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Mathematical modelling of sound transmission loss (STL) in metallic and graphite based coatings

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ABSTRACT

This study aimed to investigate the relationship between the sound transmission loss (STL) properties and surface morphologies of metallic-based and graphite-based coatings. Aluminium (22 µm average particle size (APS) and 12 µm APS), copper (12 µm APS), silver (14 µm APS) as conductive metal pigments and graphite (18 µm APS) as a semi-conductive pigment were used to create coatings and STL properties were measured with an in-house designed sound wave modulation device. Laser beams with different carrier wavelengths were used in the experiment, for which the wavelength with the highest R^2 and number of significant variables was used to create a mathematical model to help in measuring the sound transmission loss properties. Surface tension energy, conductivity, permeability, reflectivity, concentration and the type of pigment were found to be significant in determining the STL properties.

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KEYWORDS

Metallic-based coating; conductivity; sound transmission loss (STL); optoelectronics; graphite-based coating

Introduction

Coatings and paints are a part of the coating material classification which has a very wide range of functions. The function of the coating (metallic coating, solid coating, etc.), the layer of the coating (undercoat, surface, etc.), compatibility with the environment (water-based, powder, radiation curable, etc.), the processing conditions (oxidative curable coating material etc.) or decorative finishes (automobile paints, decorative paints, industrial paints, etc.) are some of the sub-sections of these.¹ Metallic-based coatings can provide an aesthetically appealing display since pigments like aluminium, copper, copper–zinc alloys, etc. have flakes/platelets structure. With the growing waterborne technology, the metallic-based coating industry has an increased demand day by day.² As a matter of fact, aluminium and copper pigments are gaining more popularity due to their unique ability to make a gold-bronze effect.³ In industries like textile, plastics, printing inks, painting, coating, furniture and many others, mostly platelets of aluminium, copper, copper alloys, bronze, nickel or stainless steel metallic pigments are used.⁴ The platelets of metallic substance settle in a layer, sit parallel to each other and form a light reflecting surface; this causes the shining and appealing look. In addition, the layer creates a surface hardly penetrable by liquids when it is hardened.¹

The most important metallic pigment is the aluminium pigment, which is prepared by grinding the metal in steel rolling mills.⁵ The application of pigments creates two types of coating surfaces which are leafing and non-leafing coatings.⁶ In the leafing aluminium coating, because of the low surface tension, the platelets are forced to the surface of the coating. This causes the bright metallic appearance and prevents oxygen and water vapour penetration.⁷ Since aluminium coatings create a non-penetrable barrier on the top layers of the steel-based structures, aluminium coatings are used for anti-corrosive purposes. Non-leafing aluminium

pigments surface tension is higher, which makes them more usable in automobile coating.⁸

For the moving parts of a machine, where there is not enough lubricant, a soft coating technique is used in which graphite is one of the usual selections.⁹ There are many studies about the graphite coatings lubrication and thermal properties.^{10–12} In addition, there are many studies with graphite foams sound absorption properties yet the coating is not widely used for absorption purposes.⁹

Chemical formula, physical shape and optical properties constitute the differences for the pigments. Most of the properties change only with the chemical structure.¹³ Colour, particle size, particle and crystal structure, surface area are all affected by the molecular structure of the pigment. Therefore, the pigment used in the coating material becomes decisive in properties such as conductivity, surface tension energy and optical properties.¹⁴

One of the surface tension energy measurement methods is the contact angle method which is defined as calculating the free energy on the surface of material for the unit quantity.¹⁵ This method helps to determine the hydrophilicity-hydrophobicity of the surface by using a liquid, the surface tension energy of which is known.¹⁶

The particle size of the pigment changes the colour strength, permeability of the pigment changes its opacity and durability of pigment changes the solvent resistance. The energy absorbance increases as the particle size of the pigment decreases. Therefore, energy transparency of the pigment is an indicator for the area of use for the coating.¹⁷

Every colour can be represented with three main colours in the RGB model. The International Commission on Illumination (CIE) in 1931 stated that red, green and blue are the three main colours and the other colours can be measured with the R (red), G (green) and B (blue) values. Therefore, the pigments' differentiation can be linked with RGB values.^{18,19} With

Euclidean distance and vector angle methods the amount of the pigment colour (RGB) difference can be calculated and represented with ΔE_{RGB} .²⁰ Thus, definitive criteria are created for the type of the pigment which causes the difference in permeability, reflection, surface tension energy, conductivity, etc.

Technological developments in the telecommunication and electronics field in the most recent decades created a problem called electromagnetic interference. Shielding materials are mostly used to overcome this problem, which means mainly covering the structures with electromechanically conductive materials.²¹ For conductivity (σ) measurements the four-point-probe method is a well-established method. The main aim is to measure the resistivity (ρ) of various geometries where conductivity is $1/\rho$.²²

This study contains aluminium, copper, silver and graphite-based coatings produced in the laboratory, which are applied on a 1 mm thick glass surface (lamellae) with the thickness of 60, 90 and 120 μm . The main components of the coatings are the metallic and graphite pigments, since 83% pigment together with 17% binder and solvent are used in preparation of the coating. In addition, three different concentrations of the pigment are used to learn the effect of the concentration. Lasers with three different energy levels are used for determining the permeability of the coatings, as pigments with different particle sizes can create a difference in the permeability. For the surface morphology, the surface tension energy is determined and the sound transmission loss properties are found by an in-house laboratory system. Sound transmission loss (STL) relationships to surface tension energy, conductivity, pigment type (RGB), pigment concentration, permeability, reflectance and thickness of coating are assessed.

Materials and methods

In this study, caradol oil and MDI, a binder for the composite structures, were mixed in a 50–50 ratio for a total of 0.1 grams and then mixed with two different particle size aluminium (22 and 12 μm APS), copper (12 μm APS), silver (14 μm APS) as conductive metal pigments and with graphite (18 μm APS) as a semi-conductive pigment. Three different ratios were used to understand the effect of pigment ratio, by mixing 0.1 grams of binding agent together with respectively 0.5, 0.7 and 0.9 grams of pigments in total in laboratory conditions (1 atm, 25°C). The pigment ratio was increased by 0.2 grams for each coating surface and three different coatings were created.

In order to create homogenous coating mixtures, the coating materials were mixed for 15 min in ultrasonic blenders (Qsonica Q500). The mixtures were coated on to glass lamellae with the dimensions of 76 mm \times 26 mm \times 1 mm, which were cleaned before coating with 10% dilution sulphuric acid and then with deionised water. The coating materials were applied to the surfaces with three different thicknesses with a Dr. Blade. A laboratory designed equipment shown in Figure 1 was used to create homogenous surfaces by moving the Dr. Blade with a constant speed on the glass surface. 60, 90 and 120 μm homogeneously coated surfaces were created in this process.

In order to determine the surface tension energy, the contact angle method was used. The stove-dried coated surfaces which had different concentrations and thicknesses

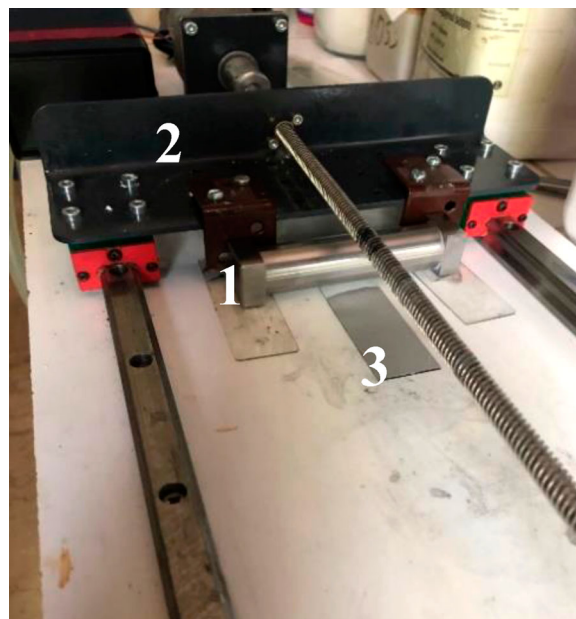


Figure 1. Surface coating equipment with Dr. Blade: 1. Dr. Blade, 2. Constant speed equipment and 3. Coated surface.

were subjected to 10 μL distilled water, paraffin and glycerol as described previously.²³

In Figure 2, an in-house permeability measurement device is shown. The device takes measurements from 180 points, with four different 4.5 mW laser diodes (ThorLabs CPS450, CPS532, CPS670F, CPS980, Dachau, Germany) with the wavelengths of 450, 532, 670 and 980 nm. The device moves the coated surface with a constant speed in between the laser diode and the light receptor with the receptor recording the power data. The amount of power passed through the coated surface is recorded for 180 points behind the surface and the arithmetic mean is taken as permeability amount.

Figure 3 shows the reflection measurement equipment designed in the laboratory. The 4.5 mW laser beams with 450, 532, 670 and 980 nm wavelengths, were pointed to coated surfaces with a 45° angle and the measurements were recorded through reflection from the surface. The lasers are fixed in space as shown in Figure 3, where

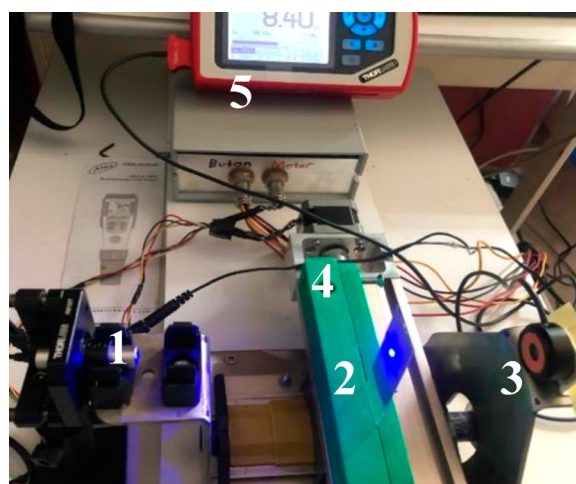


Figure 2. Permeability measurement device: 1. Laser diode, 2. Coated surface, 3. Light receptor, 4. Coated surface moving device and 5. Light power recording device.

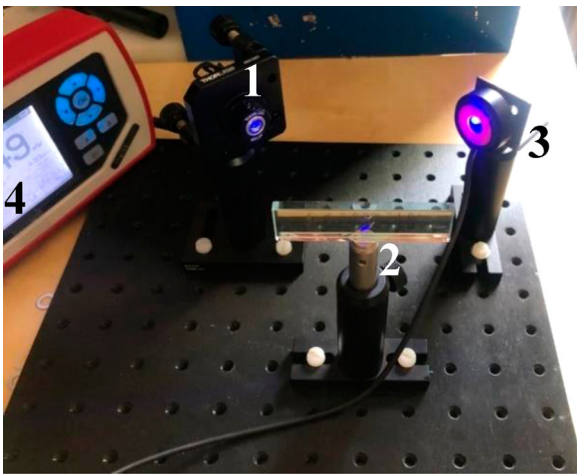


Figure 3. Reflection measurement device: 1. Laser diode, 2. Optical mirror, 3. Light receptor and 4. Light power recording device.

optimal positions were found with an optical mirror. During the study, measurements were repeated for three times and the mean values were used.

Another property of metallic coatings is the conductivity. During this study, a four-point conductivity measurement device was designed in the laboratory. Measurements were done under 1.2 kV and current was measured to determine the conductivity. To prevent corrosion affecting the measurement results, the coated surfaces were dried in an oven at 275°C and the measurements were recorded directly, immediately after the surfaces were removed from the oven. Figure 4 shows the conductivity measurement equipment with four-point detection needle probes.

Aluminium, silver, copper and graphite coated surfaces were subjected to permeability, reflection, conductivity, surface tension energy and sound transmission loss (STL) tests. For the mathematical modelling, thickness and pigment concentration were also considered as variables as well as the pigment type. For this study, the RGB colour codes were used as proxy for the pigment type as the coated surfaces had different colours. The RGB values were determined with the Adobe Photoshop Program's colour sampler tool using ten sample points and their average value. The images were taken in the laboratory under the same conditions with a fixed angle. ΔE_{RGB} values, which were used as the pigment type selector in the mathematical modelling, were calculated using black colour (0:0:0) as the vector origin point.

Sound transmission loss (STL) level of the coatings was measured by a method called amplitude modulation,²⁴ where the intensity of the laser beam is altered with the sound signal.²⁵ Laser beams (ThorLabs CPS450, CPS532, CPS670F, CPS980, Dachau, Germany) were modulated with a UNI-T UTG 9010C function generator creating sinusoidal signals with an amplitude of 80 dB (dB(A)) at 10, 20, 50, 100, 300, 500, 1000, 4000, 10,000, 16,000 and 20,000 Hertz (Hz) frequencies. The aim is to identify the effect of the wavelength on the sound transmission loss. The mean of the measured STL percentage was used for determining the sound transmission loss and the error was estimated with the eleven different frequencies. Each signal was sent through the coated surfaces and the transmitted signal was measured with a photodetector in which sound wave and the laser beam were separated with an in-house designed

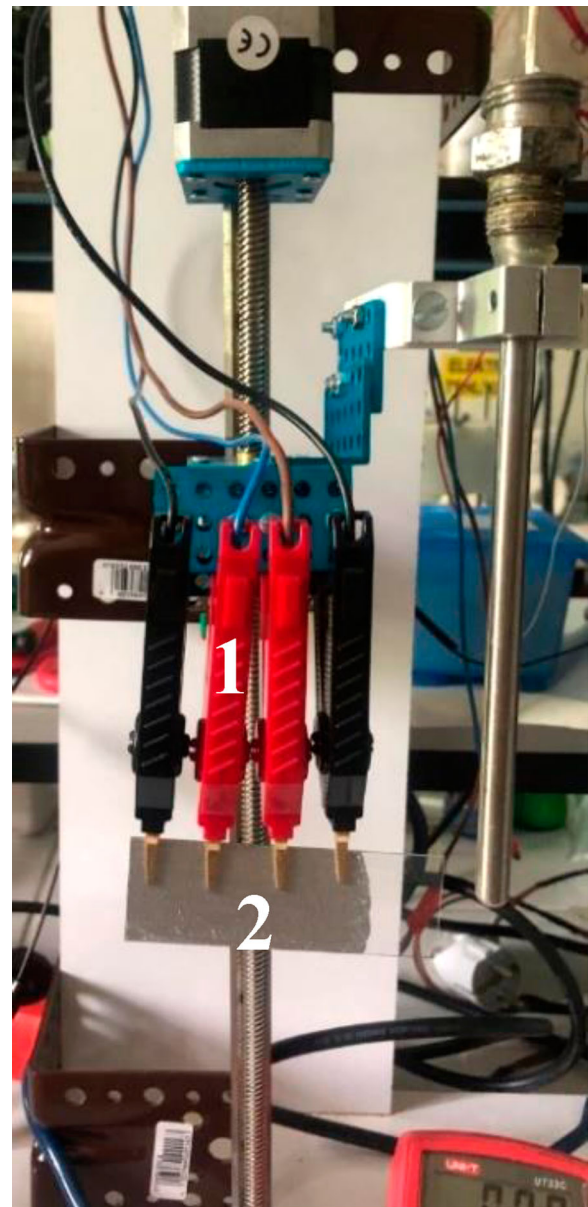


Figure 4. Conductivity measurement device: 1. Four-point probes and 2. Coated surface.

electric circuit. For each case, sound wave was then sent to a speaker with a decibel meter (Cem DT-8850) measuring the corresponding sound transmission loss. A photographic image of the experimental equipment is depicted in Figure 5.

Reflection, conductivity, surface tension energy and RGB pigment determination experiments were repeated three times with each surface, and the corresponding mean values were used for the modelling and the rest of the data sheet.

Results and discussion

In this study, surface tension energy, concentration of the pigment, permeability, reflectivity, thickness of the coating and conductivity values of the coatings were used to determine the relationship between sound transmission loss (STL) properties of silver, copper, graphite and two different sized particle aluminium coatings prepared in the laboratory. Each experiment was done at least three times to make sure the measurements were repeatable. For the STL level, the standard deviation (SD) values are between 0 and 0.098,

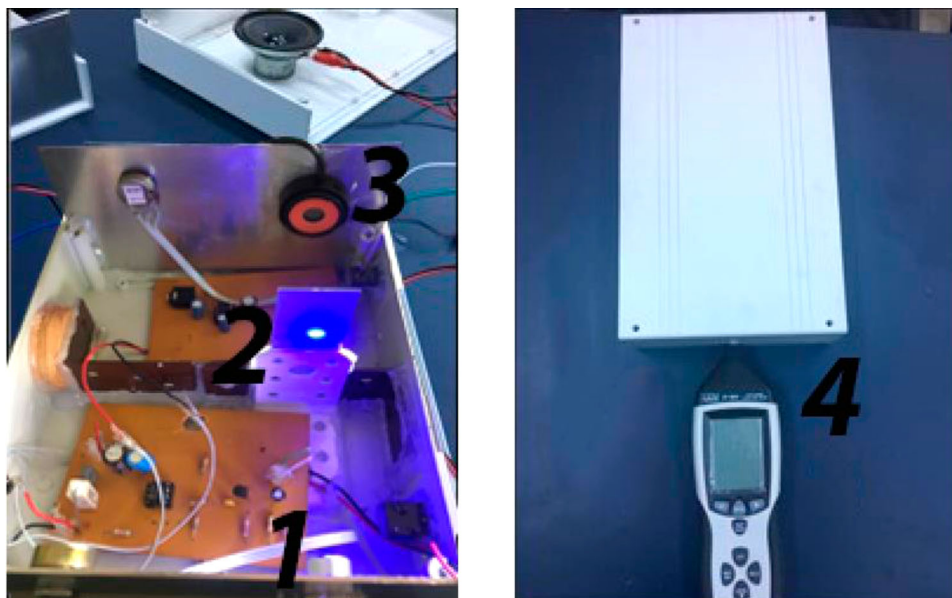


Figure 5. Image of the STL measurement device: 1. Laser beam device, 2. Coated Surface, 3. Photodetector and 4. Decibel meter.

where the coatings were subjected to eleven different frequencies of sound waves. For the rest of the experiments the SD values change between 0.012 and 0.108 with an acceptable repeatability value for the experiments. In addition, for each kind, and each particle size, of material, three different concentrations of coatings were prepared and applied with three different thicknesses on to the glass surfaces. The measurements are given in mean and SD values for each kind of material.

The surface tension energy values were calculated using a MATLAB program where the Fowkes Method, OWRK/Owens-Wendt Method, Wu Equation and Neumann's Method²⁶ are integrated. The mean value of the surface tension energy (STE) was taken for each coated surface. The results show that aluminium with 22 μm APS has the highest STE level. Aluminium with 12 μm APS has the second highest STE value, close to the bigger particle sized aluminium. These are followed by silver, graphite and copper respectively. Figure 6 shows the mean values of STE levels.

With the 450, 532 and 980 nm wavelength laser beams the reflection property of the coated surfaces were determined as can be seen in Figure 7 with standard deviations (SD) marked on the graph. The most reflective surfaces were found to be the ones coated with aluminium with an APS of 12 μm . The 22 μm APS aluminium coated surfaces was second with little difference for all three beams. For the reflection property in general silver, copper and graphite are broadly equal with only one interesting exception. Graphite coated surfaces are slightly more reflective in non-visible light than copper coated surfaces.

The permeability properties of the surfaces were also determined in this study and the mean values of nine different surfaces for each kind of material is given in Figure 8 with the SD values marked. In the case of permeability the effect of the light wavelength is more significant. For the green beam (532 nm) the highest permeable surfaces are copper ones where the lowest ones are the aluminium 12 μm , whereas for non-visible light the highest

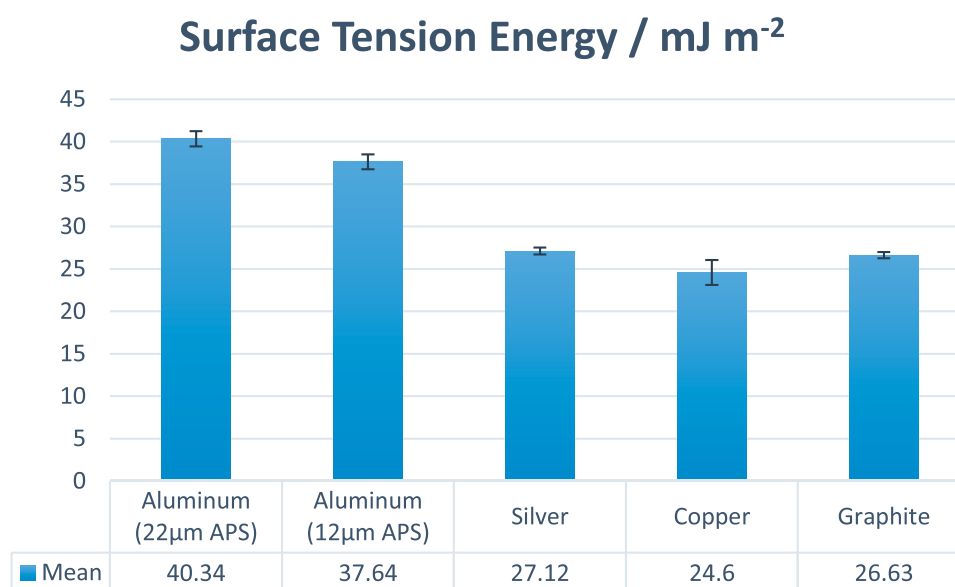


Figure 6. Surface tension energy of different coatings.



Figure 7. Reflectivity of different coatings.

permeable surfaces are the silver ones and the lowest ones are graphite surfaces. Also, for the blue beam (450 nm) permeability of copper surfaces is the highest and that for graphite surfaces is the lowest.

The aim of this study was to determine the sound transmission loss (STL) property of the surfaces which are given in Figure 9 with the SD values marked. The highest STL level can be obtained with non-visible spectrum (980 nm) laser, blue beam (450 nm) and green beam (532 nm), respectively in the order mentioned. For comparison purposes, 670 nm wavelength data are added to the STL graph from Esen Ergin *et al.*²⁵ whose investigation used only the red laser beam (670 nm). STL values of red beam are second highest for graphite and silver and third highest for aluminium 12 μm APS and copper. For the material type of the

surface the highest STL level was achieved with aluminium with 22 μm APS. Also, on average, aluminium with 12 μm APS surfaces have the second highest level of STL except with the 980 nm beam. Copper surfaces have the second highest STL level with the 980 nm beam, whereas in general they have the third highest.

Esen Ergin *et al.* in 2020²⁵ studied the STL values with 670 nm (ThorLabs CPS670F, Dachau, Germany) laser beam with the same experimental equipment. The average results of STL values for coloured paints were between 4.5 and 6.6 where green coloured paint has the highest and yellow coloured paint has the lowest STL values. In this study the material with the highest STL value, 7.77%, in 670 nm is aluminium (22 μm APS). Green and orange coated surfaces have a higher STL value than silver, copper and graphite

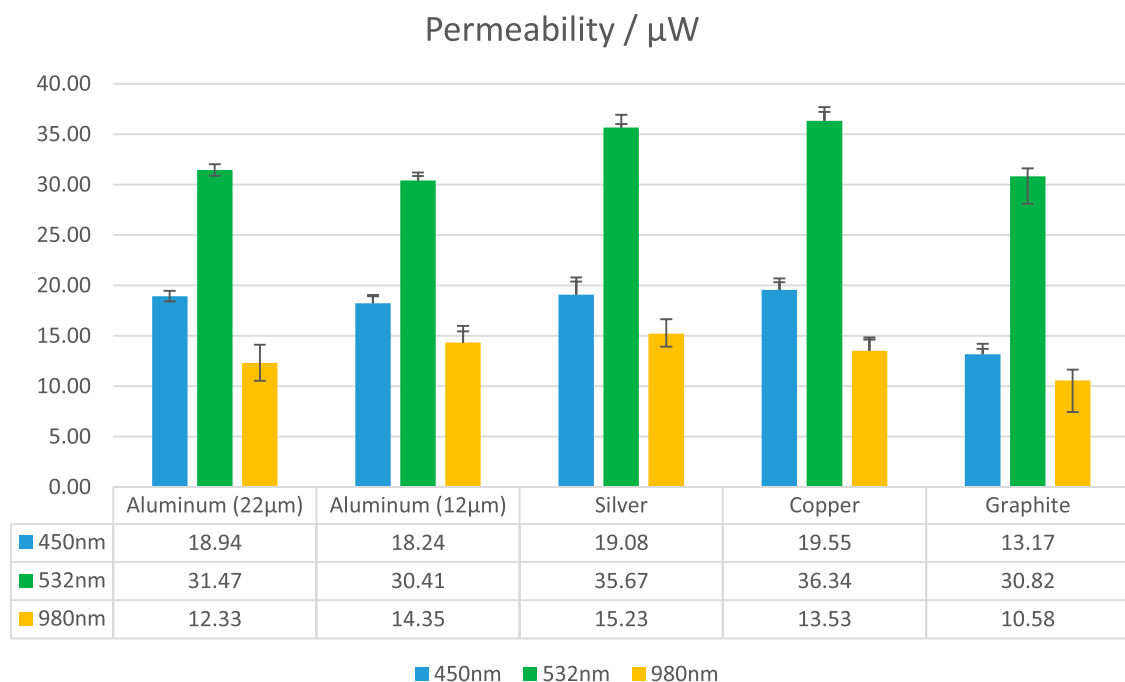


Figure 8. Permeability of different coatings.

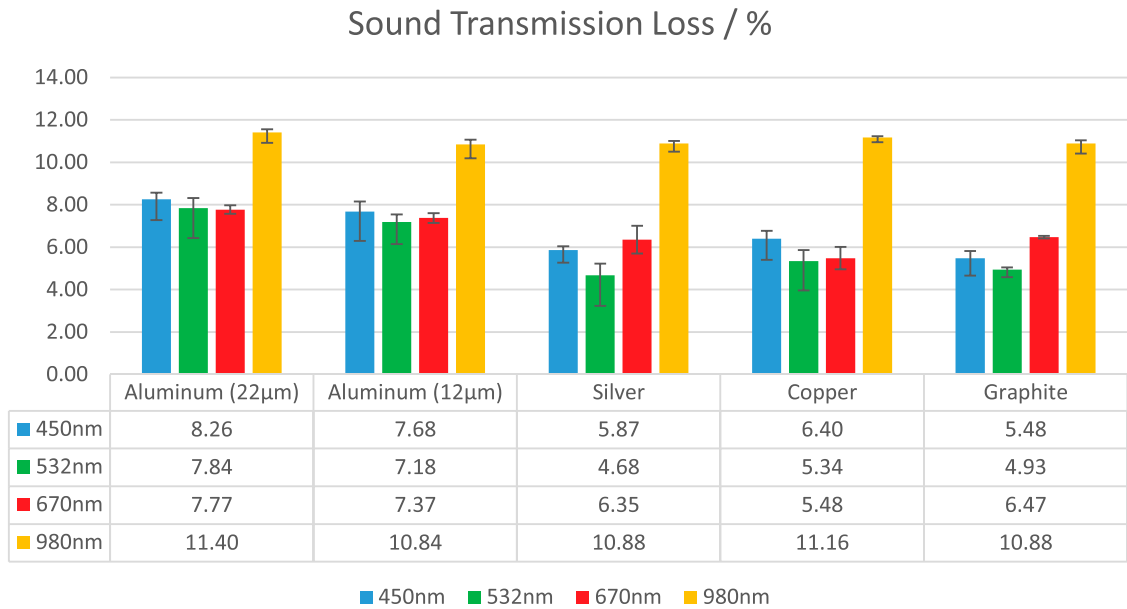


Figure 9. STL in different wavelengths.

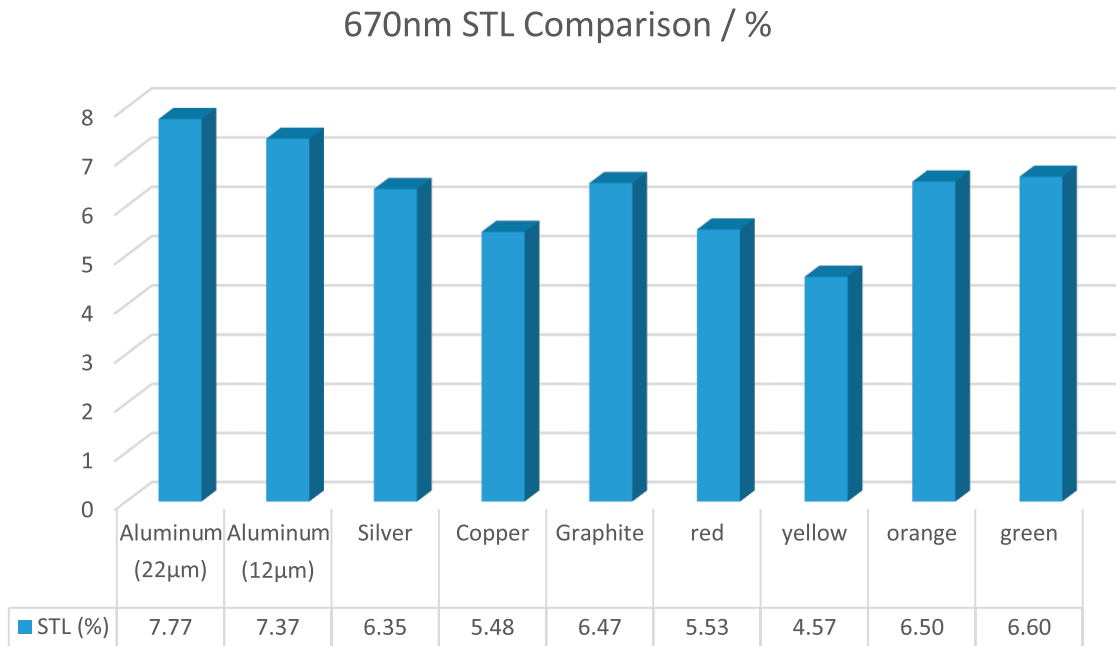


Figure 10. STL comparison with literature.

coated surfaces where red and yellow coated surfaces have lower STL values in average. The comparison of STL values in 670 nm are given in Figure 10.

The relationship between sound transmission loss (STL) and surface tension energy (STE), conductivity (K), pigment type (ΔE_{RGB}), pigment concentration (C), permeability (P), reflectance (R) and thickness of coating (T) was studied with multivariable linear regression using SPSS 25 program (Statistical Package for the Social Sciences). R^2 values for

multilinear regression and P values were determined for the different laser beams. The highest R^2 value and the highest number of significant variables (P values lower than 0.05) were found to be in the 980 nm laser beam equation. In the 980 nm beam all variables except the thickness of coating (T) are significant in the equation. The R^2 values are given at the end of Table 1 and variables with P values lower than 0.05 are marked with 'signif.'. Table 1 shows that surface tension energy (STE), pigment concentration (C) and

Table 1. SPSS results for different wavelengths.

Wavelength	Surface tension energy/mjm ⁻²	Conductivity/Sm ⁻¹	Pigment type/ ΔE_{RGB}	Pigment concentration/%	Permeability/ μW	Reflectance/ μW	Thickness of coating/ μm	R^2
980 nm	Signif.	Signif.	Signif.	Signif.	Signif.	Signif.	—	96.2
532 nm	Signif.	—	—	Signif.	—	Signif.	—	93.1
450 nm	Signif.	—	—	Signif.	—	Signif.	—	91.8

Table 2. Sound transmission loss in 980 nm (STL), surface tension energy (STE), conductivity (K), pigment type (ΔE_{RGB}), pigment concentration (C), permeability (P), reflectance (R) and thickness of coating (T).

980 nm Sound transmission loss (STL)	Surface tension energy/mjm ⁻²	Conductivity/Sm ⁻¹	Pigment type/ ΔE_{RGB}	Pigment concentration/%	Permeability/ μW	Reflectance/ μW	Thickness of coating/ μm
0.115	42.5	0.14931	382.9112	90	15.20	88.11	60
0.114	37.9	0.10580	352.9773	90	11.18	99.89	90
0.113	37.8	0.10548	369.3684	90	10.40	98.51	120
0.114	39.0	0.11275	369.7596	87.5	12.08	97.53	60
0.111	43.9	0.11714	370.9609	87.5	15.10	98.97	90
0.113	39.7	0.07455	365.3026	87.5	10.98	89.74	120
0.115	44.1	0.09056	388.2293	83.33	11.97	86.67	60
0.115	44.6	0.07581	369.0016	83.33	13.11	81.43	90
0.115	40.3	0.09343	365.3026	83.33	10.90	80.20	120
0.112	38.7	0.03424	385.5451	90	11.74	93.08	60
0.111	41.1	0.00607	370.8719	90	13.04	97.42	90
0.109	35.2	0.02293	356.6076	90	12.01	97.49	120
0.107	36.9	0.00332	394.5238	87.5	15.00	97.14	60
0.108	37.6	0.03963	380.5903	87.5	15.30	91.25	90
0.109	39.8	0.05258	362.7671	87.5	15.65	90.07	120
0.108	35.7	0.04179	385.571	83.33	15.13	100.59	60
0.107	35.9	0.02458	387.697	83.33	15.34	102.29	90
0.105	35.9	0.02748	363.0344	83.33	16.01	95.00	120
0.109	28.6	0.16061	368.4277	90	17.60	68.02	60
0.109	27.8	0.09789	359.1852	90	17.21	59.69	90
0.109	26.4	0.11236	366.0724	90	17.49	61.04	120
0.109	26.6	0.10157	396.178	87.5	15.00	59.69	60
0.109	26.7	0.13718	397.6355	87.5	16.35	59.97	90
0.107	27.9	0.11124	380.4261	87.5	18.24	63.21	120
0.107	26.2	0.11581	353.6015	83.33	17.21	61.12	60
0.109	27.2	0.08784	341.4293	83.33	15.20	60.12	90
0.110	26.7	0.10579	377.3486	83.33	13.35	59.44	120
0.111	17.9	0.15967	401.9577	90	13.02	54.65	60
0.112	26.9	0.08518	408.8838	90	13.59	57.08	90
0.112	21.7	0.11086	398.5398	90	12.19	56.03	120
0.111	25.7	0.08596	391.7716	87.5	14.65	51.30	60
0.111	26.9	0.14489	363.0937	87.5	15.34	55.90	90
0.113	26.1	0.13424	362.9449	87.5	14.74	49.60	120
0.111	25.4	0.12915	361.0249	83.33	12.92	58.99	60
0.112	25.6	0.09213	332.9565	83.33	12.61	55.90	90
0.112	25.0	0.10921	343.9724	83.33	12.74	54.98	120
0.105	26.4	2.6087E-05	153.0653	90	13.94	62.01	60
0.109	27.2	0.000138	149.0168	90	10.12	57.03	90
0.109	26.8	0.000135	164.0183	90	11.72	55.64	120
0.109	27.6	0.000213	143.248	87.5	10.03	57.98	60
0.109	25.6	0.000224	125.9365	87.5	10.09	54.98	90
0.109	25.6	0.000971	114.3766	87.5	10.14	54.33	120
0.110	27.2	0.000525	120.7187	83.33	9.916	53.94	60
0.109	27.3	0.000328	106.2826	83.33	10.12	53.54	90
0.109	25.9	0.000146	108.0046	83.33	10.05	54.20	120

reflectance (R) values are significant in all the different wavelengths. All the experiments in this study were conducted with four different amplitude modulated laser beams (Thor-Labs CPS450, CPS532, CPS670F, CPS980, Dachau, Germany), however only the 450, 532 and 980 nm results are given, except for the STL values, because the R² value of 670 nm is around 82.5 percent, whereas the rest are over 91 percent.

The data recorded with 980 nm wavelength laser beam are given in Table 2 and Table 3.

The mathematical equation created using multiple linear regression with all the variables affecting the sound transmission loss level (STL) is given in Equation (1).

$$\begin{aligned} \text{STL} = & 0.105071 + 0.000388 * \text{STE} + 0.02538 * \text{K} \\ & + 1.14\text{E}^{-05} * \Delta E_{RGB} \\ & + 9.42\text{E}^{-05} * \text{C} - 0.00088 * \text{P} - 0.00012 * \text{R} - 4.5\text{E}^{-08} * \text{T} \quad (1) \end{aligned}$$

In summary, the surface tension energy, reflectance and pigment concentration are the main parameters affecting the sound transmission loss level in all three wavelengths. The data and the equation show us all three are significant parameters in determining the STL, with STE having a higher effect. On the other hand, thickness of the coating is

not a significant value in all the different wavelengths. In further studies, thickness of the coating parameter may be removed to simplify the data and the experiment itself. For aluminium, it is possible to say the increasing average particle size creates increased roughness in the coated surface,²⁷ which results in higher sound transmission loss (7.77% for 22 μm APS and 7.37% for 12 μm APS); further studies are needed to explain this relationship for silver, copper and graphite. However, with the same particle size aluminium is more effective in sound transmission loss than copper (12 μm APS). This shows the pigment type is also an important parameter in STL. In comparison to polyurethane-based coatings studied by Ergin Esen *et al.*²⁵ the STL values

Table 3. SPSS Results 980 nm.

	B	Std. error	p-value	Significance
Constant	0.105071	0.003944	9.61E-26	
STE	0.000388	3.74E-05	1.65E-12	Significant
K	0.02538	0.003817	8.34E-08	Significant
ΔE_{RGB}	1.14E-05	2.48E-06	4.58E-05	Significant
C	9.42E-05	4.38E-05	0.037967	Significant
P	-0.00088	6.33E-05	3.42E-16	Significant
R	-0.00012	1.61E-05	3.86E-09	Significant
T	-4.5E-08	4.82E-06	0.992517	

are higher for metallic-based coatings on average. Furthermore, the highest STL value can be reached with the aluminium-based coatings within the scope of this study.

Conclusions

This study shows that the sound transmission loss (STL) is related to the wavelength of the laser beam used. The mathematical model created using the results of the experimental study, which was carried out using an in-house experimental set-up and equipment, reveals that when a 980 nm wavelength (non-visible) laser beam is used, surface tension energy (STE), conductivity (K), pigment type (ΔE_{RGB}), pigment concentration (C), permeability (P) and reflectance (R) are significant variables in the equation. The only variable not significant is the thickness of the coating. However, in 450 and 532 nm wavelengths, there are only three significant variables, in addition to a smaller R^2 .

A parallel study conducted in 2020,²⁸ shows the STL values for different coloured polyurethane-based surfaces. This extends the study of Esen Ergin *et al.*²⁵ with different metallic and graphite-based coatings. The results show that the metallic-based coatings have higher STL values overall although green and orange coloured surfaces have slightly higher STL than silver, copper and graphite-based coated surfaces in a 670 nm laser beam. In this study, three different laser beams have also been added to the system; for a 980 nm laser beam the STL values are the highest in all different coated surfaces.

During this study, a new mathematical equation has been created to help measuring sound transmission loss percentage where the sound signal is carried with a laser beam with the help of an in-house experimental set-up to examine the performances of different coating materials. The research team is also trying to determine the relationship between carbon, graphite and graphene as an extension of this study.

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Disclosure statement

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