of adhesion by using an apparatus reported separately (8). A small (radius of curvature = 1.31 to 1.34 mm) hemispherical lens of PDMS and a flat sheet of PDMS were allowed to come into contact in the absence of an applied load. For two surfaces of unmodified PDMS, the pull-off force was 0.034 dyne for an initial area of contact at zero load of 4.45×10^{-4} cm². We characterize this adhesion as "tacky," because the area of contact decreased with increasing negative load. The pull-off force for the two gold films supported on PDMS was 3.33 dyne for an initial area of contact of 3.53×10^{-4} cm². In this case, the area of contact did not decrease with increasing negative load; rather, cohesive failure occurred within the flat sheet of PDMS. We conclude that this pull-off force is a lower limit of the strength of adhesion across the gold-gold interface and that these results rule out the possibility that the welding is actually tacky adhesion arising from organic contaminants at the interface.

Chemisorbed monolayers of alkyl thiolates at the surfaces of these gold films prevented welding. Treatment of one of the films with ethanoethiol vapor for 5 to 10 s greatly reduced the strength of adhesion; that is, the films were easily separated and adhesive failure occurred at the "gold-gold" interface. This "weak" adhesion was similar in strength and character to the tackiness between sheets of unmodified PDMS (16). As expected, gold films bearing ordered SAMs of longer chain alkyl thiolates (17)

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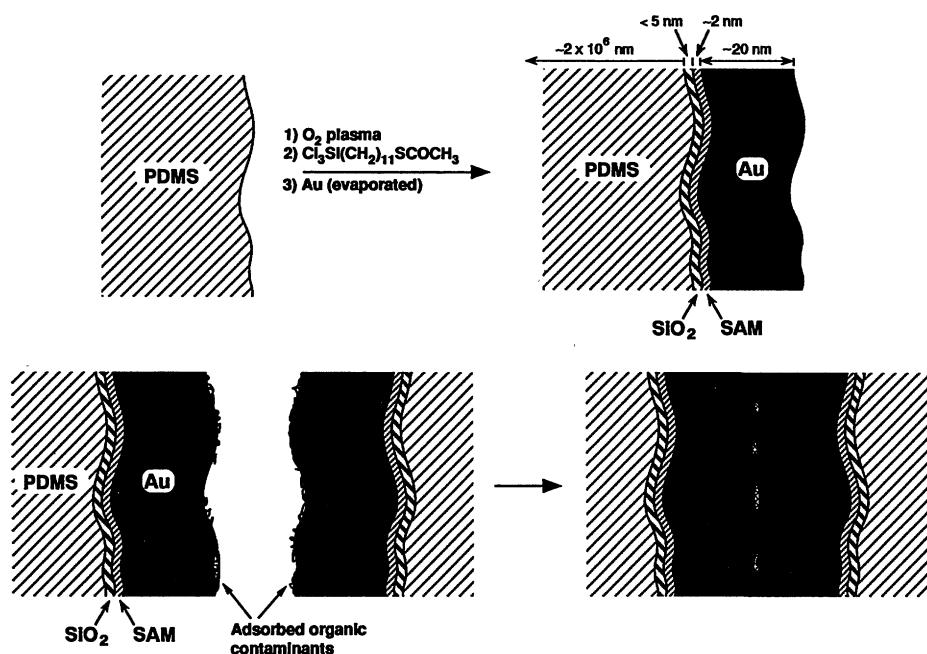


Fig. 1. Schematic illustration of the structure of the supported films of gold and gold-gold welding. PDMS = poly(dimethylsiloxane); SiO₂ = silicon dioxide overlayer; SAM = self-assembled monolayer prepared by adsorption of 11-trichlorosilylundecyl thioacetate; Au = evaporated film of gold.

[HS(CH₂)₁₀CH₃, HS(CH₂)₁₁CH₃, and HS(CH₂)₁₅CH₃] also showed only weak adhesion. The pull-off force for two of these surfaces [bearing SAMs of HS(CH₂)₁₁CH₃] was 0.037 dyne for an initial area of contact of 3.50×10^{-4} cm². We presume that these thin (5 to 25 Å) films exert their influence by preventing atomic contact of the gold surfaces.

It seems to be necessary to have at least one gold film supported on a compliant elastomer in order for cold welding to occur under these conditions. We observed welding between a sample of the gold on a PDMS square 1 cm by 1 cm and gold condensed very slowly (0.3 to 0.4 Å/s) onto a glass, microscope cover slip (18). We observed only weak (tacky) adhesion, however, between samples of gold on PDMS and gold condensed at or above 1 to 3 Å/s onto a glass microscope slide (19).

The composite films (PDMS/SiO₂/monolayer/Au/Au/monolayer/SiO₂/PDMS) described in this report were optically transparent and provide an opportunity for optical microscopic and spectroscopic (ultraviolet-visible) analysis of the gold-gold interface. Preliminary experiments, in which we used simple patterned surfaces formed by shadowing portions of the polymer film during evaporation of the gold, have established that the gold-gold contacts show little electrical resistance (<0.4 ohm/cm²) (20).

Cold welding requires atomic contact between the surfaces that are joining. These clean surfaces are probably generated in the system described here by lateral displace-

ment of contaminants on the gold surface. This displacement is facilitated by the elastomeric and compliant support. Although cold welding is a well-known phenomenon in other circumstances, its occurrence between "dirty" metal surfaces, under ambient atmospheres, at very small applied pressure is unexpected. A number of characteristics of this system make it a particularly attractive one with which to study cold welding. The mechanical properties of the PDMS elastomer are easily varied. The thicknesses of the SiO₂ layer, the gold film, and the SAM coupling layer can be controlled. The system is very sensitive to alkane thiolates adsorbed on the gold, and the techniques developed in studying these SAMs (17) are applicable to understanding this sensitivity. The entire system is optically transparent and can be examined by absorption spectroscopy. The welds are electrically conducting: this conductivity may be useful in characterizing them and may also provide the basis for methods of fabricating novel types of electrical circuits.

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10. Gold was condensed onto the substrates at rates of 0.3 to 3 Å/s at $20 \pm 2^\circ\text{C}$.
11. Scanning electron micrographs of PDMS/SiO₂ bearing monolayers and 200 Å of thermally evaporated gold were featureless on a length scale greater than 1 μm. A gridlike distribution of fine cracks (<0.1 μm wide) separated by 20 to 40 μm was observed in the gold film. XPS analysis indicated the presence of silicone contaminants on the gold surface.
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16. The strength of adhesion between two sheets of unmodified PDMS is less than 1% of that between two of the gold surfaces (without monolayers). Pull-off forces were measured by the procedure described in (8).
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20. We made these measurements using a four-probe method to avoid errors due to the resistance of the

electrical contacts (silver epoxy) to the gold films. In addition to the possibility of practical applications of these experiments, careful measurement of resistivity may allow an estimation of the degree of atomic (gold-gold) contact between the two surfaces.

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Thymocyte Expression of RAG-1 and RAG-2: Termination by T Cell Receptor Cross-Linking

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The expression of the V(D)J [variable (diversity) joining elements] recombination activating genes, RAG-1 and RAG-2, has been examined during T cell development in the thymus. In situ hybridization to intact thymus and RNA blot analysis of isolated thymic subpopulations separated on the basis of T cell receptor (TCR) expression demonstrated that both TCR⁻ and TCR⁺ cortical thymocytes express RAG-1 and RAG-2 messenger RNA's. Within the TCR⁺ population, RAG expression was observed in immature CD4⁺CD8⁺ (double positive) cells, but not in the more mature CD4⁺CD8⁻ or CD4⁻CD8⁺ (single positive) subpopulations. Thus, although cortical thymocytes that bear TCR on their surface continue to express RAG-1 and RAG-2, it appears that the expression of both genes is normally terminated during subsequent thymic maturation. Since thymocyte maturation in vivo is thought to be regulated through the interaction of the TCR complex with self major histocompatibility complex (MHC) antigens, these data suggest that signals transduced by the TCR complex might result in the termination of RAG expression. Consistent with this hypothesis, thymocyte TCR cross-linking in vitro led to rapid termination of RAG-1 and RAG-2 expression, whereas cross-linking of other T cell surface antigens such as CD4, CD8, or HLA class I had no effect.

DURING DEVELOPMENT, THE THYMIC rudiment is seeded by bone marrow-derived stem cells that have not yet undergone TCR gene rearrangement (1). As thymic cells mature, each cell assembles the genes that encode a TCR α - β or γ - δ heterodimer through the recombination of individual variable (V), diversity (D), and joining (J) germline elements for each chain (2). This assembly process is known as V(D)J recombination. In general, mature T cells express only a single TCR (3). It is the specificity of the TCR that appears to determine the fate of the thymocyte as it undergoes thymic selection (4). Recently, two genes that appear to encode components of the V(D)J recombinase, RAG-1 and RAG-2, have been cloned (5). Coexpression of RAG-1 and RAG-2 is both necessary and sufficient to induce V(D)J recombination in fibroblasts, and their

expression in lymphoid cell lines correlates precisely with the presence of V(D)J recombinase activity (5, 6). These findings suggested that the regulation of RAG-1 and RAG-2 expression might contribute to the control of TCR recombination during lymphoid ontogeny. To explore this possibility, we have examined RAG expression in lymphoid cells developing in the thymus.

The intrathymic expression of RAG-1 and RAG-2 mRNA transcripts was analyzed by in situ hybridization (Fig. 1). Sections of thymus from a 32-day-old mouse were hybridized with RAG-1 antisense (Fig. 1B) or RAG-2 antisense (Fig. 1C) RNA probes. Control probes included RAG-1 sense and RAG-2 sense (Fig. 1D) RNA probes. Both the RAG-1 and RAG-2 transcripts were detected throughout the cortex although the RAG-1 hybridization was consistently more intense. The stronger signal for RAG-1 compared to RAG-2 seen by in situ analysis was also seen in RNA (Northern) blots hybridized with cDNA probes of roughly comparable size labeled to equal specific activities (see below). A demarcation of RAG-1 and RAG-2 expression was observed between the thymic cortex and medulla (Fig. 1, E and F). Little or no specific

antisense hybridization was seen with RAG-1 or RAG-2 in the medulla. The extensive specific hybridization for both RAG-1 and RAG-2 in the cortex was surprising in that a large percentage of cortical thymocytes express TCR on their surface (2), an indication that they have already carried out productive V(D)J recombination. This suggested that productive TCR gene rearrangement and surface TCR expression was by itself not sufficient to terminate RAG expression.

To confirm that the expression of RAG-1 and RAG-2 by TCR⁺ cortical thymocytes was a general feature of mammalian T cell development, we investigated human thymocyte subpopulations (Fig. 2A). After productive TCR gene rearrangement, the resulting TCR heterodimer is expressed on the cell surface in close association with several other invariant polypeptides, and forms a structure referred to as the TCR-CD3 complex (7). Like that of the mouse, the human thymic cortex contains both TCR-CD3⁻ cells, which have not completed productive TCR gene rearrangement, and immature TCR-CD3⁺ cells that have undergone successful V(D)J recombination (1). As expected, RAG-1 and RAG-2 were co-expressed at high levels in unseparated human thymocytes (Fig. 2B). Cell fractionation revealed that both RAG-1 and RAG-2 transcripts were expressed in TCR-CD3⁻ cells, as would be expected if these cells are undergoing V(D)J recombination. In addition, as was predicted from the in situ hybridization analyses, RAG-1 and RAG-2 were also expressed in TCR-CD3⁺ cells (Fig. 2C).

Immature cortical TCR-CD3⁺ thymocytes express both the CD4 and CD8 accessory molecules. Thus, they are frequently referred to as "double positive" cells. About 90% of TCR-CD3⁺ cells in human thymic tissue are double positive cells, but the majority of double positive thymocytes die in the thymus before further maturation (8). The continued maturation of a small number of double positive cells is apparently dependent on the ability of their TCR-CD3 complex to interact with a self-MHC class I or II molecule expressed on thymic epithelial cells (Fig. 2A) (4). Such an interaction, although poorly understood, results in cell survival and "positive selection" and ensures that the T cell response to foreign antigen will be restricted by self-MHC molecules. If positive selection occurs as a result of TCR interaction with a class I MHC molecule, thymocytes differentiate to "single positive" cells that express only the class I-restricting antigen CD8 and lose CD4 expression. In contrast, if selection occurs through interaction with an MHC class II antigen, the

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