Beyond Fresnel: absorption of fibre laser radiation on rough stainless steel surfaces

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Abstract

The standard procedure for the calculation of the angle-dependent absorptivity consists of applying the well-known Fresnel formulas to the material property refractive index \(n\) and extinction coefficient \(k\). However, the Fresnel formulas are only applicable for perfect smooth surfaces. Hitherto the effect of rough surfaces on absorptivity values and profiles are not thoroughly explored. Yet, this knowledge is vital for many kinds of theoretical consideration in computational models as they are applied in laser material processing, imaging and metrology. Therefore, we present experimental results on the angle-dependent absorptivity of stainless steel with five different surface finishes for fibre laser radiation with a 1.07 \(\mu\)m wavelength and for s- and p-polarisation separately. We can show that for probes which fulfil the modified Fraunhofer smoothness criterion at normal incidence \((R_z < \lambda/32)\), the Fresnel formalism is applicable. For the tested probes with higher surface roughness, the real absorptivity differs considerably from this solution: the maximum of absorptivity at the pseudo Brewster angle is decreased and shifted towards lower inclinations. Furthermore, other effects are observed which are not explained by theoretical approaches like Fresnel or ray-tracing models.

Keywords: absorptivity, rough surfaces, optical properties

(Some figures may appear in colour only in the online journal)

1. Introduction

Knowledge of the absorptivity of a material is one of the fundamental necessities for any kind of theoretical consideration in the field of industrial processes using light matter interaction. It is needed for the calculation of the energy input from a laser beam to a workpiece and can therefore be used to calculate the thermal load of the workpiece. Further, the angle-dependent absorptivity is the starting point for the modelling of processes using laser beams as a heat source. For these purposes, either tabulated literature values of the optical constant refractive index \(n\) and extinction coefficient \(k\) are deployed from standard text books [1–3], or \(n\) and \(k\) are calculated using the Drude theory with an application adapted extension [4–7]. Then, using the well-known Fresnel formulas, the absorptivity of the workpiece can be estimated. However, three problems arise from this procedure for real processes. First, the literature values for \(n\) and \(k\) are only valid for very special conditions which are linked to the underlying measurement method. For example, \(n\) and \(k\) are reported for room temperature [8], while real processes mostly involve melting or even evaporation. The applicability of these values is therefore questionable. Second, the Drude model only performs well in the intermediate infrared region while it struggles for short wavelengths [9] due to the anomalous absorption from surface imperfections. Third, the usage of Fresnel’s formulas can have an influence on the accuracy of the estimates. It is commonly known that these are only applicable for perfect smooth surfaces, whereas the range of validity of them and the influence of surface roughness is up to now unknown. So even if \(n\) and \(k\) values are available for the evaluated material–wavelength–environmental combination, there is a large uncertainty in estimating the absorptivity through the
application of the Fresnel equations. This article addresses these open questions.

Bennett et al [10] first tried to experimentally evaluate the effect of rough surfaces on the reflectivity of metals. The study showed that the absorptivity at normal incidence increases at higher surface roughness for the evaluated range of $R_{\text{ms}} < 0.01 \, \mu m$—the corresponding roughness values and wavelengths of the cited articles can be found in table 1. These findings were later validated for surface roughness heights up to 3.5 $\mu m$ [11], while in some experiments only a correlation at higher surface roughness levels was found [12, 13]. Therefore, the relation between the surface roughness and the wavelength of the light is thought to have an influence on the absorptivity [10, 14, 15]. Other studies demonstrated an absorption maximum at specific surface roughness heights [16]. Endriz and Spicer [17] evaluated the reflectance of micro rough aluminium surfaces ($R_{\text{ms}} < 0.01 \, \mu m$) and observed local peaks at specific wavelengths, which were shifted to other regions for a changed surface roughness. These findings were explained with the excitation of surface plasmons.

Beaglehole and Hunderi [18] evaluated the scattering on rough surfaces and demonstrated that the reflectivity of the parallel polarised (p-polarised) light component is more influenced by the surface roughness than the perpendicular polarised (s-polarised) radiation. Elson and Sung [19] theoretically demonstrated that peaks at different inclination angles of the p-polarised component occur at higher surface roughness, which cannot be explained by the Fresnel formulas. The authors related these peaks to surface plasmons due to the damped surface periodicity of the diamond turned surface. Further, results of other theoretical considerations indicate a shift of the pseudo Brewster angle to lower inclination angles caused by rough surfaces [20–22].

The experiments of Baria and Bautista [23] validated the effect of the surface roughness on the absorptivity at higher temperatures with the result of a more intense absorption peak at specific temperatures at higher surface roughness. Wieting and DeRosa [24] measured a 30% higher absorptivity of a CO2 laser beam on ground than on polished surfaces at room temperature, while this difference disappeared at higher temperatures.

Hence, while for some dependencies experimental results can be found, others, like the behaviour at high inclination angles, are only theoretically evaluated. Some results in literature even contradict themselves. However, the availability of approved absorptivity values is crucial for any theoretical consideration of technical surfaces. This study therefore experimentally evaluates the validity of the Fresnel formulas in terms of the angle-dependent absorptivity for standard-type 304 stainless steel with five different surface finishes (as received, K240 ground, matt polished, shiny polished and sandblasted). Further, the dependence on the polarisation state of the fibre laser is evaluated. As a result, the range of validity of the Fresnel equation can be analysed and assessed for this material. The paper is structured as follows. In section 2 the determination method for the estimation of the absorptivity is described. In section 3 the results are presented and discussed regarding the validity of the Fresnel relation. Section 4 summarises the main results of the article.

### 2. Material and methods

Due to its wide application in industry, metal sheets of AISI 304 (X5CrNi18-10) stainless steel with different surface finishes were used for the determination of the absorptivity behaviour.

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1 Since for opaque materials like metals the transmission can be neglected, the reflectivity and absorptivity are directly linked through the equation $R = 1 - A$.

4 In article [23] the effect of surface roughness on emissivity is studied. According to Kirchoff’s law of thermal radiation the direct spectral emissivity is equal to the direct spectral absorptivity, wherefore it can be analysed here in terms of direct spectral absorptivity.
We used cold rolled steel plates which were pickled after the hot rolling process to remove the oxidation layer. The probe sheets were cleaned with ethanol before experimentation to remove any existing dirt or grease. The absorptivity of 304 stainless steel is assumed to be temperature-independent for a temperature range up to 700 °C for light with a 1.07 µm wavelength [25], wherefore the temperature dependence, which might have an influence on absorptivity [23, 24], may be neglected in our experiments. The analysed surface finishes and the corresponding roughness heights in terms of $R_{\text{rms}}$ as well as the correlation length $a$ of the surface roughness are listed in Table 2. These values have been measured using confocal microscopy along the plane of incidence $R_{\text{rms}}(X)$. For the isotropy test, roughness values perpendicular to the plane of incidence $R_{\text{rms}}(Y)$ have been measured. If $R_{\text{rms}}(X)/R_{\text{rms}}(Y) \approx 1$, the surface is regarded as isotropic.

Table 2. Summary of the experimental results for the absorptivity at normal incidence, at the pseudo Brewster angle, the location of the pseudo Brewster angle, as well as the surface finish-induced absorptivity.

| Surface Finish | $R_{\text{rms}}$ [µm] | $a$ [µm] | $\lambda/R_{\text{rms}}$ | $A(\theta = 0)$ | $A_{\text{SFIA}}(\theta = 0)$ | $A(\theta = \theta_B)$ | $\theta_B$ [$^\circ$] |
|---------------|-----------------|------|-----------------|----------------|-----------------|-----------------|----------------|
| Polished shiny | 0.0008 | 2.4 | 1338 | 22 | — | 70 | 82 |
| Polished matt | 0.016 | 2.91 | 67 | 26 | 18% | 69 | 81 |
| As received | 0.176 | 4.09 | 6.1 | 28 | 27% | 66 | 80 |
| K240 ground$^b$ | 0.776 | 14.3 | 1.39 | 33 (p-pol) | 50% | 63 | — |
| Sandblasted$^c$ | 2.31 | 37.1 | 0.46 | 56 (p-pol) | 154% | 88 | — |

$R_{\text{rms}}$ and $a$ are measured along the plane of incidence $R_{\text{rms}}(X)$. For the isotropy test, roughness values perpendicular to the plane of incidence $R_{\text{rms}}(Y)$ have been measured. If $R_{\text{rms}}(X)/R_{\text{rms}}(Y) \approx 1$, the surface is regarded as isotropic.

$^a$ Cold rolled—pickled.

$^b$ Surface anisotropic: $R_{\text{rms}}(X)/R_{\text{rms}}(Y) = 2.3$.

$^c$ Surface slightly anisotropic: $R_{\text{rms}}(X)/R_{\text{rms}}(Y) = 1.9$.

We test if these criteria can be used to assess the validity of the Fresnel formalism for technical rough surfaces. Therefore we classify the surfaces of our probes as smooth or rough using these criteria at normal incidence ($\theta = 0$). In contrast to the literature, the roughness value of $R_z$, which is the average of the distance between the highest peaks and valleys in five measuring sections (according to DIN EN ISO 4287 ASME B46.1), is used to approximate $\Delta h$, since it is less influenced by scratches or impurities in the roughness profiles than the maximum surface roughness height at one point.

The method for the absorptivity measurement is described in detail in [27], and the setup is illustrated in figure 1. A linear polarised 400 W fibre laser beam with a wavelength of $\lambda = 1.07$ µm is projected onto the workpiece using a linear polariser.
collimating lens with a focal length of 50 mm. This results in a collimated beam with a radius of \( \omega_0 = 3.35 \) mm. The laser beam can be inclined from 0 to 86°. The plane of incidence is perpendicular to the workpiece surface and lies in the direction of the translation motion. The electrical field \( \mathbf{E} \) is adjusted to be either parallel (p-polarisation) or perpendicular (s-polarisation) to the plane of incidence. The rolling direction of the cold roll process is for all samples perpendicular to the plane of incidence. However, only the ground and sandblasted probes show a surface roughness anisotropy (see table 2), wherefore it is believed that the effect of rolling orientation for the other probes is small.

For the determination of the absorptivity, temperatures inside the defined measuring areas on the workpiece are recorded using a thermographic setup. The measuring areas are located apart from the processing zone to avoid fluctuations of the measuring signal as reported by Gonçalves et al [28]. The temperatures are recorded during the irradiation and the subsequent 120 s to capture the heating as well as the cooling regime. The workpiece is thermally isolated and moved under the laser beam with a translation speed of 1 m min\(^{-1}\). The path of motion is 150 mm long, so that possibly existing local impurities or surface defects should not primarily influence the measuring result.

The resulting temperature–time curves of the irradiation are then used to adjust the computational heat flow model. This model is evaluated in detail in [27], where it is demonstrated that a fit between the experimental curves and theoretically calculated ones is only given for one particular value of absorptivity. The total measurement duration of the model is stated to be better than 2% [29]. The model has to be adjusted for the inclination of the laser beam during the experimental series in this study. This leads to a projected elliptical spot on the workpiece surface with enlarged radius and thus, a decreased laser beam intensity. The heat source definition of Goldack et al [30] is modified and used as a heat flux distribution:

\[
q(x, y) = \frac{2A_{opt}P_l}{ab\pi} \exp\left(\frac{2(x - x_0)^2}{a^2}\right) \exp\left(\frac{2(y - y_0)^2}{b^2}\right)
\tag{3}
\]

with \( A_{opt} \) the absorptivity, \( P_l \) the laser power, \( x_0 \) and \( y_0 \) the position of the heat source at \( t = 0 \), and \( a \) and \( b \) the spot radii in the \( x \) and \( y \) direction (see figure 1), respectively.

Further, for the computational model and the thermographic measurement of temperatures, the emissivity is needed. The emissivity values of the probes with the different surface finishes were experimentally determined in oven experiments from room temperature up to 200 °C. The measured emissivity showed no temperature dependence.

For a better quantitative comparability of the absorptivity for the different surface roughness heights as well as to already published literature values, \( n \) and \( k \) were determined by a graphical method using the simplified Fresnel equations in the formulations [31]

\[
A_\text{s} (\theta) = \frac{4 \ast n \ast \cos \theta}{(n^2 + k^2) + 2 \ast n \ast \cos \theta + \cos^2(\theta)}
\tag{4}
\]

and

\[
A_\text{p} (\theta) = \frac{4 \ast n \ast \cos \theta}{(n^2 + k^2) + 2 \ast n \ast \cos \theta + 1}.
\tag{5}
\]

which are valid in the range of

\[n^2 + k^2 \gg 1.\tag{6}\]

The unknown \( n \) and \( k \) values are evaluated using the Levenberg–Marquardt algorithm [32] for a fit between the experimental absorptivity values and the Fresnel formulas. As the input values of the Levenberg–Marquardt algorithm as a non-weighted least squares method, \( n \) and \( k \) from the literature [33] are used. Since the phenomenon of light absorption at rough surfaces also induces multiple reflections, the calculated values for \( n \) and \( k \) using this method only equal the fundamental optical parameters if multiple reflections are absent. In all other cases, the calculated \( n \) and \( k \) are integrated values over all actually occurring reflections and are therefore termed \( n_i \) and \( k_i \). Since this study also seeks to answer the open question about the applicability of Fresnel at rough surfaces, this method is nevertheless useful. Using test statistics for the Levenberg–Marquardt algorithm one is able to estimate the error by applying the classical Fresnel theory.

The method is also applicable for other materials and wavelengths by using the complex form of the Fresnel formulas (see for example [34]) as a fit function. In this work the introduced error because of the usage of the simplified expression (2) and (3) was estimated and is negligible.

3. Results and discussion

For the determination of the absorptivity, temperature–time profiles were measured and then matched with the corresponding computational model. A characteristic temperature profile inside a measuring area in comparison to the computed profile is illustrated in figure 2. The fit between the experiment and computation is good over the full period of the measuring time of 150 s. The peak signal of the presented experimental
run is 8 °C, resulting in a signal-to-noise (SNR) ratio of 12. For the trial with the lowest temperature rise, the SNR is still 4, which is sufficiently large for a reliable evaluation. The maximum temperature inside the spot area was below 500 °C during all experiments, wherefore oxidation of the surface can be excluded.

3.1. Polished probes (shiny)

The measured absorptivity profiles for the smooth classified probes (according to the modified Fraunhofer criterion) are depicted in figure 3(A). The measuring accuracy amounted to ±2% during the experiments, which is drawn in figure 3(A) as the confidence band. Further, the calculated $n$ and $k$ for the polished probes and the resulting absorptivity curves are plotted in figure 3(A).

It is evident from figure 3(A) that the fit of the Fresnel formula to the experimental values is good, resulting in an $R^2$ of 0.903 and an adjusted $R^2_{\text{adj}}$ of 0.891. This indicates that the evaluated probes (figure 3(B)) can be considered as smooth and the Fresnel equations describe the behaviour of the angle-dependent absorptivity well. The absorptivity at 0° inclination is evaluated at 22% and therefore lies under the absorptivity of the main alloy elements of Fe (35.3% [8]), Cr (36.99% [35]) and Ni (27.9% [36]). The pseudo Brewster angle $\theta_B$ is observed during the experiments at 82° inclination reaching a maximum value of $A_P = 70\%$. One reason for the deviation from the fitted curve at inclination angles around 70° may be the imperfect linear polarisation of the used laser beam, whose polarisation ratio is >50:1. Another reason may be the state of the surface and small surface defects (see figure 3(B)). Also, measuring errors due to a possible fluctuation of the laser beam diameter or the measuring signal can cause such differences. In figure 4, the experimental absorptivity values are compared to values which can be sparsely found for 304 stainless steel in literature. At normal incidence, the measured absorptivity is much smaller than the one in [25, 37]. The values in [25] are measured at probes with a surface roughness of $R_a = 0.331 \ \mu m$. According to the modified Fraunhofer criterion, these probes must be classified as rough. Further, the surface roughness of the probes in [37] is not specified. A comparison is therefore difficult. The values from Boyden and Zhang [33] were derived from theoretical calculations based on the Drude theory. This was adjusted to allow the calculation of alloys by using the law of mixtures for the determination of the number of free electrons $N$, the optical mass of the electrons $m^*$ and the damping frequency $\tau$. Although the evaluated 304 stainless steel has complex Fermi surfaces and structural disorders [38], the simple Drude theory, which neglects the effect of interband transitions [39], is capable of estimating the optical constants of stainless steel in the near-infrared region to be in good agreement with the experiments presented here.

3.2. Polished probes (matt)

The measured absorptivity profiles for the matt polished probes are shown in figure 5. The absorptivity at normal incidence is evaluated at $A = 26\%$, which is 18% more than the
shiny polished probes. At the pseudo Brewster angle $\theta_B$ of 81°, the absorptivity reaches a maximum of $A_P = 69\%$. Thus, the level of this maximum is comparable with that of the polished probes. However, the pseudo Brewster angle is slightly shifted to a smaller inclination. The theoretical predictions of a shifted pseudo Brewster angle in [20–22] are therefore confirmed by the experiments presented here.

The calculated values for $n_i = 3.89$ and $k_i = 6.15$ differ considerably from the ones for the smooth surfaces. While the fit for the p-polarisation is a good approximation of the experimental profile, the one for the s-polarisation shows qualitative deviations. This results in a coefficient of determination $R^2$ of 0.54 and an adjusted $R^2_{adj}$ of 0.48. In other words, only 50% of the observed dependencies are captured by the underlying Fresnel model. Interestingly, the s-polarisation differs more from the Fresnel curves than the p-polarisation, which contradicts the theoretical considerations in literature [18, 19]. Thus, already at very small surface roughness heights of $R_{rms} = 0.016\ \mu m$ and a wavelength-roughness relation of $\lambda/R_{rms} = 67$, the experimentally observed absorptivity differs considerably from the Fresnel solution. Since the probes still meet the condition for a smooth surface according to the modified Rayleigh criterion at normal incidence $R_Z < l/8$, this definition is not suitable for the assessment of the validity of the Fresnel equations. It is worthwhile mentioning here that 304 stainless steel is susceptible to surface deformation [24], which can be seen in figures 5(B) and 6(B). However, the observed change in absorptivity cannot be separated in parts being induced by roughness or surface deformation. Therefore, the change in absorptivity is related to the previous mechanical surface finish.

3.3. As-received probes

The tendencies observed for the matt polished probes (3.2) are more pronounced for the as-received probes (see figure 6(A)). At normal incidence, the absorptivity amounts to $A = 28\%$, while the maximum at the pseudo Brewster angle $\theta_B$ of 80° amounts to $A_P = 66\%$. Hence, the absorptivity of the as-received probes at normal incidence is 28% more than that for a polished shiny surface, while the observed absorptivity maximum at the shifted pseudo Brewster angle is lower. Further, the absorptivity of the s-polarisation does not show the expected dependence on the incidence angle anymore.
The absorptivity only decreases from $A = 28\%$ at normal incidence to $A_S = 16\%$ at $80^\circ$.

It is obvious from figure 6(A) that at the roughness of the as-received probes (see figure 6(B)), the Fresnel solution for the angle-dependent absorptivity does not reflect the tendencies observed in the experiments. This is quantitatively confirmed by a negative adjusted $R^2_{\text{adj}}$ of the fit model. This means that the applied model for the fit is not suitable to explain the observed effects. Therefore, at roughness heights in the region of the wavelength ($\lambda/R_{\text{rms}} \approx 1–10$) and consequently for the most commercially available metal sheets, the Fresnel formulas are not applicable to estimate the real absorptivity.

3.4. K240 ground/sandblasted probes

The measured angle-dependent absorptivity of the ground (K240) probes is plotted in figure 7(A). Further, the angle-dependent absorptivity of the sandblasted probes is shown in figure 7(C). The fit of the Fresnel equations to the experimentally measured absorptivity diverges for these two surface roughness states, wherefore no values of $n$ and $k$ can be determined using the Levenberg–Marquardt algorithm. At normal incidence, the absorptivity of the p-polarisation evaluates to $A_p = 33\%$ for the ground probes and $A_p = 55\%$ for the sandblasted probes. The maximum absorptivity for the ground probes is decreased to $A_p = 63\%$, while it is increased for the sandblasted probes to $A_p = 88\%$ compared to the smooth surfaces.

For these probes, three additional effects come into play, which are not represented by the classical Fresnel formulas and make the fit to the experimental values unusable. First, the absorptivity for s- and p-polarisation does not show the same value at normal incidence. It is assumed that this phenomenon is directly linked to the surface profile. While the surfaces of the probes previously presented (3.1–3.3) can be regarded as isotropic, the probes with the ground and sandblasted surface finish show a pronounced anisotropy, as illustrated in figures 7(B) and (D) as well as table 2. During the experiments, the translation motion of the probes and therefore either the s- or p-polarisation plane was perpendicular to the characteristic grooves. Hence, the polarisation-dependent absorptivity must be anisotropic, resulting in the spread of s- and p-polarisation at normal incidence.

The second effect relates to the angle-dependent absorptivity in the case of the s-polarisation. Over the complete range of evaluated inclination angles, the absorptivity of the s-polarisation remains on the same level for the K240 ground probes and even increases for the sandblasted probes. This
may be reasoned by the characteristics of the surface. At high inclination angles the laser beam hits the surface in the middle of the hills. Therefore, the effective angle of the impinging laser beam is reduced, resulting in higher absorptivity values compared to smooth probes. Additionally, through the characteristics of the surface, the beam is now not purely polarised in respect to the surface structure, it is rather a mixture of s- and p-polarisation. Therefore it is expected that the s-polarisation is increased while the p-polarisation decreases, which can be observed from the polished shiny probes to the sandblasted probes with increasing strength. The effect of angle-independence was also predicted by theoretical considerations for circular polarisation [40, 41]. Our experiments show that this phenomenon also occurs at specific roughness characteristics ($R_{\text{rms}}/a = 0.05$) for the absorptivity of the s-polarisation of linear polarised light.

Third, the absorptivity for the p-polarisation shows local peaks at specific incidence angles, which are more pronounced for the sandblasted probes. In the literature, the observed local absorption peaks are explained by the coupling to so-called spoof surface plasmon polaritons (SPPs) due to damped surface periodicity [19]. In the presented case, natural surface plasmon excitation can be excluded. The excitation of natural surface plasmons only happens when, for metals with a negative real part of the dielectric function, the relation

$$\frac{\omega_p}{\sqrt{2}} < \omega_L,$$  (7)

with $\omega_p$ the plasma frequency of the bulk material and $\omega_L$ the frequency of the laser beam, holds true [42]. In the case of the evaluated 304 stainless steel, $\omega_p$ equals $2.266 \times 10^{16}$ 1/s [33]. Coupling to natural surface plasmons therefore occurs at laser wavelengths near $\lambda = 117$ nm and can be excluded for the laser frequency $\omega_L = 1.762 \times 10^{15}$ 1/s [43] of the used fibre laser. In contrast, spoof SPPs, e.g. observed by [44] for 2D grooves on a metallic surface, cannot be excluded a priori. However, the strength of the contribution of the possibly occurring spoof SPP is out of reach for a quantitative assessment but is included in the overall absorptivity enhancement due to the previous mechanical surface preparation. The phenomenon of local peaks, either due to spoof SPP, surface deformations or geometrical conditions, was not predicted by ray-tracing analysis in the literature (e.g. in [20]).

### 4. Summary and conclusion

In this study the effect of rough surface conditions on the angle-dependent absorptivity was evaluated. As a result, the validity of the Fresnel formulas for the calculation of the angle-dependent absorptivity on technical rough surfaces could be assessed. Table 2 summarises the quantitative results of this analysis in terms of absorptivity at normal incidence, absorptivity at the pseudo Brewster angle and surface finish-induced absorptivity ($A_{\text{SFIA}}$) as the absolute percentage change in absorptivity due to the previous mechanical surface finish in comparison to the polished shiny surface. $A_{\text{SFIA}}$ therefore includes the roughness-induced absorptivity effects due to surface deformation and defects as well as due to possibly occurring spoof SPPs. Further, following conclusions can be drawn from the experimental evaluations:

- Results showed that for the probes which fulfil our modified Fraunhofer criterion at normal incidence ($R_{\text{rms}}/a < \lambda/32$), the Fresnel equations can be used for the determination of the angle-dependent absorptivity if n- and k-values are known.
- At higher surface roughness, the pseudo Brewster angle is shifted towards smaller inclination angles and the corresponding absorptivity maximum is decreased.
- At specific surface roughness levels ($R_{\text{rms}}/a = 0.05$), the absorptivity of the s-polarisation can be considered as angle-independent.
- The observed experimental effects of local absorption peaks are up to now not predicted by ray-tracing analyses.

These conclusions are of special interest for the wide range of research dealing with laser material processing. The understanding of underlying physical phenomena and involved interactions with predefined environmental conditions like surface roughness supports the interpretation of processing results.

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