# Material Issues in Layered Forming

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## Abstract

A brief overview of key issues in layered thermal processing is given. Incremental sintering and layered fusion of powder and molten droplets are discussed. The criteria for remelting the solid substrate are derived from a one dimensional heat transfer model. Temperature gradients which occur during solidification and subsequent cooling are responsible for the build up of internal stresses which can be estimated through establishing an elastic beam model. The difficulties as well as opportunities regarding the generation of multi-layer multi-material structures are also described in this article.

## Key Words

Layered manufacturing, sintering, melting, thermal modeling, residual stress, multi material structures, stress cracking.

## Introduction

Solid freeform fabrication through layered material deposition appears to be an attractive method for 3D object generation[1,2,3]. This method offers the possibility of expanding the design space with respect to geometric complexity, material diversity, and traditional cost/time constraints. However, building up materials in layeres poses significant challenges from material science, heat transfer and applied mechanics viewpoint.

Depositing materials onto a solid substrate can typically be accomplished through sintering, local melting, chemical synthesis (e.g. photo polymerization), or otherwise gluing, brazing, and soldering. The issues associated with each of these processes can be summarized as follows:

- Local melting requires significant energy input to the semi-finished part which may result in the buildup of internal stresses and consequently distortions.
- Sintering requires less energy to establish bonding of the added layers but local voids may be left unless external forces are applied
- The practical applicability of chemical synthesis is limited to certain derivatives of organic substances
- Gluing, brazing, and soldering have the disadvantage of adding bonding materials to the part which are not necessarily desirable for its function or performance.

Some of these difficulties can be overcome by adopting post processing steps such as annealing, sintering, and material infusion. Building parts through layered forming is further complicated if one attempts to deposit dissimilar materials on to the substrate. In particular, differences in the coefficient of thermal expansion (CTE), and misfit dislocations (due to differences in atomic radii) can lead to even greater distortions of the atomic lattice in comparison with layered material structures of the same kind. This paper discusses some of these underlying issues in layered forming rather than attempting to offer specific solutions to these problems.

## Process Classification

Common to all layered forming techniques is the incremental nature of the material build up process. Stepwise material build up requires bonding between layers. Obviously, the material quality of a part is determined by the quality of each deposited layer as well as the quality of the bond between the layers. The following classification for material deposition processing in layered manufacturing is chosen. Processes are listed with respect to the temperature regimes in which they operate at and issues of concern regarding the resulting articles. This list is by no means exhaustive with important problems like speed, surface quality and accuracy not being addressed.

| **Process** | **Temperature** | **Issues** |
|---|---|---|
| Sintering | $T<T_M$ | Density<br>Postprocessing |
| Melt On | $T>T_M$ | Residual Stress<br>Warpage<br>Debonding<br>Postprocessing |
| Glueing<br>Powder<br>Sheet | $T \sim T_R$ | Strength<br>Postprocessing |
| Photocuring | $T \sim T_R$ | Limited Material Range<br>Residual Stress |

In the following, we limit our discussion mostly to thermal processing issues (e.g.sintering, melting) some of which are also relevant for processes occurring at room temperature.

### Sintering

Layered powder deposition followed by laser sintering has become an established prototyping process; for more details see [1]. The physics of any sintering process is based on particle fusion at temperatures below the material melting point. During sintering necks form between adjacent powder particles thus reducing the surface area and increasing the density of the powder aggregate. The driving force for this process is the reduction of the particle surface free energy. The densification rate is proportional to that reduction.

In order to change the shape of the powder particles, matter or vacancies need to flow. (Vacancy flow can be considered as the counterflow of matter, both concepts are equivalent). The densification rate depends further on the combination of the transport path of the matter as well as the source of the matter. Ashby [4] distinguishes six different path/source combinations e.g.: surface diffusion from surface, boundary diffusion from boundary, or volume diffusion from boundary. At different temperatures different path/source combinations dominate the flow of matter.

During pressureless sintering (i.e., no external force applied) the densification rate decreases as the aggregate density increases due to a decreasing rate of surface reduction. A quick inspection of the theoretically established sintering maps by Ashby such as the example of copper in Figure 1 indicate that close to full density (i.e. when the neck radius is comparable to the particle radius) can only be reached asymptotically. Also, the times required to achieve high densities are significantly higher than the mean time that a selective heat source (e.g. laser) will for practical reasons dwell in a certain location. Hence, selectively sintered powder aggregates need to be subjected to further postprocessing procedures such as hot isostatic pressing to achieve full density.

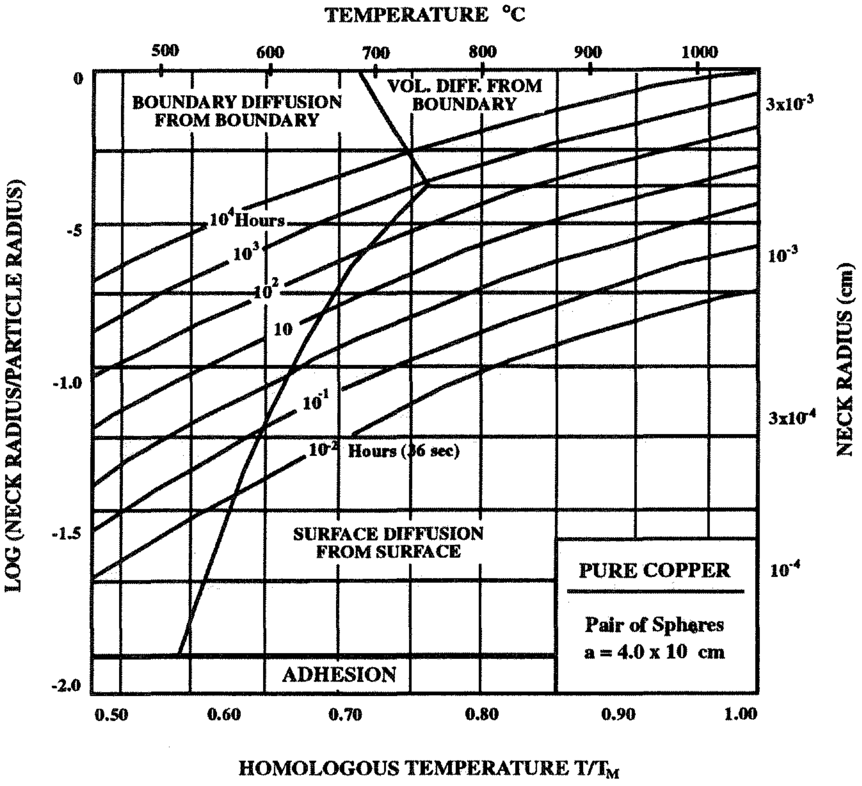


Figure 1: Sintering map of copper particles [4].

## Melting On

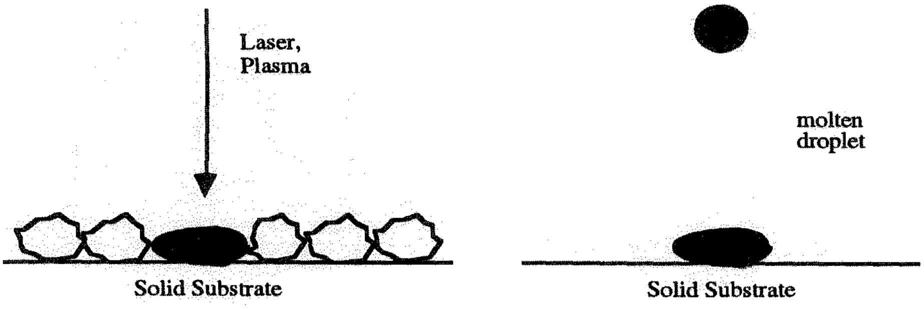
To further enhance and accelerate the bonding of a layer to the substrate one can locally melt already deposited powder particles or deposit molten droplets as done for example in thermal spraying [2]. Two scenarios can be envisioned: In the first one the molten droplet adapts to the shape of the underlying substrate. In the second the particle has sufficient energy to remelt the substrate and form a solid bond. In the first case two possibilities for bonding exist. The molten droplets simply form a mechanical interlock as commonly observed in thermal spraying [4]. Alternatively, the droplets may also bond to the substrate through a sinter mechanism in which necking occurs by shape adaptation of the molten droplet and diffusion within or on the surface of the substrate. The rate of bond formation will obviously be higher compared to the pure sinter case as described earlier.

In addition to the structure of the bond, the microstructure resulting from the solidification of the droplets is key to the strength of the layered article. Therefore an understanding of the entire temperature history is important for planing layered manufacturing processes. Also, higher temperature gradients involved in melting compared to pure sintering tends to lead to the formation of higher residual stresses.

In the following sections we address the issue of predicting the thermal history of the melting process as well as the build up of residual stresses after solidification.

## Thermal Modeling

This section presents a numerical modeling of the thermal history of a molten metal particle on a solidified substrate. The particle can be melted by a laser or plasma alternatively, molten droplets can be sprayed on a solidified substrate.as depicted in Figures 2a and 2b. This model is useful for investigating the conditions needed to achieve partial substrate remelting, to create an accurate predictive tool of the particle melt of the thermal spray process, and to investigate the effect of operating conditions such as initial molten particle/droplet and substrate temperatures, size, surface heat transfer and sprayed material properties on the resulting melting front migration rate and thickness, temperature distribution, and overall cooling rates.



(a) (b)

Figure 2 (a) molten powder particle (b) molten droplet deposition.

In the following we refer to molten particles as well as droplets from spraying just as droplets. Application parameters such as laser energy or spray gun power input, and deposition rates may then be modified to optimize the deposited material microstructure.

The numerical model for determining process temperatures and remelting conditions is simplified to a one-dimensional, heat transfer problem by assuming that the impacted droplet width is sufficiently greater than height, and that the droplet flattening time scale is much shorter than the droplet solidification time scale.This phenomena is modeled by the governing equation of the form:

$$\rho\ c_p \frac{\partial T}{\partial t} = k(T) \left[\frac{\partial^2 T}{\partial x^2}\right] + \frac{\partial k(T)}{\partial T} \left[\frac{\partial T}{\partial x}\right]^2 \tag{1}$$

for temperature T, density r, specific heat $c_p$, and thermal conductivity k. The $\partial k/\partial T$ term is omitted since the thermal conductivity variation is slight for the materials and temperature ranges considered, although temperature dependent thermal properties are used [6]. This equation is valid for both the liquid region as well as the solid region. Above the top liquid surface, combined convective and radiative boundary conditions exist, while the energy balance:

$$\rho\ L \frac{\partial x}{\partial t} = k_{sol} \frac{\partial T}{\partial t} - k_{liq} \frac{\partial T}{\partial t} \tag{2}$$

is applied at the interface between the liquid and solid regions, balancing the energy flux into and out of the interface with the release of latent heat (L) For the lower boundary of the solid region a constant substrate temperature is assumed at a remote distance from the surface.

The energy equation is discretized using an Eulerian explicit formulation. To track the location of the melting front during the solidification process, a three-point Lagrange interpolation formula is used to approximate the temperature function [7] at the nodes preceding and following the melting point. This assumes a form that can be readily incorporated into the finite difference formulation used, but permits the location of a varying "node" point corresponding to the melting front. The new front location is calculated after each iteration using the discretized interface energy balance equation. To approximate the initial interface temperature when the known liquid droplet first strikes the known solid substrate the analytical Stefan interface solution is used:

$$T_{inter.} = [RATIO * T_{liq} + T_{sol}] / [1 + RATIO] \tag{3}$$

$$\text{RATIO} = \sqrt{(k\, c_p\, \rho)_{liq} / (k\, c_p\, \rho)_{sol}} \tag{4}$$

For the complete duration of the thermal system modeling, the Stefan solution is not an accurate representation of the actual boundary conditions. However, for the initial interface condition the solution above can be used because boundary conditions corresponding to two semi-infinite bodies in contact remain valid until the temperature fluctuation propagates to the liquid surface.

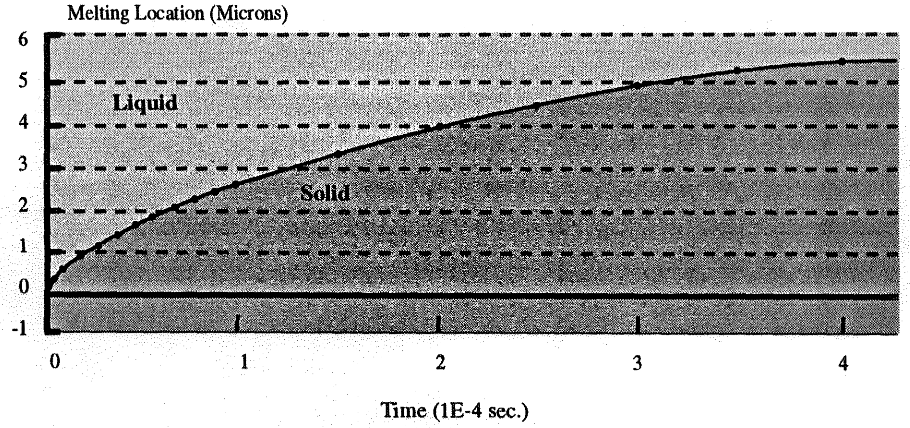
For the first droplet, the initial conditions assumed are uniform droplet and ambient substrate temperatures. When prior molten droplets heat the substrate, the model is then modified to incorporate the two-dimensional effects of substrate preheating arising from the diffusion of energy from previously molten droplets:

$$\rho\, c_p \frac{\partial T}{\partial t} = k(T) \left[ \frac{\partial^2 T}{\partial x^2} + \frac{\partial^2 T}{\partial y^2} \right] \tag{5}$$

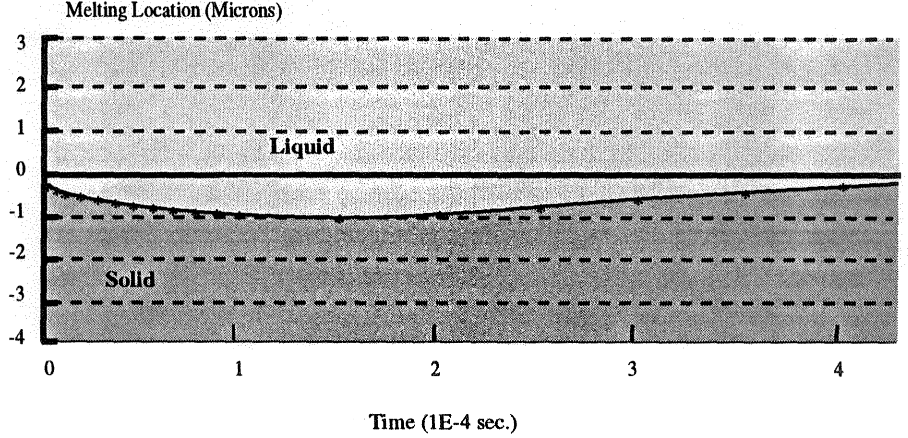
This model reflects the process where the laser source or spray gun is moving across the substrate. As with the one-dimensional solidification model, an Eulerian explicit algorithm is used to solve the two-dimensional energy equation. Because remelting does not occur with this lateral case there is no Lagrangian approximation terms required. The substrate temperature profile resulting from this two-dimensional model is then used as the substrate initial condition for the solidification model.

Simulations are made for the model of single droplets of carbon steel, stainless steel and zinc which are residing or have landed on similar substrates, and for a steel droplet on a zinc substrate. This latter case simulates the building up of sprayed materials onto a sacrificial substrate. Parametric studies of remelting sensitivity to surface convection and radiation changes, variations of impacting droplet temperature, droplet size and existing substrate temperature have been performed. The solidification process is completed so rapidly (on the order of milliseconds) that the heat transfer is basically a conductive process, and the surface convective and radiative effects are negligible. Numerical results also indicate that substrate remelting will not occur with realistic droplet temperatures (having less than several hundred degrees centigrade of superheating) on an *unheated* substrate. A remelting condition requires a substrate heated several hundred degrees above ambient temperature. This condition does exist when the preheating effect caused by previous droplets is included with the two-dimensional model.

For the case of a stainless steel droplet landing on a stainless steel substrate, numerical simulations are performed with initial "droplet" thickness of 100 microns and substrate temperature of 1100°C. The time-dependent solidification of this layer is shown in Figure 3 for two cases: a 1550°C and a 1650°C initial droplet temperature. The y-axis of Figure 3. indicates the location of the melting front; 0 represents the interface between the impinging droplet and the substrate, with the droplet extending in the positive direction.



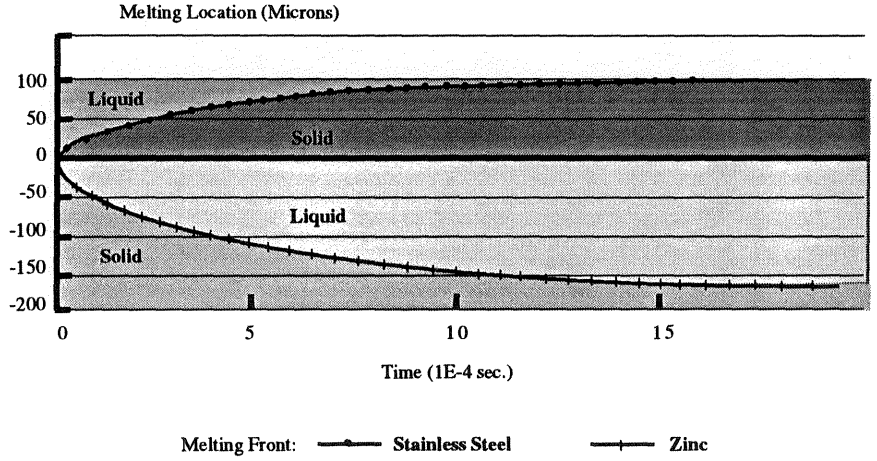
Initial Droplet Temperature 1550 Degrees



Initial Droplet Temperature 1650 Degrees

Figure 3: Substrate Remelting
(304 Stainless Steel Preheated substrate, Drop Size 100 Microns

At the start of the simulation the entire positive y region is liquid and the negative region solid. For the 1550°C temperature no substrate remelting occurs, while for the 1650°C case a small amount of remelting does occur. In Figure 4 the results for a stainless steel droplet on a zinc substrate are shown.



(Initial 1450 Degree SS droplet on 25 Degree zinc substrate)

Figure 4: Substrate Remelting (Stainless steel on zinc substrate)

The stainless steel droplet, initially at its melting temperature, solidifies while the lower melting zinc melts and actually vaporizes slightly. This result demonstrates the need to protect sacrificial support material.

## Mechanics Issues in Shape Deposition Processes

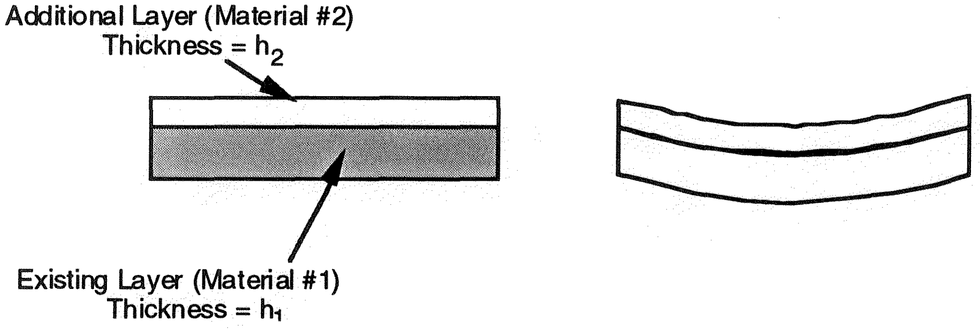
A current limitation of layered processing is the build-up of residual stresses as artifacts are manufactured. Residual stresses can affect artifact performance (response and life) and are also the root cause of specific deleterious effects including artifact warping, artifact delamination and stress cracking of brittle layers. Understanding the build-up of residual stresses and how to minimize them and their effects are thus the focus of current mechanics research into layered manufacturing.

### *Residual Stresses and Artifact Warping*

In the layered processing, residual stresses are built up as new layers are deposited onto existing layers of the artifact. This build-up is due to the contraction each new layer experiences as it solidifies and cools and occurs even in the successive application of layers of the same material. The process is illustrated in Figure 5 where, for simplicity, a single layer of one material is shown applied to a single existing layer of another material. The layer thicknesses may differ, however, it is assumed that each layer is beam-shaped. It is also assumed that the new layer experiences a uniform contraction as it solidifies on the existing layer and that the contraction can be characterized by a temperature-independent coefficient of thermal expansion, $\alpha$. Under these assumptions, the elementary analysis of Timoshenko [8] for the stresses in a uniformly heated bimaterial strip can be applied to predict the residual stresses in each layer and the curvature of the two-layered artifact caused by the contraction of the newly applied layer. The predicted curvature, $\kappa$, takes the form

$$\kappa \equiv \frac{1}{\rho} = \frac{-\alpha\,\Delta T}{\dfrac{(h_1+h_2)}{2} + \dfrac{(E_1h_1^3+E_2h_2^3)}{6(h_1+h_2)}\left(\dfrac{1}{E_1h_1}+\dfrac{1}{E_2h_2}\right)} \qquad (6)$$

where $\rho$ is the radius of curvature of the artifact. In eq (6) $\alpha$ is the coefficient of thermal expansion of the new layer and $\Delta T$ is the difference (negative in sign) between the solidification temperature of the new layer and the operating temperature. The layer thickness is designated by h and E is the Young's modulus of the layer.



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a) At solidification temperature of new layer

b) After new layer cools

Figure 5 Curvature induced by the thermal contraction of a new layer (material #2) after its application to an existing layer (material #1).

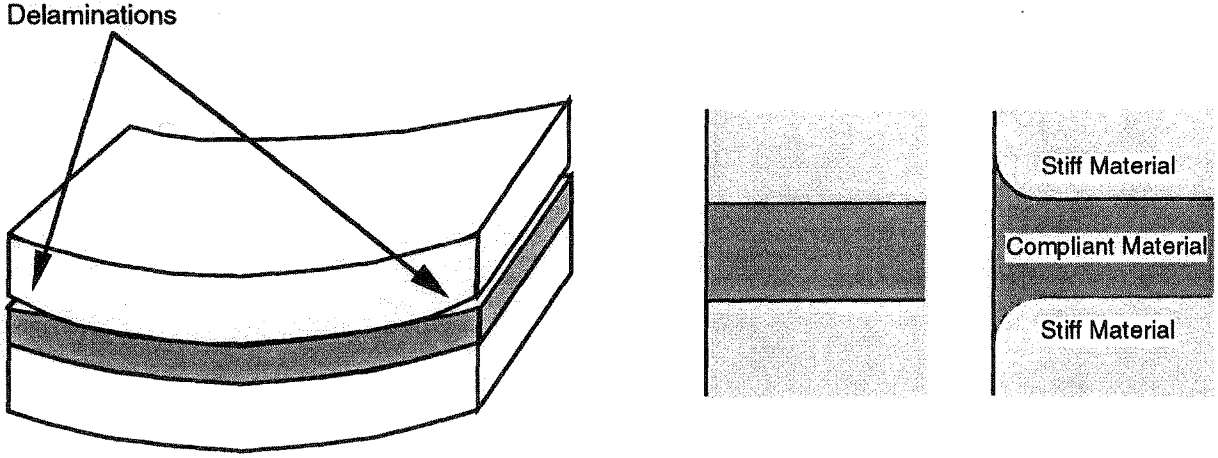
For this simple model the stresses in each layer of the bilayer artifact are composed of axial and bending components and thus vary linearly through the thickness of each layer. In layered manufacturing, this interaction between newly applied and existing layers is repeated for each additional layer applied. The curvature of the artifact and the residual stresses in it are increased with the addition of each new layer.

There is a need in layered manufacturing to experimentally quantify residual stresses created during the process by measuring curvature changes caused by the addition of new layers. Results can be compared to simple models of layer interaction such as the one above. It is expected that enhancements to the model will be necessary, including accounting for the temperature dependency of coefficients of thermal expansion and the modeling of non-uniform thermal contraction of individual layers. In particular, results from thermal modeling of the solidification of layers (see previous section) is expected to yield more precise layer residual stress distributions for use in the solid mechanics model. The goal would be to not only predict residual stress and curvature effects in geometrically complex artifacts, but to also shed light on material combinations and process procedures that minimize them. An example of a layered copper steel tube manufactured by Carnegie Mellon's MD* process [2] is shown in Figure 6

Figure 6. Layered copper steel tube manufactured in MD*

*Interfacial Debonding*

In addition to warping, residual stresses can cause delaminations between layers by acting as the driving force in the propagation of interfacial cracks from the edges of the artifact toward its center (see Figure 7 a). The delamination may propagate through the entire length of the artifact, separating it into two pieces. This is particularly a problem in the case of artifacts made of layers of different materials, due to the large stress concentrations that exist at the intersection of an uncracked bimaterial interface and a free edge.



(a) (b)

Figure 7 a) Edge delaminations and b) Untailored and tailored bimaterial interfaces

A fully elastic asymptotic analysis of a bimaterial interface intersecting a free edge (see Bogy [9] or Hein and Erdogan [10]) predicts that stresses are, in general, singular there. Using the notation that the stresses vary as r $^{(\lambda-1)}$ as r approaches zero, where r is measured from the intersection point, $\lambda$ can take on values from $\lambda$=1.0 (no singularity) to roughly $\lambda$=0.60. The value of $\lambda$ is a function of the relative mismatch in elastic properties of the two materials and the angle that the interface makes with the free edge. The stress singularities that result from an elastic analysis of the bimaterial problem are important for two reasons. First, they indicate that actual interfaces in manufactured artifacts exhibit very high concentrations of stress at their free edges. Second, from an analysis standpoint, because the strength of the singularity in the elastic stresses is a function of the material combination studied, analytical comparisons of delamination driving force between different material combinations is difficult if an uncracked interface model is used.

We are investigating several approaches to help minimize delaminations. For example, one approach involves attempting to tailor the geometry of the interface to eliminate undesirable elastic stress singularities. This approach was suggested to the authors by G. B. Sinclair and follows the work of Okajima [11] on the bimaterial interface problem and the role of interface geometry in adhesive tensile tests. In Okajima [11] it is shown that the stress singularity at a bimaterial free edge in adhesive specimens can be eliminated if the interface is made to intersect the free edge tangentially, as shown in Figure 2b. In fact, the angle of intersection with the free edge need not be tangential and is a function of the relative elastic mismatch between the layers.

*Stress Cracking*

Another problem associated with residual stresses layered manufacturing is the cracking of newly applied brittle layers as they cool and contract after being applied to existing layers of the artifact (see Figure 8). A separate but related problem is the cracking of brittle layers after they are embedded between ductile layers within the artifact, typically due to a combination of residual and applied tensile stress. It is important to predict, for a particular brittle material, the maximum allowable thickness of a newly applied layer so that stress cracking will not occur. Similarly, for embedded layers, it is desired to determine the relative thickness of an embedded layer so that no cracking will occur.

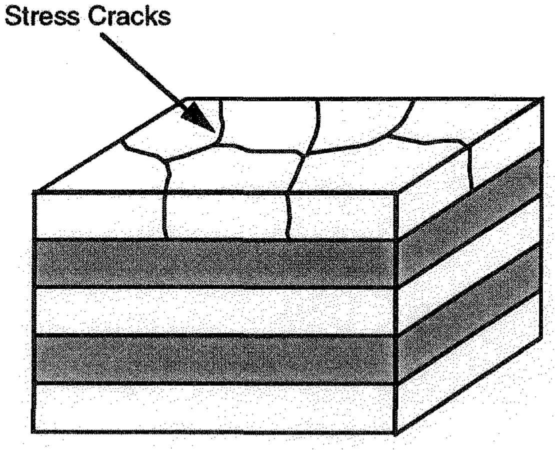


Figure 8. Stress cracking in a newly applied brittle layer.

## Summary

Layered manufacturing offers new opportunities for product design. This is true from a geometry and from a materials perspective. Objects of arbitrary geometric complexity can be built from a larger variety of material combinations than with conventional manufacturing methods. However, frequently the quality of the built articles (bond strength between layers and material density) and the rate at which they are created does not meet industrial demands. A better insight into the physics of the underlying bonding processes between layers and the resulting residual stress accumulation due to temperature gradients is expected to lead to improved performance of ojects my through layered manufacturing.

## Acknoledgements

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