A METHOD FOR DETERMINING THE CHARACTERISTIC SCALE FOR ADHESION FOR A DISCRETE BONDING MODEL ON A ROUGH SUBSTRATE

C.A. Brown¹ and S. Siegmann²

¹ Surface Metrology Laboratory Director
Worcester Polytechnic Institute
Worcester, MA 01609-2280 USA

² Head of Thermal Spray Group
Materials Technology Section
EMPA Thun
Feuerwerkerstrasse 39
CH-3602 Thun / Switzerland

Abstract

A discrete bonding model for adhesion on a rough substrate is presented. A key parameter in using the model is the characteristic scale, which is not always apparent from the adhesive system. A method for determining the characteristic scale using area-scale fractal analysis is presented. The method is tested on substrates prepared by grit blasting and coated by vacuum plasma spraying. The results support the discrete bonding model and appear to indicate characteristic scales for bonding.

1. Introduction

The objective of this work is to develop a method for determining the characteristic scale of adhesion. This characteristic scale is necessary to predict the adhesive strength on a rough substrate from a discrete bonding model (Brown 1994). The model is based on fractal properties that are common to many engineering surface textures (Brown et al. 1998). The objective of this paper is to demonstrate, using a thermal spray system, how this method can be applied. The method and the discrete bonding model (DBM) have the potential to be applied to many adhesive systems, and more generally to intensities of interactions, of which bonding would be one type, on rough surfaces.
1.1 The discrete bonding model (DBM)

The DBM attempts to predict the adhesive strength on a rough surface by summing the contribution of individual bonds. It assumes that the macroscopic adhesive strength is due to a large number of individual bonds.

\[ S_t = N \frac{S_s}{A_p} \quad [1] \]

Where \( S_t \) is the macroscopic adhesive strength in units of force per area, \( N \) is the number of adhesive bonds, \( S_s \) is the strength of an individual, i.e., single bond, and \( A_p \) is the nominal area over which the bonding is taking place. The nominal area is essentially the projection of the rough substrate surface onto a nominally horizontal plane.

The number of bonds that can be made on a rough surface depends on, \( A_s \), the area occupied by the bond, i.e., the characteristic scale, and, \( A_{ts} \), the total area of the surface measured at the scale of the bond including the area due to the topography of the surface texture

\[ N = m \frac{A_{ts}}{A_s} \quad [2]. \]

The total number of potential bonding sites is \( A_{ts}/A_s \), which is likely to be large since \( A_s \) is expected to be small. \( A_{ts} \) is evaluated, or measured, at the scale of \( A_s \) and, in general, will vary with the scale of measurement.

Since not all the potential bonding will be available \( m \) is a factor between 0 and 1, accounting for the fraction of available bond sites on the rough surface that is occupied. A potential bond site might be unoccupied because of contamination, or, in a further sophistication of the model, for geometric reasons, making \( m \) a function of the local topography of the surface. In this paper \( m \) will be considered to be 1.

Substituting equation [2] in equation [1] and rearranging gives:

\[ S_t = m \left( \frac{S_s}{A_s} \right) \left( \frac{A_{ts}}{A_p} \right) \quad [3]. \]

The term \( (S_s/A_s) \) has units of force per area and characterizes the individual bond. The term \( (A_{ts}/A_p) \) is called the relative area, and it characterizes the surface texture, or roughness. The relative area is the ratio of the total measured area at the scale of the bond to the nominal area. As \( A_s \) varies with scale, so the relative area also varies as a function of the scale of measurement, or evaluation, on a rough surface. Generally the finer the scale the larger the relative area. The relative area is always equal to or greater than one. At some sufficiently large scale most surfaces of technical interest appear smooth, and at this scale and larger the relative areas will be one.

The appropriate scale for determining the relative area so that equation [3] is valid is \( A_s \), the characteristic scale for that adhesive system. The characteristic scale should be sensitive to the composition and microstructure of the substrate and of the adhesive, as well as the method of its application. In general anything that would influence the character of the individual bond, i.e., \( (S_s/A_s) \), is part of the adhesive system. Since it is not obvious what the characteristic scale should be, it is essential...
to develop a method for its determination. It is also essential to have a method for determining the relative areas as a function of scale for a rough surface.

1.2 Area-scale fractal analysis and the determination of relative areas

The relative areas as a function of scale of a rough surface can be determined by applying a virtual tiling algorithm to a measured surface (Brown 1993, Brown et al. 1993). In this method triangular tiles, or patches, with the same area, but not necessarily the same shape, are used to virtually tile a measured surface (i.e., $z = z(x,y)$ or a regular grid in $x$ and $y$). The area of the tile represents the scale. The virtual tilings are repeated so that a range of scales can be represented. The measured, or apparent, area at a particular scale is equal to the number of tiles used in the tiling, times the area of the tile. The relative area is determined by dividing the measured area by the nominal area of the surface that has been covered in the tiling. An example of a sequence of tilings is shown in Fig. 1.

That the measured, or apparent, area is not unique, but depends on the scale of measurement or observation is a fractal geometric property. The fractal dimension, which could be used to characterize the complexity of the measured surface over some particular range of scales, can be determined from the slope of a log-log plot of the relative areas versus scales (i.e., area-scale plot, Fig.2):

$$D_{as} = 2 - 2 \text{slope}$$

[4]

where the subscripts ($D_{as}$) indicate that this is the fractal dimension determined from area-scale analysis. The fractal dimension itself, however, is not used in this method for determining the characteristic scale of adhesion, nor is it used specifically in the DBM for determining the adhesive strength. According to the DBM, the relative area at the characteristic scale for that adhesive system is the essential parameter for characterizing the rough surface geometry for understanding the roughness component of adhesive strength.

The determination of the characteristic scale is central to applying the DBM. In some adhesive systems it may be possible to suggest a characteristic scale from first principles. The complexity of the interaction of a liquid droplet and a solid substrate is complex enough to suggest an experimental approach to determine the characteristic scale, in addition to theoretical calculations.

One approach would be to test adhesive strength on a perfectly smooth surface. A perfectly smooth surface in this application is one where the relative area equals 1, to scales comfortably below the characteristic scale. With such a surface it should be possible to evaluate the term characterizing the bond, ($S_d/A_d$), directly. This, combined with a method for determining the relative area and appropriate tests of adhesive strength on this surface, would allow for the use of equation [3] to predict the adhesive strength on rough substrates. Still, it is not clear when a surface is sufficiently smooth for this approach to work, and it is not clear what influence of the texture at scales below the characteristic scale might have on the term characterizing the bond, ($S_d/A_d$). While this method is not the method used here, it certainly deserves further attention.
Fig. 1. Four tiling exercises from an area-scale analysis, illustrated on a surface measured by a scanning tunneling microscope (a diamond coating on a silicon substrate, fabricated and measured at UNCC). The triangular tiles, or patches, are shown in outline. In this method all the tiles in each tiling exercise have the same area in Euclidean 3-dimensional space, although the shapes and the areas of the projection of the individual tiles on the nominal surface vary. The nominal areas covered by each tiling exercise are represented on the ceiling of each box. The measured area can be calculated as the scale times the number of tiles.

1.3 Approach
In this work a method is tested whereby a number of substrates are prepared with a number of different rough surface textures. These textures are measured and the area-scale relations are determined over an appropriate range of scales. The surfaces are then coated by thermal spray and the adhesive strengths are determined experimentally according to ISO standard 14916 (1999). The scale where the relative area best correlates with the adhesive strength is determined from a series of plots of relative area versus adhesive strength.
2. Methods

Initially Steel alloy substrates with four different compositions and hardness were polished to a mirror finish to create a standard surface that would not influence the eventual texture of the substrate. Grit blasting was performed on all substrates at different impact angles, stand-off distances, pressures and with a different number of passes, in order to produce a variety of surface types on each substrate (Seigmann and Brown 1999).

The substrate textures were measured using a scanning stylus instrument (UBM) to measure the surfaces. The stylus radius was 5 µm, the sampling interval 2 µm and the scanned region was 500x500 µm.

Initial area-scale plots were used to determine the approximate scale of the smooth-rough crossover (SRC), the scale above which the relative areas are close to or equal to one and below which the relative areas are clearly greater than 1 (Fig. 2). It was believed that the characteristic scale for adhesion would be significantly smaller than the 12 500 µm², which is approximately the value of the largest SRCs. As it was possible that the SRC itself could be an interesting parameter for understanding adhesion, the size of the scanned region was adjusted so that the SRC would be included in the subsequent analyses of the measured surfaces. This avoided expending recourses to measure larger regions where the relative areas would be one. The sampling interval was selected as small as was reasonable with a 5 µm stylus radius in the hope that it would be small enough to include the characteristic scale. In this way the recourses for data acquisition would be concentrated in the region of scales most likely to yield interesting results.

Fig. 2: Relative surface area as a function of triangular patch area (µm²) for a St 37 steel substrate after 10 passes of grit blasting under 75° angle with pressure of 4x10⁵ Pa and 40 mm stand-off distance.
Area-scale analyses were performed on the measurements from each substrate prior to coating by vacuum plasma spraying. The relative areas were determined for scales between 2 μm² and 125 000 μm², which represent the areas of triangles with bases and heights corresponding to the sampling interval and scanned region respectively.

A 200 μm thick NiCr coating was applied at a chamber pressure of 100 mbar, a standoff distance of 275 mm and 44 kW of electrical power. The primary gas was argon at 46 l/min and the secondary gas was hydrogen at 6.5 l/min. The powder grain size was –53+20 μm.

Two identical adhesion test specimens were made for each substrate material and grit blast combination. Preliminary tests indicated that the variation in adhesion strength would be small, typically about ±5 MPa, encouraging a small number of samples.

For one substrate material the relative areas at one scale are plotted versus adhesive strengths of the coatings on the several different surface textures. The least squares regression coefficients, $R^2$, are determined. And, the process is repeated at other scales so $R^2$ values are determined over a significant range of scales. Then the regression coefficients are plotted versus scale to see if there is a tendency for a maximum.

It can be postulated from the DBM that there should be a maximum on the plot of $R^2$ versus scale. If the DBM is correct, then relative areas at the large scales should tend to underestimate adhesive strength, and the relative areas at the fine scales should overestimate adhesive strength. Therefore a maximum of $R^2$ with respect to scale is expected, and the scale with the highest regression coefficient, i.e., where $R^2$ is a maximum, should be the characteristic scale for this adhesive system.

### 3. Results and Discussion

Exemplar plots of relative area versus adhesive strength are shown in Fig. 3. These plots are for one of the substrate materials at four different scales. More plots, not shown here, were necessary to generate the $R^2$ versus scale plots for each substrate material (Fig. 4). The results of the regression analyses show varying degrees of linear fit with the linear regression. It appears that the regression coefficients generally increase as the scale decreases.

Plots of the regression analyses for the four substrates as a function of scale are shown in Fig. 4. There might be a tendency for the regression coefficients to increase with a decrease in scales from the largest scales, 2 000 μm² to about 100 μm², albeit with somewhat erratic behavior. And there could be a tendency for a maximum, at least in two cases.
The plot of $R^2$ versus scale for adhesion on the St 37 substrate show what could be interpreted as a broad maximum from about 200 $\mu$m² to about 20 $\mu$m², as $R^2$ exceeds 0.70, although the maximum $R^2$ exceeds 0.80 at a scale of 1 627 $\mu$m². The regression coefficients for relative areas versus adhesion on the XCrNi18/10 substrate follow a similar pattern, although the relative maximum for $R^2$ is higher, and at a finer scale.

Fig. 3. Relative areas at four different scales (indicated on the plots) versus adhesive strength, for the 100Cr6 substrate.
The values of $R^2$ versus scale for adhesion on the 42CrMo4 and 100Cr6 substrates still seem to be increasing at the finest scale, 2 $\mu$m$^2$. If there is some kind of maximum, as might be expected, it could be in the region of erratic behavior above 200 $\mu$m$^2$, or it could be below 2 $\mu$m$^2$.

![Graphs showing regression coefficients, $R^2$, for relative area as a function of adhesive strengths on substrates with different textures, versus scale for the four substrate materials.](image)

Fig. 4. Regression coefficients, $R^2$, for relative area as a function of adhesive strengths on substrates with different textures, versus scale for the four substrate materials.

The size of the particles could be a critical scale that might be anticipated in thermal spray. In this case, assuming spherical particles the cross sectional areas would be between 314 and 2 206 $\mu$m$^2$. This is approximately the range of erratic scales on the $R^2$ versus scale plots. There may be more than one characteristic scale for an adhesive system as complex as thermal spray coatings could be. It is possible that there is a critical scale in the region of the drop size, and others at some more fundamental scales of bonding, possible below the resolution of the surface measurements.
The resolution of the measuring device could also be a factor in the form of the area-scale and $R^2$ versus scale plots. There appears to be a slight change in the slope of the area-scale plot in the region of about $30 \mu m^2$. This corresponds to a circular contact area with a radius of about 3 $\mu m$, which in turn corresponds to a height of about 1 $\mu m$ on a spherical stylus. The average roughness (Ra) on these surfaces varied from about 2 to about 7 $\mu m$. It is not difficult, especially given the nature of a grit blasted surface, that the stylus would engage asperities on the surface at a significant distance from the center of the tip, which would gradually reduce the resolution of the tip beginning at scales below about $30 \mu m^2$. This lack of resolution could lead to the apparent lack of a relative maximum at scales below 100 $\mu m^2$.

4. Conclusions

A potential method for determining the characteristic scale of adhesion on a rough substrate is demonstrated and appears to be consistent with a discrete bonding model of adhesion.

The relative area at certain scales correlates well with adhesive strength at certain scales or over certain scale ranges, with $R^2$ values as high as 0.90.

5. References