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Pre-impregnated natural fibre-thermoplastic composite tape manufacture using a novel process

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Abstract

Pre-impregnated flax and thermoplastic poly(amide) composite tapes have been produced using a novel process. The manufacturing method uses an impregnation unit with a siphon system to impregnate continuous flax yarns with the polymer in the form of a slurry. After water evaporation, the powder is sintered and the coated yarns are compressed by passing them through a pair of heated rollers. Using a parametric study of the process, tape quality has been assured using the key outcome criteria of tensile strength/stiffness, surface roughness, fibre weight fraction, width and thickness. The temperature of the air heater placed before the roller has the biggest influence on tape quality. A heating model was developed using finite element software LS-DYNA. The research novelty comes from producing composite tapes with good tensile properties and surface finish using aligned *natural* fibres; the feasibility of automated tape placement and winding has also been demonstrated.

Keywords: A. Natural fibres; B. Physical properties; C. Statistical methods; (Not in list): Tape consolidation.

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1. Introduction

An increased awareness of the environmental impact of non-recyclable and non-biodegradable plastics and other materials during composites manufacturing and their end-of-life disposal has led to strong interest in finding nature-based alternatives to synthetic materials. Composites that utilise natural fibres, which are derived from plant materials, and recyclable or biodegradable polymeric matrices make both manufacture and disposal more environment friendly. At present, the use of natural fibre composites is mainly restricted to non-structural components due to limitations in mechanical properties. The typical performance of a natural fibre composite is inferior in comparison to those of synthetic composites. However, the specific properties of some natural fibres are close to or even better than those of E-glass fibres, which indicates that their composites' performance can be improved. The primary focus of this paper is to demonstrate a new manufacturing process to fully realise the potential of natural fibre composites in the form of thermoplastic impregnated tapes. The challenge with unidirectional natural fibre composites is to align the fibres in one direction, as the fibres are only available in a discontinuous form. While the manual alignment of individual fibres is feasible on the laboratory scale, it would be difficult and time consuming to use this method to produce even moderate-sized real parts on a commercial basis. Arranging the fibres into a yarn holds individual fibres together by applying twist. This overcomes some of the difficulties of aligning discontinuous natural fibres within a composite, although the twist means that the fibres will not be straight [1]. Thus the strength and stiffness of the composite will be lower than those in the case of continuous, fully aligned fibres.

In general, the strength and stiffness of a composite increase as the fibre content increases. This occurs until a maximum value is reached, at which point the matrix can no longer properly wet out the fibres, and both strength and stiffness begin to decrease [2]. If natural fibres are to be used in structural applications, more emphasis needs to be placed on producing composites with a fibre volume fraction between 0.5 and 0.7. In areas where high quality composites are required, pre-impregnated ('prepreg') materials are generally used. They allow high quality composite parts to be produced, with low void content and high fibre volume fractions. Creating natural fibre composites using prepregging technologies should therefore enable parts to be produced with the same advantages. One elementary form of a prepreg material is thermoplastic composite tape. It consists of aligned fibres embedded in a thermoplastic

polymer and is typically available in widths ranging from 5 to 300 mm. Tapes are mainly produced by powder or melt impregnation [3-7], using synthetic materials.

Fig. 1 shows the tensile strengths and moduli of natural fibre composites, as found from a review of the literature [1,2,8-42]. Composites which use biodegradable polymeric matrices are indicated by black markers while those using other types of polymers are represented in grey. It can be seen that the majority of natural fibre composites have tensile strengths and moduli below 150 MPa and 20 GPa, respectively. Some of the best performing biodegradable polyester based natural fibre composites have been achieved with poly(lactic acid), PLA [18]. There are very few cases reported in the literature where natural fibre composites have greater than 300 MPa tensile strength and 25 GPa tensile stiffness. Among these, Hepworth et al. [17] achieved a tensile strength and stiffness of 378 MPa and 26 GPa, respectively, using bundles of combed decorticated flax fibres that were pre-treated with a 50% mixture of poly(vinyl alcohol), PVA, and water. A very high final fibre volume fraction of 0.8 was claimed to have been achieved. Ochi [35] tested Manila hemp and other natural fibres, to make composites using a starch-based emulsion resin. The best tensile properties of 365 MPa strength and 29 GPa stiffness were obtained at the highest fibre volume fraction of 0.7. Madsen [2] achieved properties of up to 320.7 MPa tensile strength and 28.7 GPa tensile stiffness with flax and poly(propylene). In this case, flax yarn was wound onto metal frames, producing fibre assemblies with high yarn alignment and controlled uniform thickness. Poly(propylene) matrix foils were then combined with the fibre assemblies using a film stacking technique and the entire composite stack was then processed by heating and vacuum consolidation. These results indicate that with the right materials, composite structure and manufacturing techniques, high mechanical properties can indeed be achieved with natural fibres.

With respect to the desired composite structure and its manufacturability, a natural fibre-thermoplastic tape manufacturing process would have a number of advantages. The tape making process can be semi or fully continuous, improving process speed and the ease of scalability for large volume production. Producing a tape material as a precursor would allow fibres in the final product to be highly aligned in the direction of loading, enabling the production of a well-designed and efficient part. The flexibility and width-wise scalability of a tape manufacturing process would also allow the same process to be used to manufacture unidirectional sheets, which are in effect wide tapes. The work presented in this paper details the development of a continuous natural fibre-thermoplastic composite tape manufacturing process using

consolidation rollers. The process has been evaluated using the experimental design methodology proposed by Taguchi [43]. This allows the effects of many different factors on the outcome criteria to be examined by a condensed set of experiments. The manufacture of natural fibre tapes may be seen as an essential step towards improving the mechanical properties of natural fibre composites, allowing a move towards a structural range of product capabilities.

2. Manufacture of pre-impregnated natural fibre thermoplastic composite tape

This section describes the manufacturing of pre-impregnated natural fibre-thermoplastic composite tapes using an innovative processing methodology. A Belgian variety of flax fibre in the form of the smallest available 3-yarn size of 82.7 tex was selected, and supplied by Jaya Shree Textiles, India. Flax fibre has superior tensile properties (800-1,500 MPa strength and 60-80 GPa modulus) [44,45]. Each yarn was approximately 0.27 mm in diameter, while the individual fibres themselves were 20-30 μm in diameter (as per manufacturer supplied data), and had lengths of several millimetres. The yarn had a specific strength of 0.29-0.33 N/tex, and a density of 1.49 g/cm^3. Yarns spun from bleached flax fibres were selected to avoid fat, wax, pectin and other impurities, which could cause mechanical and chemical adhesion problems between fibres and the matrix. To explore the possibility of producing truly biocomposite tapes, a biodegradable polymer, poly(lactic) acid (PLA) was initially chosen as the matrix material. Polymer 4042D, an extrusion/thermoforming grade of PLA from NatureWorks LLC, USA, has a strength and stiffness of 60 MPa and 3.5 GPa, respectively (manufacturer datasheet). The PLA was dissolved in Tetrahydrofuran (THF) solvent, and a solution impregnation method was used to coat the PLA onto the flax yarn. A thermoplastic composite tape making process using continuous compression moulding of solution impregnated flax yarns was developed [46]. Fig. 2 presents some sample tapes manufactured by this technique (tensile strength of 332 MPa and stiffness of 45 GPa). However, to automate the production method, and to avoid the use of organic solvent THF, a continuous thermoplastic composite tape manufacturing process using heated rollers to consolidate powder-coated flax yarns was developed. The details are discussed in the following section.

2.1. Use of consolidation rollers

Full consolidation of the yarns normally requires compression at high pressures and elevated temperatures. However, a suitable natural fibre tape manufacturing process does not need to ensure full consolidation of the polymer into the yarns but only requires the correct volume fraction to be present because consolidation can be achieved during the latter stages of manufacturing. The PLA pellets were ground into fine powder employing a Retsch SM100 grinder and using a fine sieve, an average particle size of 80 µm was achieved. The flax fibres were then PLA coated by passing them through an aqueous slurry of the PLA powder and impregnation was achieved under heated consolidation rollers. However, due to the difficulty of getting PLA supply of small granule size and for studying the future scale-up possibilities, a poly(amide) matrix, Vestosint 2157 PA12 powder (readily available in the necessary granule size) was used for the later part of the study. Vestosint 2157 PA12 had an average particle size of 56 µm and a melting temperature of 184 °C. It had a tensile strength at yield of 43 MPa, while its density was 1.016 g/cm^3 (as per the material data sheet). It is to be noted that the process is equally applicable to both polymers when the correct powder size and processing conditions are used.

The manufacturing facility developed had five main stages – yarn feed, yarn impregnation, heating unit, consolidation unit and a haul-off unit, Fig. 3. The yarns were held on individual spools and fed through a tensioning system which applied the minimal tension necessary to prevent the spools from overspinning and tangling the yarns. Twenty-five spools were used to create a 6 mm wide tape. A powder impregnation unit utilising a siphon system [47] was adapted to coat the yarns, Fig. 4. A Vestosint 2157 PA12 powder-water slurry mixture, kept agitated using a magnetic stirrer, was pumped into the impregnation head. Flax yarns were pulled through the head inside curved channels, which created pressure to help with impregnation. It was found that a concentration of 30 g powder per 100 ml water corresponded to an initial fibre volume fraction of 0.4 - 0.5, which was the target level for this process. Fig. 5 shows an SEM image of a coated yarn, following the partial impregnation and heating stages. It can be noted that a smooth, consistent coating of sintered polymer on the surface of the yarn has been achieved. The coated yarn passed beneath two ceramic heaters (each 250 mm long, together spanning the entire length of the heating tunnel), which were used to evaporate excess water and sinter the polymer onto the yarn. A steel channel section was placed below the ceramic heaters with its open side facing downwards enclosing the coated yarn. This formed a heating tunnel which reduced temperature

fluctuations and provided a more even heating via convection. This arrangement also enabled any excess water or un-sintered polymer to fall safely onto a collection tray. A gap existed between the end of the heating tunnel and the nip point of the rollers in the consolidation section. The sintered flax yarn/PA12 would cool to below the melting temperature of PA12 in this gap, thus preventing it from forming into a tape when it reached the rollers. Adding a controllable air heater (Leister LHS 41) above the tape maintained its temperature around the melting temperature of PA12 until the nip point of the roller was reached. Heated rollers which had a number of different sized grooves to set the width of the tape (2, 4, 6, 8 and 12 mm) were used for consolidation. The rollers were heated using an oil heater to a maximum temperature of 135 °C. When tightened, the rollers applied a constant force of 25 N to the tape using a spring and lever system.

3. Experimental design and analysis of the tape manufacturing process

The various parameters of the manufacturing equipment were adjusted using an iterative development process until good quality tape could be produced and the process run for an extended period of time. The orthogonal array experimental design proposed by Taguchi [43] was then applied to the tape manufacturing process, using the parametric values determined after the iterative development as baseline factors. Rather than having to test all possible combinations, as in a full factorial design, the Taguchi fractional factorial method tests pairs of combinations. On the basis of experience gained during the development of the process, five main factors (parameters) which are expected to have the largest influence on tape quality were chosen for further investigation: temperatures of ceramic heaters 1 and 2, roller oil temperature, air heater temperature and haul-off speed. The baseline values of these factors are shown as Level 1 in Table 1. Levels were taken at intervals both below and above these base levels, and are presented as Levels 2-4 (Table 1). This allowed four levels to be investigated and reveal any sinusoidal effects present, as seen during the process development stage. An L-16 orthogonal array (Table 2) was chosen, with the aim of determining the combination of parametric values that would yield the tape with the best overall quality. An L-16 array with five factors only allows the main effects, those due to the main factors, and not interactions, to be analysed. The parameters which remained unchanged throughout were pump flow rate (100 mL/min), roller force (25 N), roller speed (0.09 m/min) and powder

concentration (30%). Based on preliminary experiments [46], seven key criteria were selected for quality evaluation of the thermoplastic tape: tensile strength, tensile modulus, top and bottom surface roughness, width consistency, thickness consistency and fibre weight fraction. Tensile strength and modulus directly affect the mechanical properties of the part to be manufactured. Surface roughness is important because when parts are produced, undulations can occur if surface roughness is not consistently low [48]. Width should also be kept as constant as possible so that there is no overlap or underlap in tape laying. It is also necessary to keep close control of the volume fraction to produce tape with consistent properties. Approximately 10 m length of tape was produced at each of the 16 parametric value combinations.

3.1. Taguchi analysis

The key findings of the Taguchi analysis are presented under five sub-headings, one for each parameter (ceramic heater 1 temperature, ceramic heater 2 temperature, roller oil temperature, air heater temperature and haul-off speed). For each parameter, a main effect plot for every key outcome criteria (tensile strength, tensile modulus, top surface roughness, bottom surface roughness, width consistency, thickness consistency and fibre weight fraction) was generated. A main effect plot, Fig. 6, was created by plotting evaluation criteria against factor levels. Each data point is represented by a grey triangle and error bars are plotted for each level indicating one standard deviation above and below the mean. Following the standard procedure, the mean values of the results at each level are joined by continuous straight lines for depiction only. The generated graph shows the influence trend of the factor as it changes from one level to another. The dashed straight line indicates the global mean that covers all factor influences. When the line connecting the mean values at different levels is not equal to the global mean line, it indicates that the factor has an effect. The data points and error bars representing one standard deviation provide some idea of the relative variance between factors. The outcome criteria of tensile strength, tensile modulus, top surface roughness, bottom surface roughness and fibre weight fraction were either normally distributed or could be transformed to be normally distributed. They were analysed using the ANOVA and Tukey's tests [43]. Thickness and width could not be transformed to have a normal distribution and were hence analysed using the non-parametric techniques of the Kruskal-Wallis test and the Wilcoxon rank-sum test. The p-values were calculated to determine the strength of the evidence for a difference between levels in

each factor [43]. The signal-to-noise-ratio is also shown on the plot, and can be calculated in three ways: bigger-is-better, smaller-is-better or by specifying a target value. In this analysis, tensile strength and tensile modulus have been calculated using the 'bigger-is-better' target. Surface roughness, thickness and fibre weight fraction were calculated using the 'smaller-is-better' target. Finally, width was calculated using a target of 6 mm, the width of the mould. Four signal-to-noise ratios are shown on each plot, one for each level. Combining the main effects plot, analysis of variance and signal-to-noise ratios into one plot simplifies the analysis. The parametric influences on tape quality are described in the following paragraphs.

Ceramic heater 1 temperature: The overall best results were achieved at a temperature of 205 °C. Tape width, fibre weight fraction and tape thickness were most consistent at 205 °C, with 210 °C being the second best option. In general, both tensile strength and tensile modulus were higher at 205 °C with little variation. However, roughness values of the tape bottom surface were consistently lower at 210 °C.

Ceramic heater 2 temperature: It appears that ceramic heater 2 is best set at 210 °C. Tape width was most consistent at 210 °C, followed by 205 °C as the next best setting, while tape thickness, tensile strength and modulus showed the least variation at 210 °C. Tape bottom surface roughness was the lowest at 205 °C.

Roller oil temperature: Roller oil temperature had some effect on three outcome criteria – tensile modulus, thickness and top surface roughness. Tensile modulus and thickness values were the best at the highest temperature of 135 °C. Fig. 6 makes it clear that the thickness of the tape is the smallest at 135 °C. The most consistent thickness results are observed at the lowest and highest temperatures. Values for top surface roughness were the best and most consistent at the lowest temperature of 120 °C.

Air heater temperature: The temperature of the air heater was the factor that had the most influence on tape quality. Five outcome criteria (tensile strength, tensile modulus, top surface roughness, bottom surface roughness and thickness) showed some evidence of variation with respect to different parametric levels. The best results overall were achieved at the highest air heater temperature of 600 °C. Fig. 7(a) shows that as the temperature of the air heater increases, the tensile strength also increases. There is strong or very strong evidence of a difference between the majority of the levels. Slightly higher variations in strength are present at the highest and lowest temperatures. An increase in air heater temperature also leads to an increase in the tensile modulus, Fig. 7(b). Fig. 7(c) shows that the tape

bottom surface roughness reduces as temperature is increased, and is the lowest at 600 °C. The improvement in bottom surface roughness occurred at 500 °C, while for top surface roughness, the improvement came earlier at 400 °C. This shift in improvement can be explained through the position of the air heater, which was above the tape, making the top surface hotter than the bottom surface.

Haul-off speed: The four levels of haul-off speed that were investigated had minimal effect on the outcome criteria. The only evidence of a difference between levels occurred in the thickness, with the speed at the baseline value of 0.09 m/min producing the best quality tape, with the lowest variability.

3.2. Key criteria: output values

The quality of the tapes manufactured is evaluated in this section, based on the seven key criteria of tensile strength, tensile modulus, top and bottom surface roughness, width consistency, thickness consistency and fibre weight fraction. Each 10 m length of tape (one 10 m length for each of the 16 parametric value combinations) was cut into ten 150 mm long specimens at equal distances along its full length.

Tensile strength: Tensile tests were conducted on five specimens of each tape according to ASTM D3039 on an Instron 5567 universal testing machine, at a relative humidity of 50%. The results are presented in Fig. 8(a). The highest sample mean was 222 MPa for trial #4.

Tensile modulus: The tensile chord modulus was measured between 0.1 and 0.3% strain, again using five specimens of each tape. A maximum sample mean of 23.1 GPa was obtained with trial #4, Fig. 8(b).

Surface roughness: Surface roughness of the samples was measured using a FRT white light profilometer. The arithmetic average of 3D surface roughness, S_a, was measured on a 2 mm square section in the middle of the tape, in five equally-spaced places along each length of tape, on both the top and bottom surfaces. The results of bottom surface roughness measurements are shown as an example in Fig. 9, along with 3D renderings of the tape surface to provide a visual interpretation of the range of roughness values presented in the plots. On the whole, surface roughness was quite variable; it was not consistent along the length of the tape and may be quite specific to the small area sampled. Generally the bottom surfaces of the tapes had lower roughness than the top. There are two possible reasons for this result. First, the air heater was only blowing air on top of the yarn, which may have removed some un-

sintered powder, resulting in the exposure of gaps between yarns. Second, the haul-off unit was positioned slightly below the roller; hence the bottom surface of the tape would have spent more time in contact with the lower consolidation roller than the top. Tape from trial #1 had the lowest top surface roughness (mean value of 11.5 µm), while trial #4 produced tapes with the lowest bottom surface roughness values (mean value of 8.5 µm).

Width and thickness consistency: The widths and thicknesses of the tapes were measured on five specimens of each tape using Vernier calipers and a micrometer, respectively, at three locations on each specimen. Width measurements were quite consistent, with most results around 6 mm. However, there was a spread in thickness values, which was present for all the experiments. Trial #4 had the lowest average thickness of 0.41 mm, while trials #11 and #15 produced the most consistent thickness, with a standard deviation of 0.1 mm.

Fibre weight fraction: To calculate fibre weight fraction, 25 m lengths of raw yarn were dried for 24 hours at 70 °C in the presence of a desiccant. The average of these samples was used as the base weight for dry flax yarn. The full length of the tape produced in each trial (10 m) was measured and weighed after being dried, also for 24 hours at 70 °C. Any additional weight was presumed to be due to Vestosint PA12. Most fibre weight fraction values were fairly similar, around 0.78 (fibre volume fraction of 0.70). The values are all relatively high but within the range commonly seen in commercially available synthetic tapes.

Taguchi Analysis does not necessarily result in one set of parametric values that produces the best quality product in every respect. Engineering judgement has to be employed to determine which trial resulted in the best quality tape overall. Tapes from trial #4 (heater 1: 210 °C; heater 2: 220 °C; roller oil: 135 °C; air heater: 600 °C; and haul-off speed: 0.08 m/min) were found to have the most desirable properties on the whole, including the key properties of tensile strength and tensile modulus. However, trial #1 produced tapes with the least top surface roughness, while trials #11 and #15 had the most consistent thickness.

3.3 Thermal modelling

In order to be fit for commercialisation, the natural fibre tape manufacturing process must be fast. At

present the limiting factor of the process is the time it takes to evaporate the water and sinter the polymer powder onto the yarn. This occurs while the yarn is passing through ceramic heaters 1 and 2. To understand the heating process, a model has been developed using the finite element (FEA) software LS-DYNA. This model could be incorporated into a larger process simulation FEA model which encompasses the entire tape making process. Fig. 10 shows the model broken down into four parts. Each part was modelled using solid elements, which allowed the study of temperatures in the cross section of the yarn and could also include the study of temperatures along the length of the yarn in expanded models. The results (nodal temperatures) from each part were averaged at the shared nodes to produce an output. Each of the four parts uses the thermal isotropic phase change material model; this allowed temperature dependent isotropic properties with a phase change to be defined. The latent heat of the material (water or polymer powder) was defined together with the temperature at which the phase change occurs and which can be obtained by Differential Scanning Calorimetry analysis. Two different geometric spaces were present in the model. One was filled by water and yarn, and the other was filled by polymer powder and water. Both materials were assigned the full geometric space, but in effect will only occupy a portion of the space, depending on how much of the other material is present.

The size and proportion of materials of the inner region were calculated by assuming the yarn would absorb water while it was in the siphon impregnation unit until it was fully saturated. In this research, on average the wet yarn was 350% heavier than the dry yarn, giving a proportion of 28% fibre and 72% water by mass. Using the density of water and flax fibre, the circular diameter of the cylindrical volume required to contain the weight of flax and water present was calculated to be 0.68 mm. This was the size of the area given to the inner section of the solid elements in the model. The average fibre weight fraction of the 16 Taguchi trials was 0.75 (fibre volume fraction of 0.67). Powder lost during the process was assumed to be minimal; yarn would therefore retain the same weight fraction while it was being heated. It was also assumed that water and PA12 remained in the same ratio surrounding the yarn as they were in the original slurry. It must be noted that water content had a direct influence on the evaporation time; higher water content would obviously require more time. As expected, it was found that the convection coefficient was the most important parameter as small changes in this parameter significantly affected the heating rate. It was important to determine this directly from experiments or by using an empirical formulation and the typical temperature data from a thermocouple, Fig. 11(a), embedded in the material

as it moves through the tape making process. Water content in the flax was the second most important parameter. Water content in the powder and boundary temperature had a relatively small effect on the processability.

The processing window for natural fibre composites is relatively small. The temperature applied must be sufficient to melt the polymer, but the fibres must be kept below 200 °C to prevent degradation. On the other hand, PA12 fully melts around 190 °C; this leaves a relatively small processing window for the tape manufacture, Fig. 11(b). When leaving the heaters, the temperature of the tape should be greater than 190 °C so that the PA12 was melted, but not greater than 200 °C, to avoid fibre degradation. It is to be noted that as the boundary temperature of the evaporation and sintering heating stage increases, the processing speed is increased, but the processing window is reduced. This increases the demand on the haul-off motor to work within finer tolerances. The entire thermal modelling exercise was carried out to determine the processing window with PLA as well, Fig. 11(c), without causing any fibre degradation. The details of these analyses and their effects on the tape manufacturing are given in Ref. [46].

3.4. Optical microscopy

Tape specimens from each of the Taguchi trials were prepared for viewing under the optical microscope by mounting them in epoxy resin, then grinding and polishing using standard techniques. Sample pictures of the cross-sections of the tapes are presented in Figs. 12(a), trial #4 and 12(b), trial #1. To increase image detail, approximately five images for each trial were stitched together using image processing software. The circular cross-section of the yarn presented some challenges in producing a tape structure with rectangular edges, though this has been successfully achieved in some cases. It is noted that tapes with good mechanical properties, for example trial #4, Fig. 12(a), have yarns which are more closely compacted together and edges that are relatively smooth, forming cohesive tapes with few voids. Tapes with reduced mechanical properties were found to have more gaps between yarns, particularly at the edges; furthermore there may be areas where no resin is present in-between yarns. Yarn compaction is affected by different factors, such as compaction pressure, temperature and haul-off speed. Variation in yarn compaction is a commonly observed phenomenon in fibre composite manufacturing processes [49]. It was noted that in most samples, surface roughness was highly affected by the layout of the yarn

structure inside the tape. Surface roughness could be lowered by increasing the matrix volume fraction in some cases, although reasonably low surface roughness has been achieved in some trials, as seen in Figs. 12(a) and 12(b).

4. Manufacturing applications

Following the successful development and systematic analysis of the manufacturing process, feasibility studies demonstrating two possible subsequent processing routes were carried out. Automated tape placement and tape winding were investigated in order to evaluate the manufactured tape material's response to such highly relevant manufacturing operations.

4.1. Feasibility study of automated tape placement

A study was conducted to determine the feasibility of manufacturing parts using an automated tape placement process using natural fibre tapes. The manufacturing set-up consists of a placement head attached to a robotic arm or gantry, a motion controller with positioning software, tooling for stacking the laminate and a tape spool [48]. In this process, thermoplastic tape from a spool is passed through a guide assembly and is heated by a gas torch and placed on the tool with a temperature-controlled consolidation roller. Several tapes can be placed side by side and a laminate manufactured by placing additional layers over the previously laid-up layers. When the laminate reaches the desired thickness and dimensions, the process is terminated and tool heating switched off, allowing the laminate to cool. Sheets comprising of a four layer stack of tapes, with the width of the sheets being four times the width of a standard tape, were manufactured. The gas torch applied heat continuously throughout the sheet making process, with the maximum temperature never exceeding 220 °C. Four sheets were successfully produced, as shown in Fig. 13(a). Some thermal degradation was present in the sheets, particularly around the edges of the tapes where a reduced amount of polymer was present.

Two of the sheet specimens were subjected to tensile testing. Each sheet was cut into two specimens 25 mm wide and 70 mm long and tested according to ASTM standard D3039. The sheets had an average tensile strength of 202.8 MPa and an average tensile modulus of 16.59 GPa. The strength and stiffness

values of the sheets are comparable to those of the tapes from which they were manufactured. It is thought that some nesting and further impregnation may have occurred during the sheet making process. Sheet specimens were mounted in epoxy resin, then ground and polished using standard techniques. Fig. 13(b) shows the cross section of a sheet. It can be seen that some voids are present. The voids are more prevalent between the third and fourth layers. This area would have had only one pass with the roller, while layers one and two would have experienced up to four passes. More passes with the roller may therefore reduce the number of voids present in the part.

4.2. Feasibility study of tape winding

The feasibility of applying standard thermoplastic tape winding techniques to natural fibre tape was also investigated using tape manufactured in the study. A schematic diagram of the winding process [50] is shown in Fig. 14(a), while Fig. 14(b) shows a wound tubular part that was successfully produced using natural fibre composite tape. The tube was manufactured using a winding angle of 60°, winding speed of 2 m/min and a temperature of 220 °C at the nip point of the heated placement head. Little, if any, degradation of the tape was observed and there was strong bonding between layers. This result indicates a possible alternative heating method which could also be implemented in automated tape placement, along with the promising manufacture of tubular parts with such material.

5. Concluding remarks

A novel semi-industrial continuous composite tape manufacturing facility that uses a natural fibre, a recyclable thermoplastic polymer and online powder impregnation process has been developed, avoiding the use of organic solvents. Using this method, continuous flax/poly(amide) Vestosint PA12 tapes with a highest sample mean tensile strength of 222 MPa and tensile modulus of 23.1 GPa have been produced. As mentioned earlier, this technique can also be applied to manufacture flax/PLA tapes with PLA granules of appropriate size of 70-80 µm. The Taguchi methodology of experimental design and analysis was applied to the tape manufacturing process so that parameter settings that can improve the overall quality of the tape were determined. Automated tape placement and tape winding (see Annexure for video

demonstration) were also investigated to demonstrate the successful manufacture of sheets and cylindrical parts in two important subsequent processing routes.

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Figure Captions

Fig. 1. Tensile strength and stiffness of natural fibre composites. Black markers represent biodegradable polymeric matrices, while other matrices are shown in grey.

Fig. 2. Pre-impregnated flax/PLA composite tapes.

Fig. 3. Schematic of the natural fibre thermoplastic tape manufacturing method using consolidation rollers.

Fig. 4. Diagram of the siphon impregnation unit adapted to use a thermoplastic slurry.

Fig. 5. SEM image of yarn impregnated using powder slurry siphon impregnation.

Fig. 6. Taguchi analysis: main effect plot of roller oil temperature on tape thickness.

Fig. 7. Taguchi analysis: main effect plot of air heater temperature on tape (a) tensile strength; (b) tensile modulus; and (c) bottom surface roughness.

Fig. 8. Experimental results of tape: (a) tensile strength; and (b) tensile modulus.

Fig. 9. Experimental results of tape bottom surface roughness.

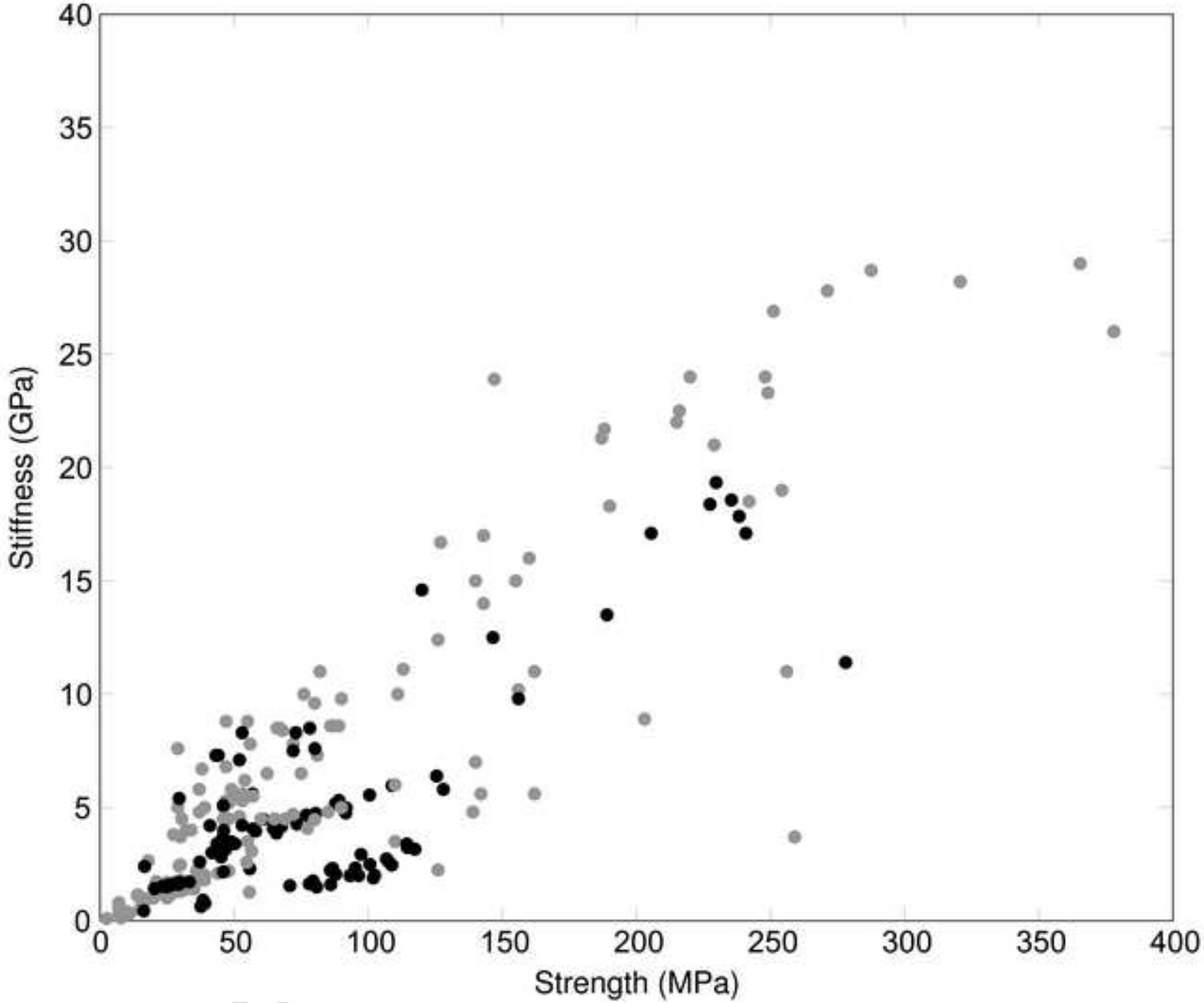
Fig. 10. The four parts of the developed LS-DYNA evaporation and sintering thermal model.

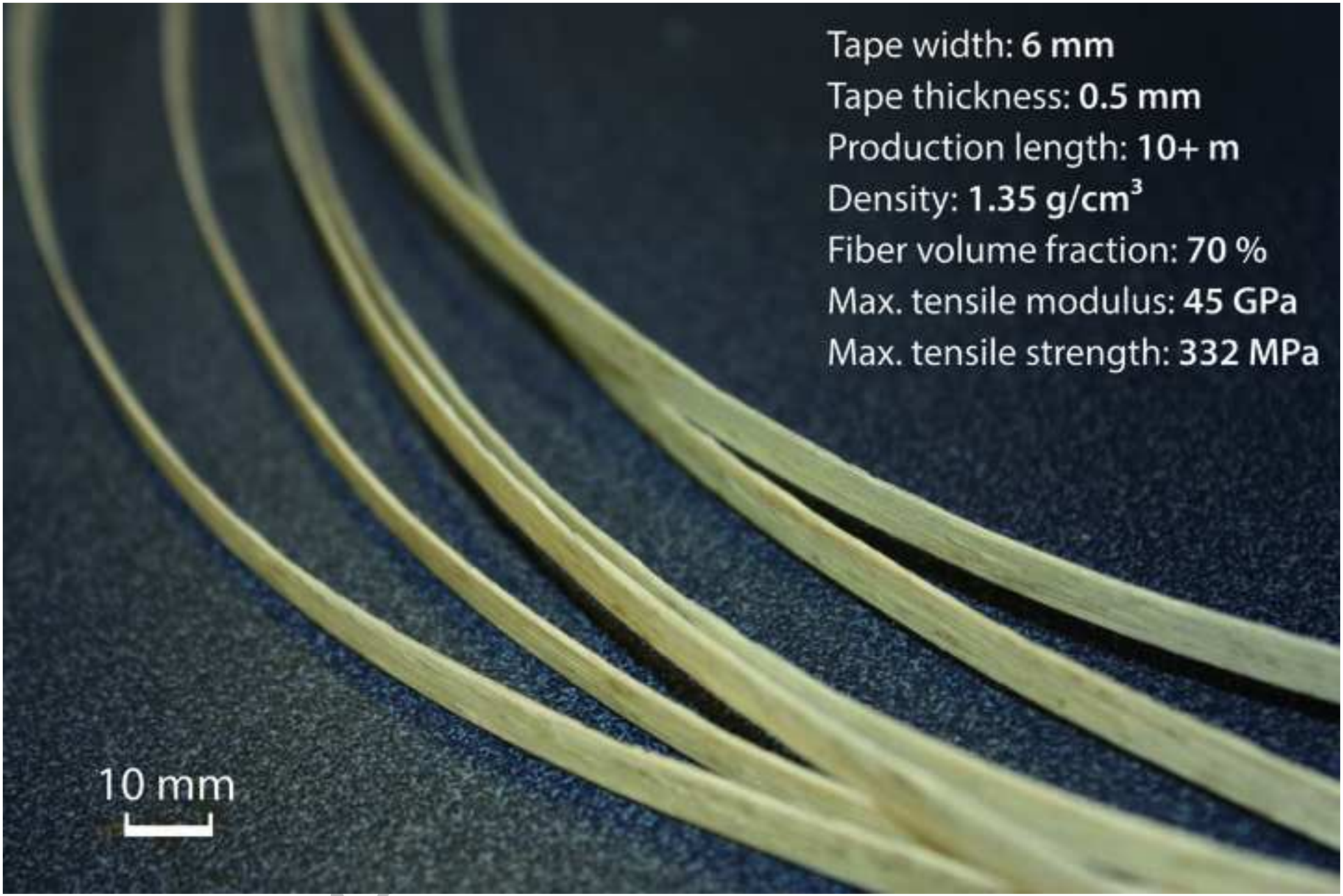
Fig. 11. (a) Comparison of simulation and experimental data at different boundary condition temperatures; (b) processing window for flax/PA12 tape at different temperatures; and (c) comparison between PA12 and PLA polymers at a boundary condition temperature of 215 °C.

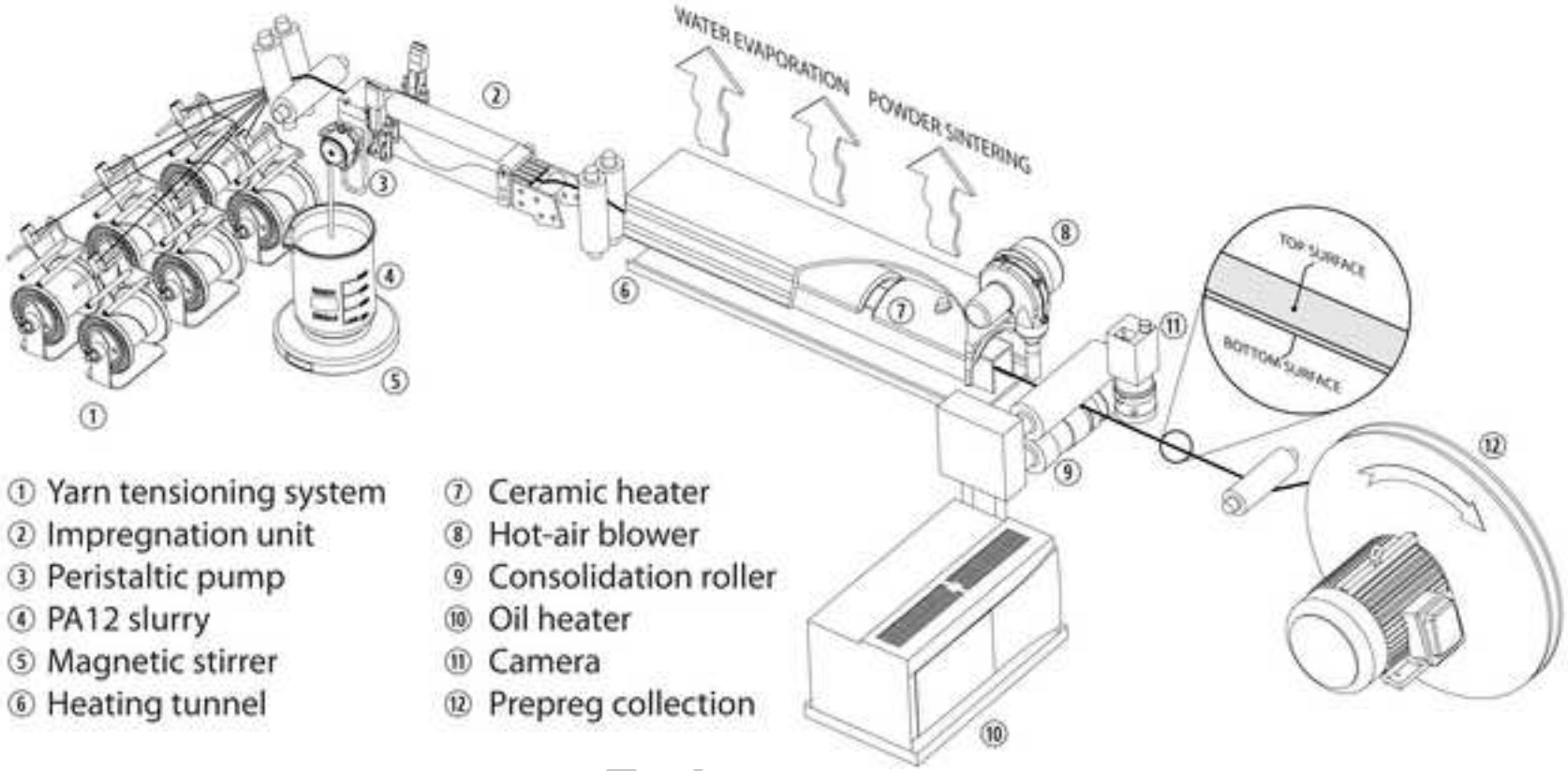
Fig. 12. Cross-section of tapes from Taguchi analysis: (a) trial #4; and (b) trial #1.

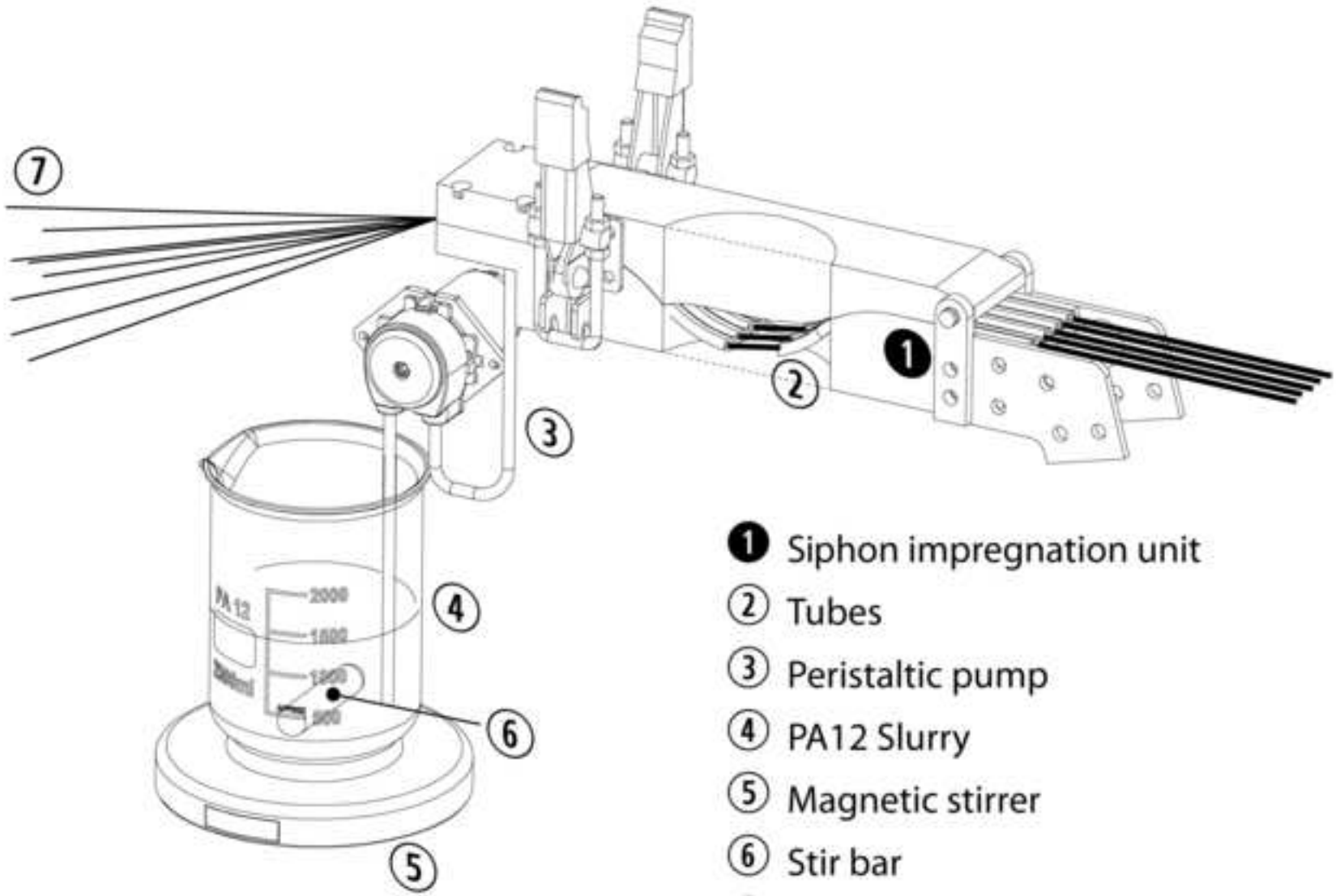
Fig. 13. (a) Sheets manufactured from flax/PLA tape using automated tape placement setup; and (b) cross-sectional view of sheets.

Fig. 14. (a) Diagram of tape winding process. Reproduced from Funck [49]; and (b) parts manufactured.

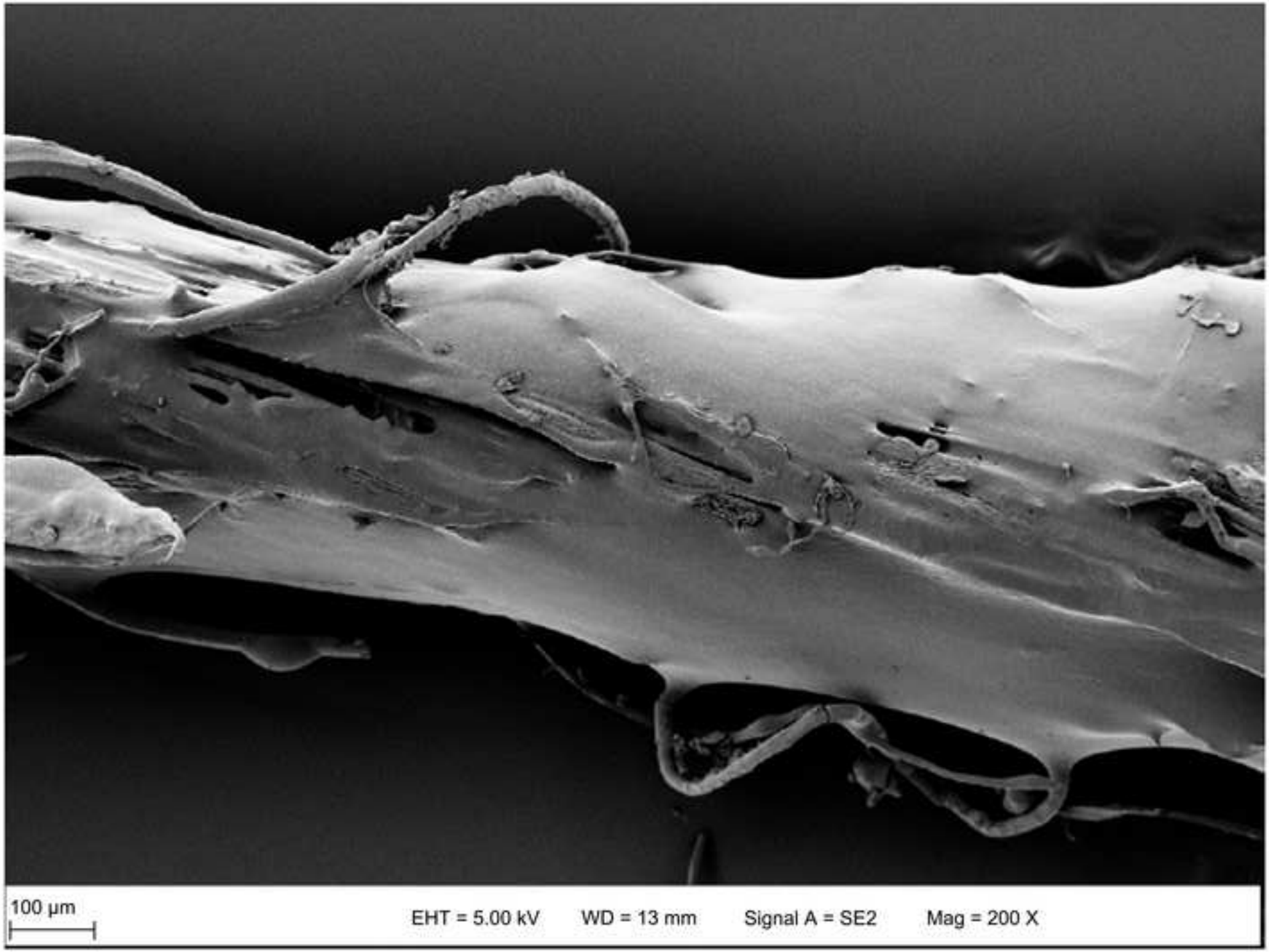


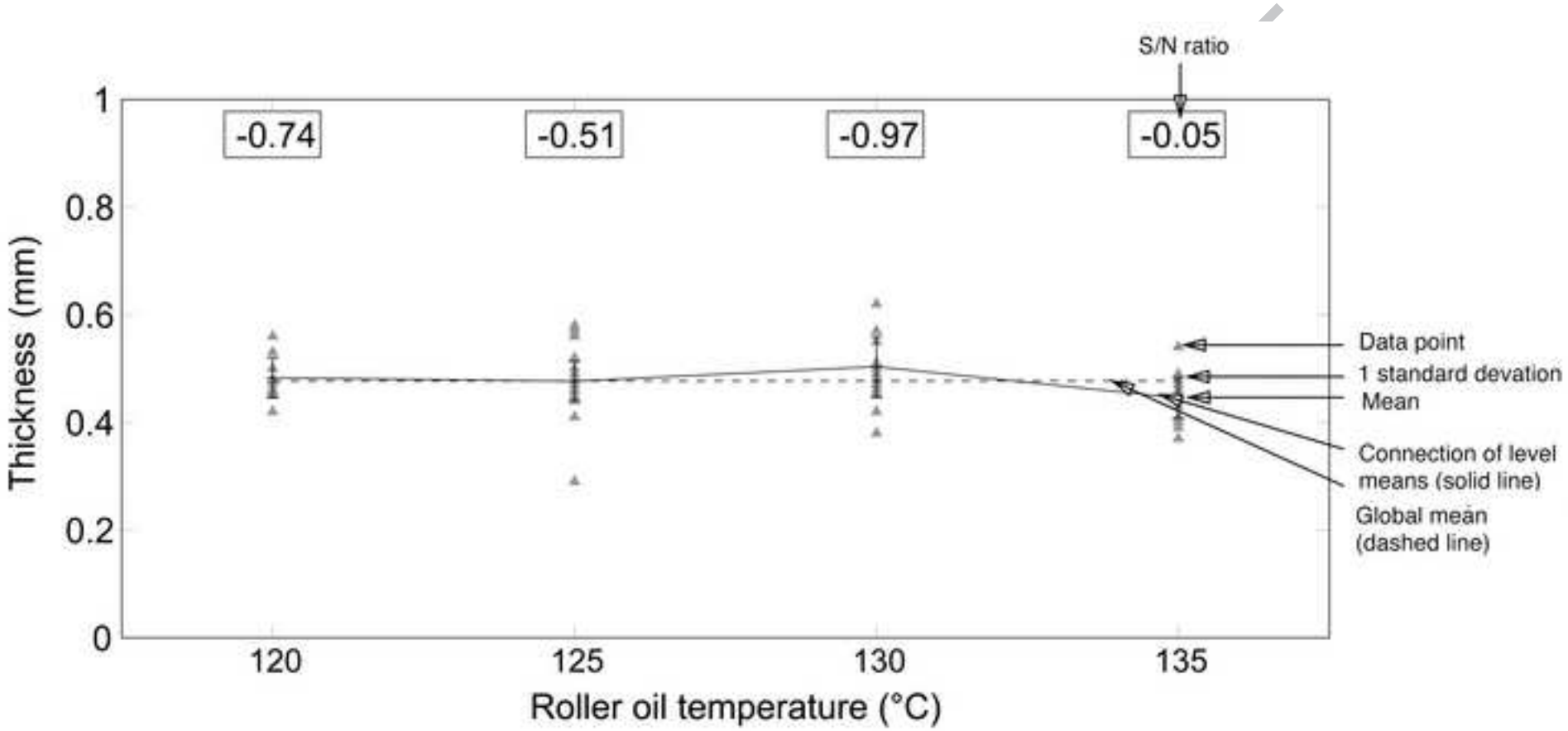


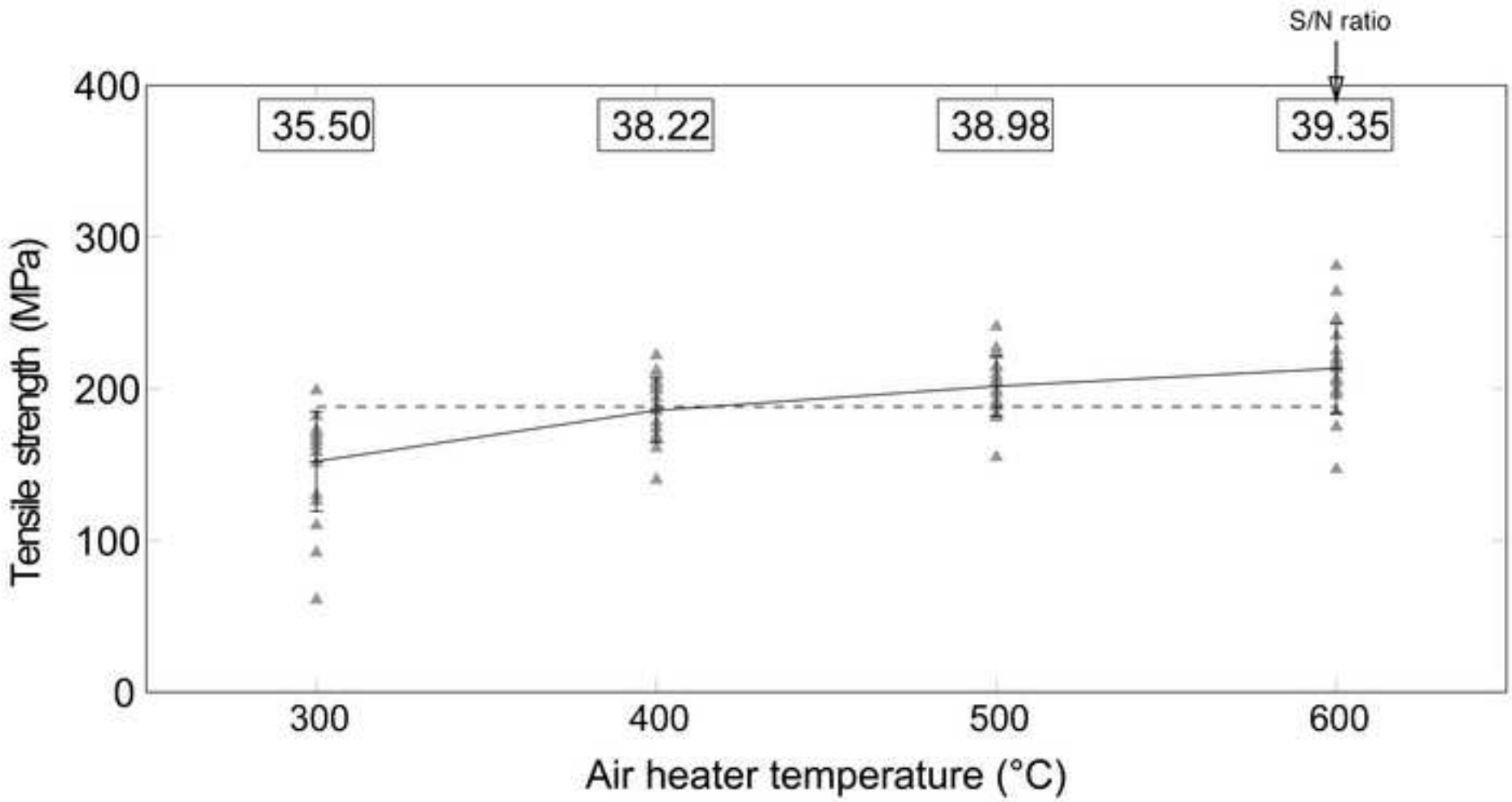


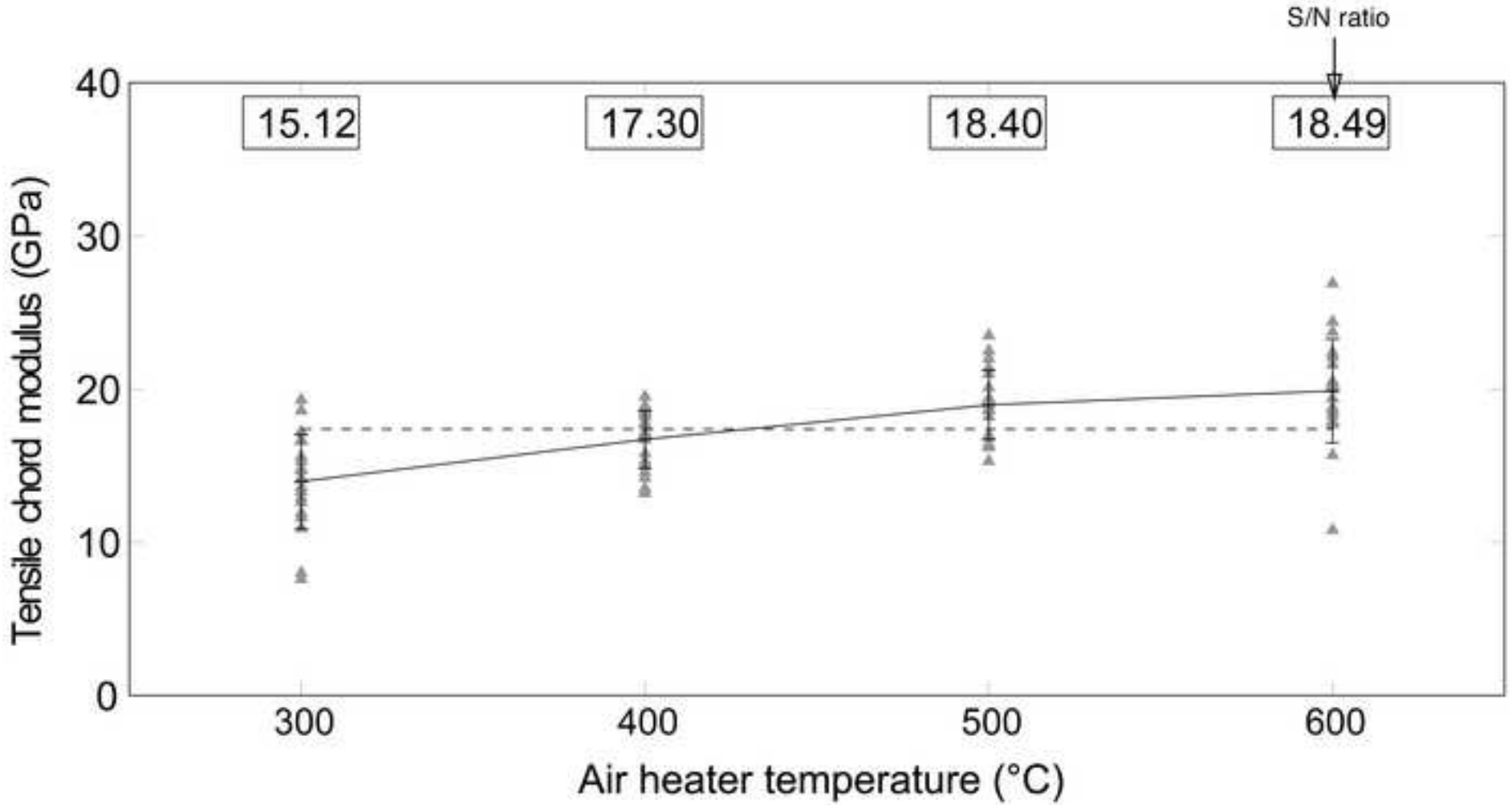


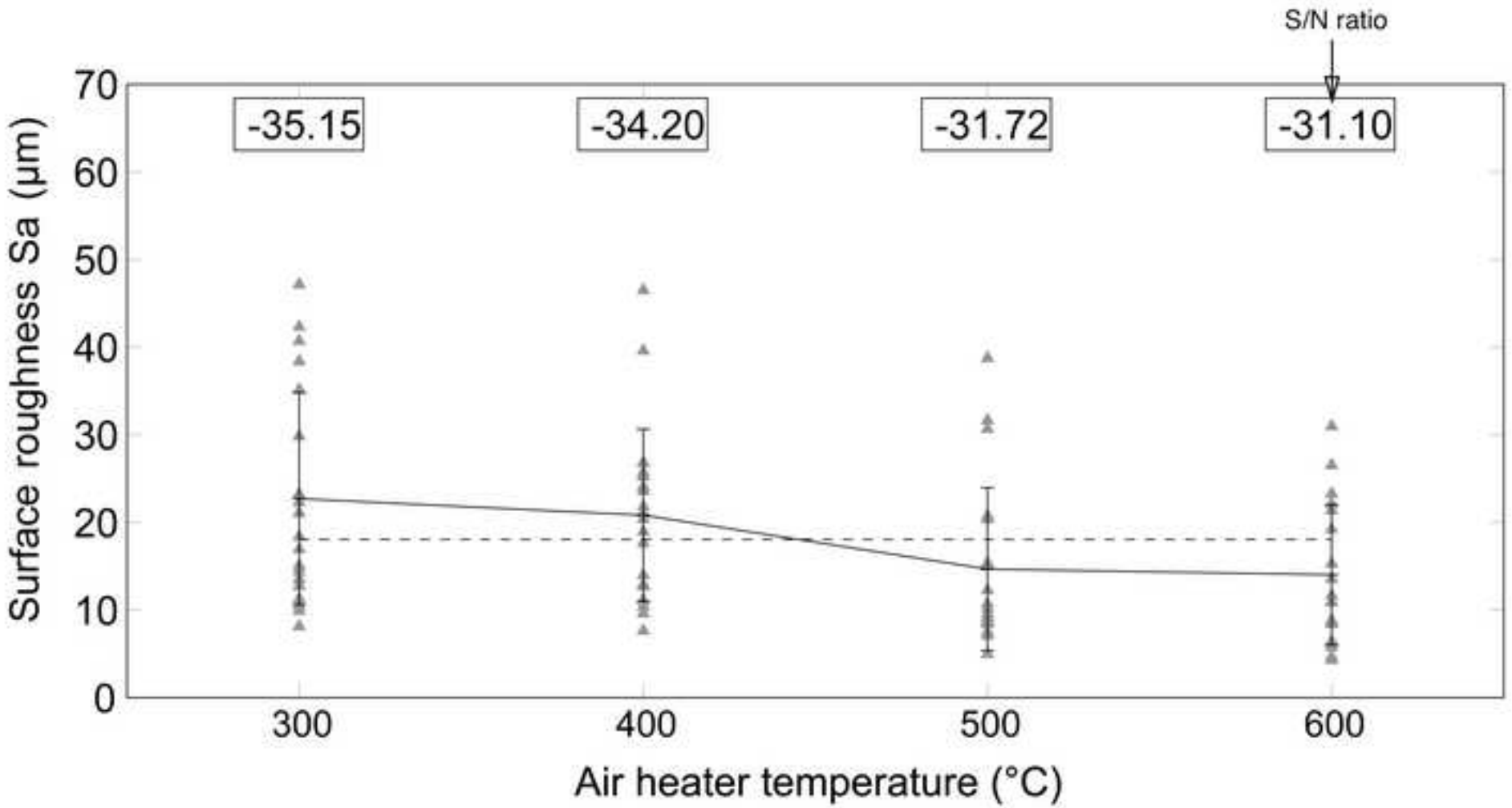
1 Siphon impregnation unit
2 Tubes
3 Peristaltic pump
4 PA12 Slurry
5 Magnetic stirrer
6 Stir bar
7 Yarn

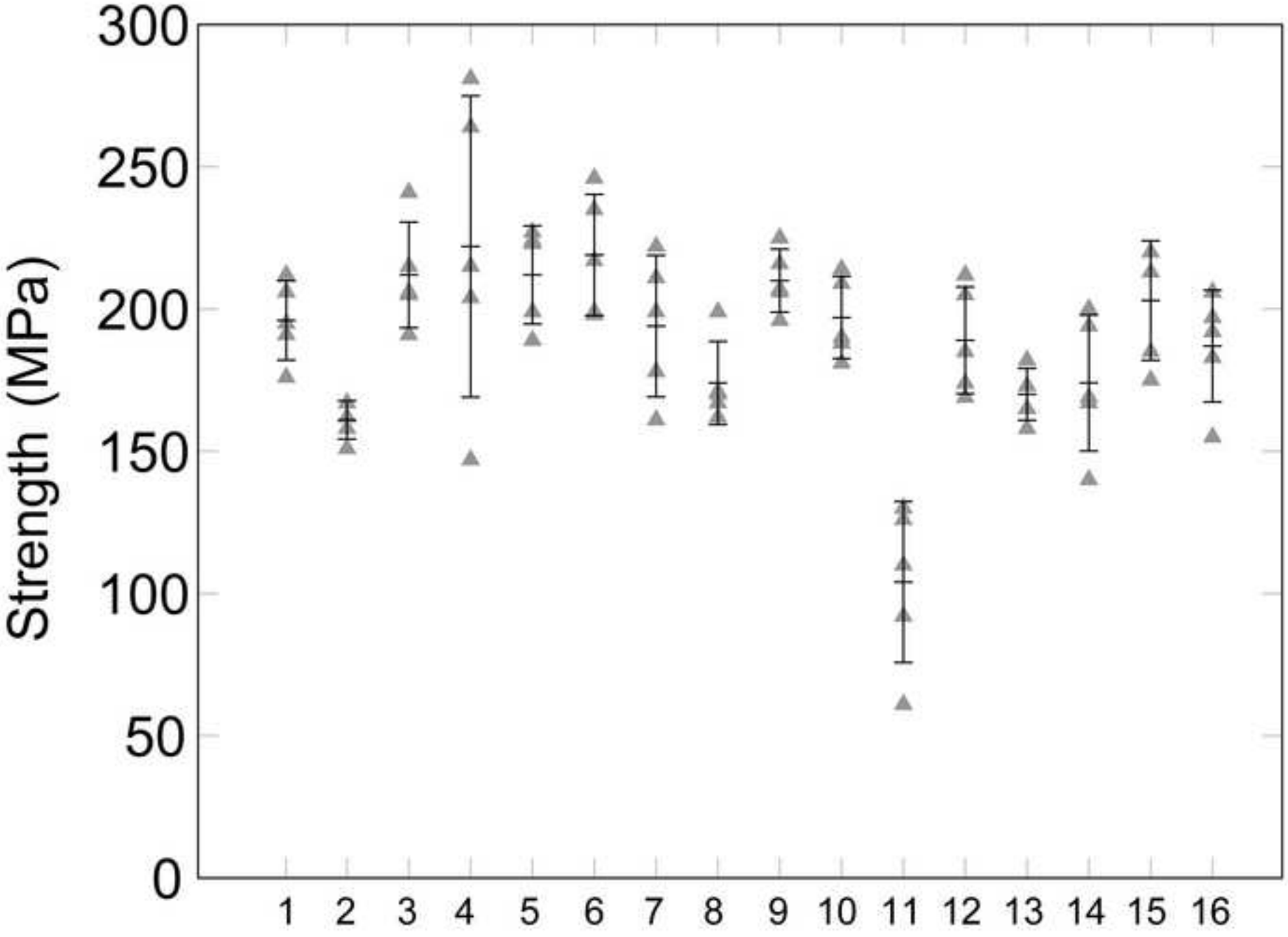


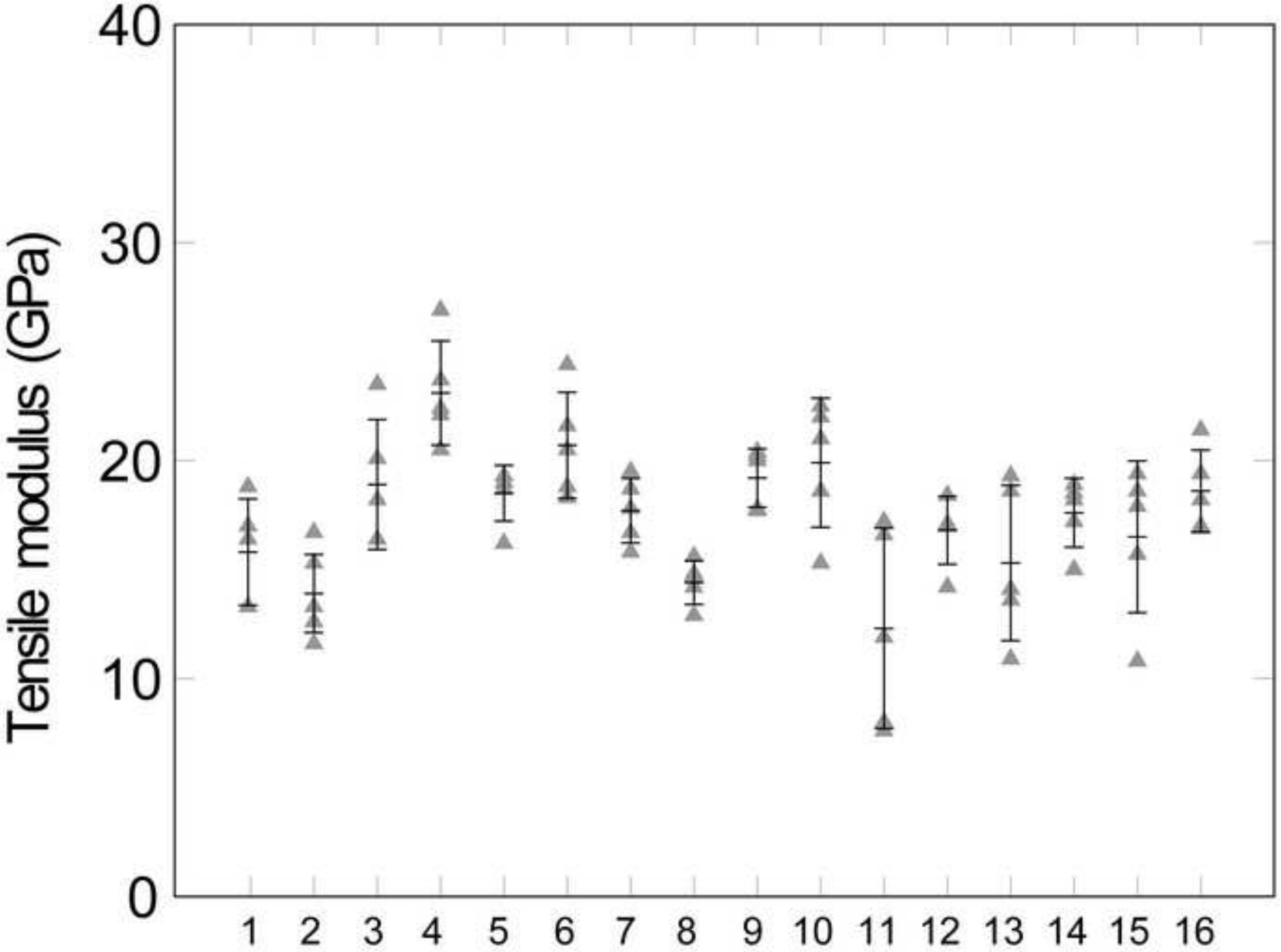


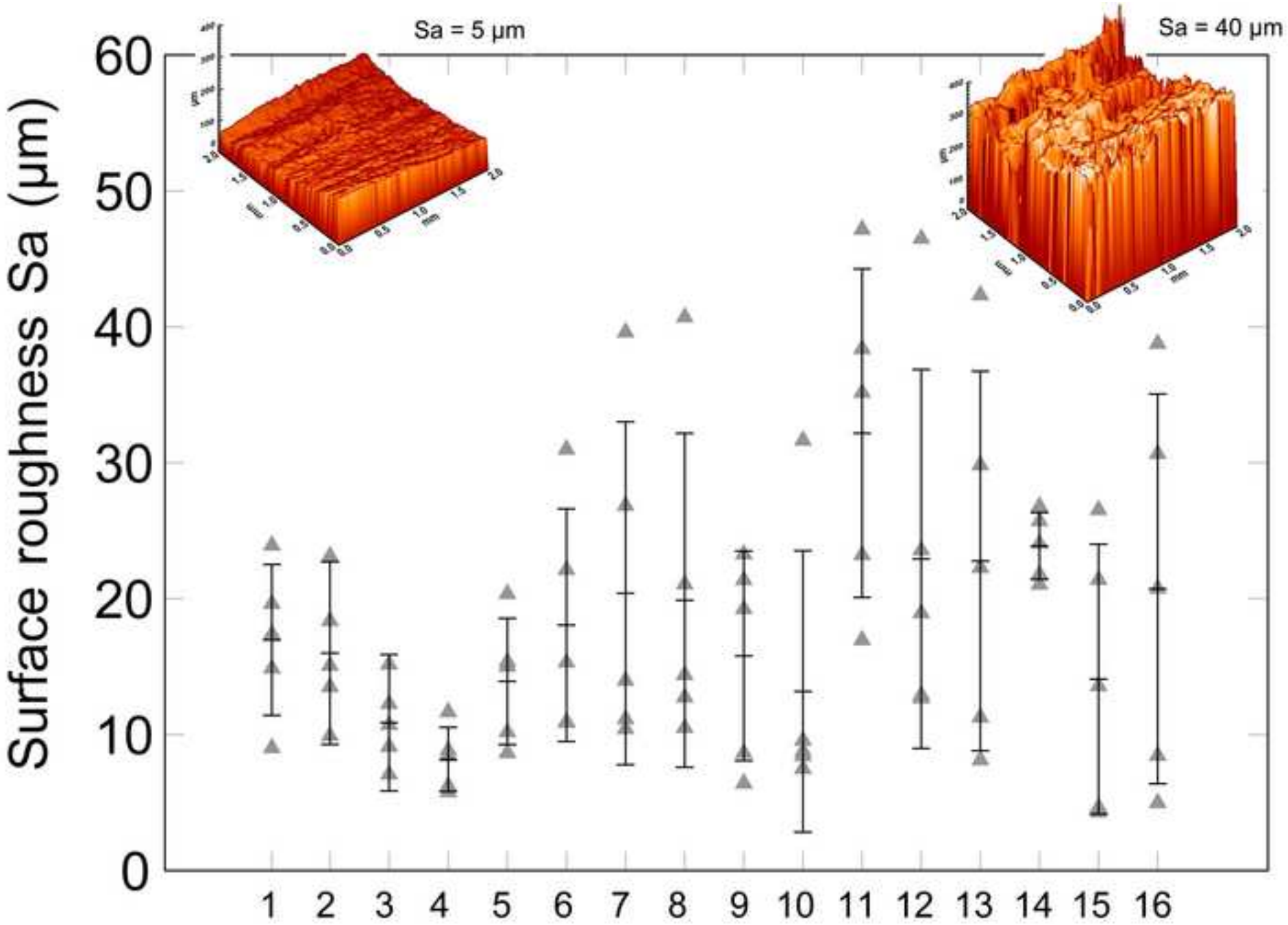


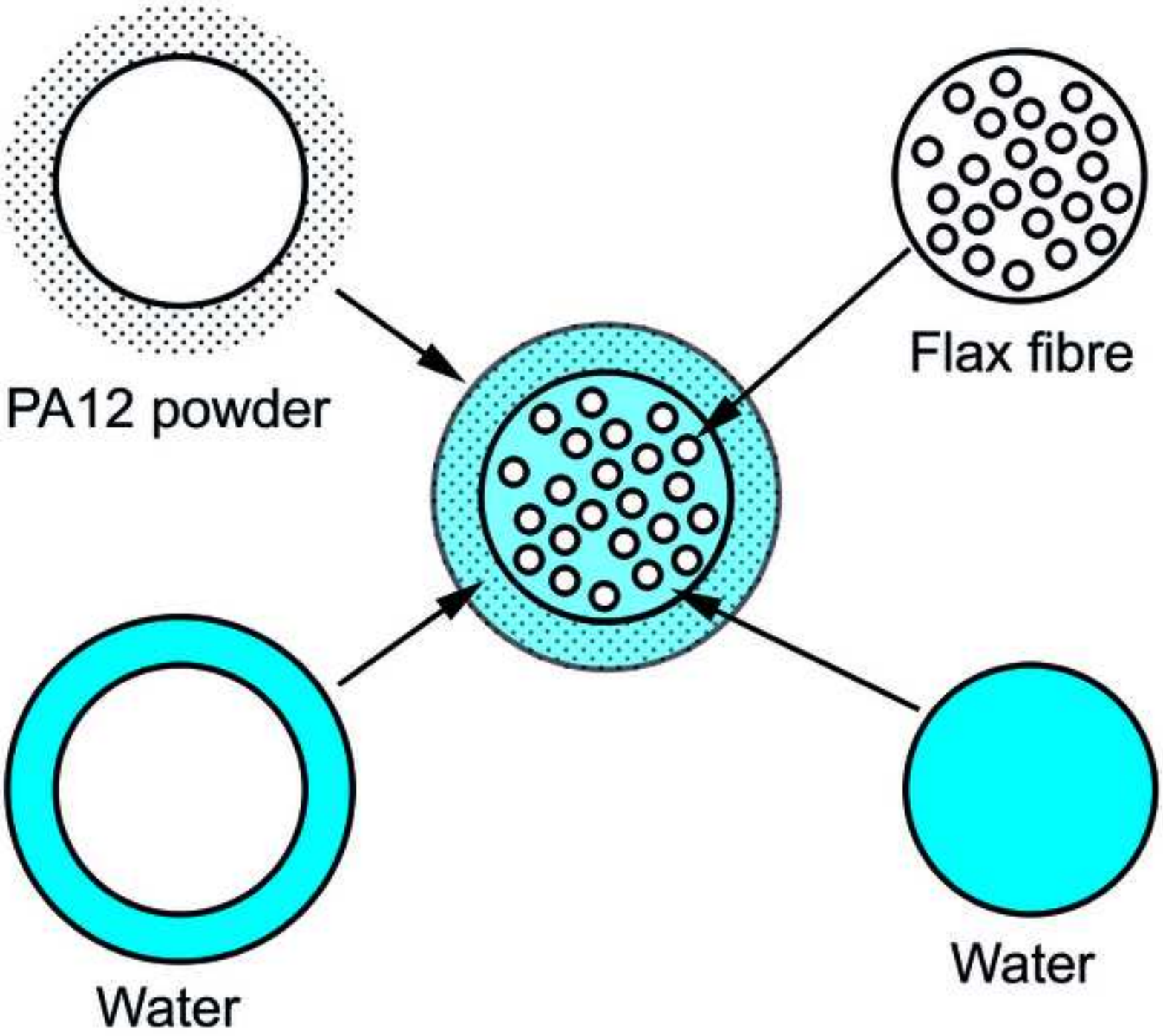


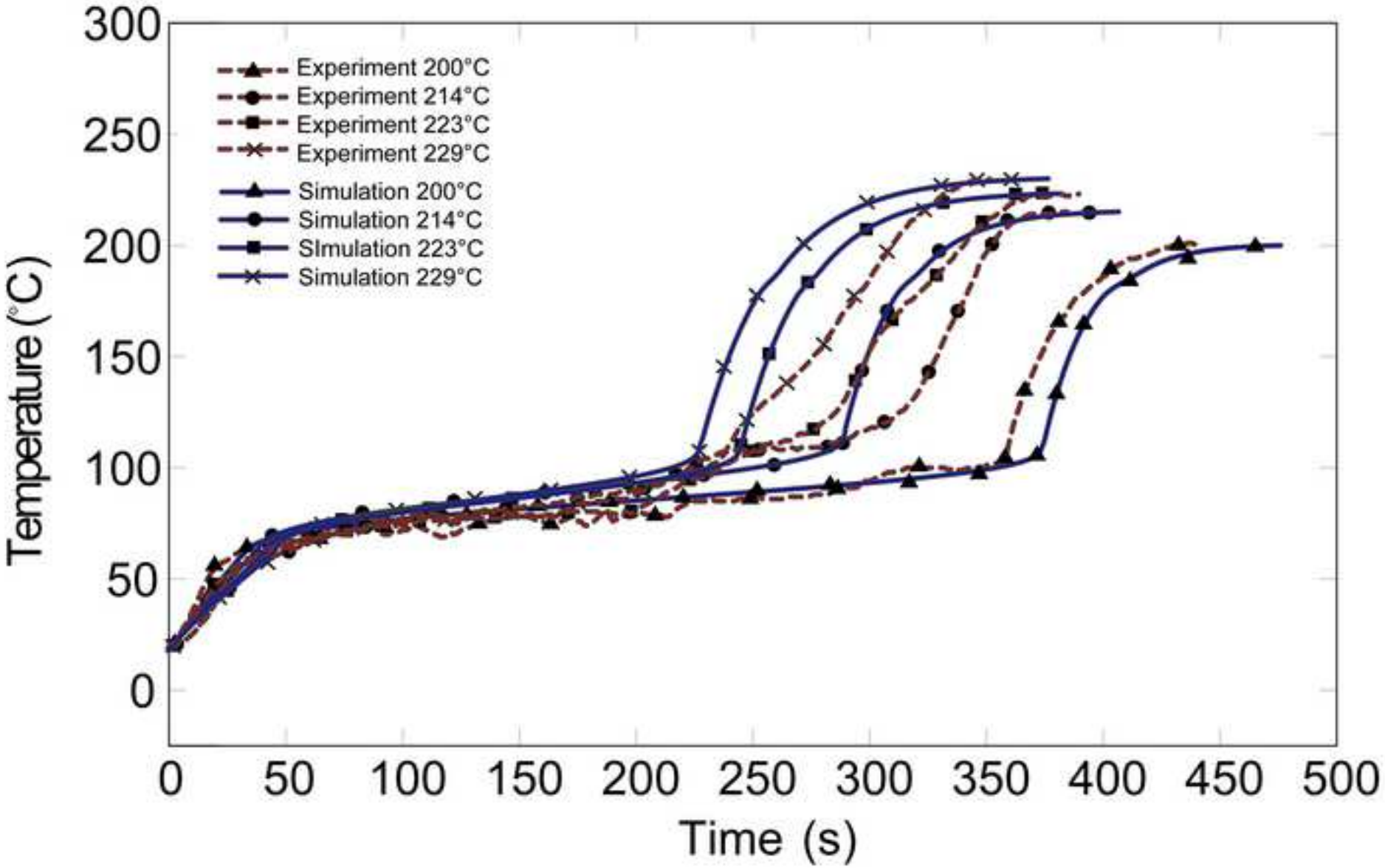


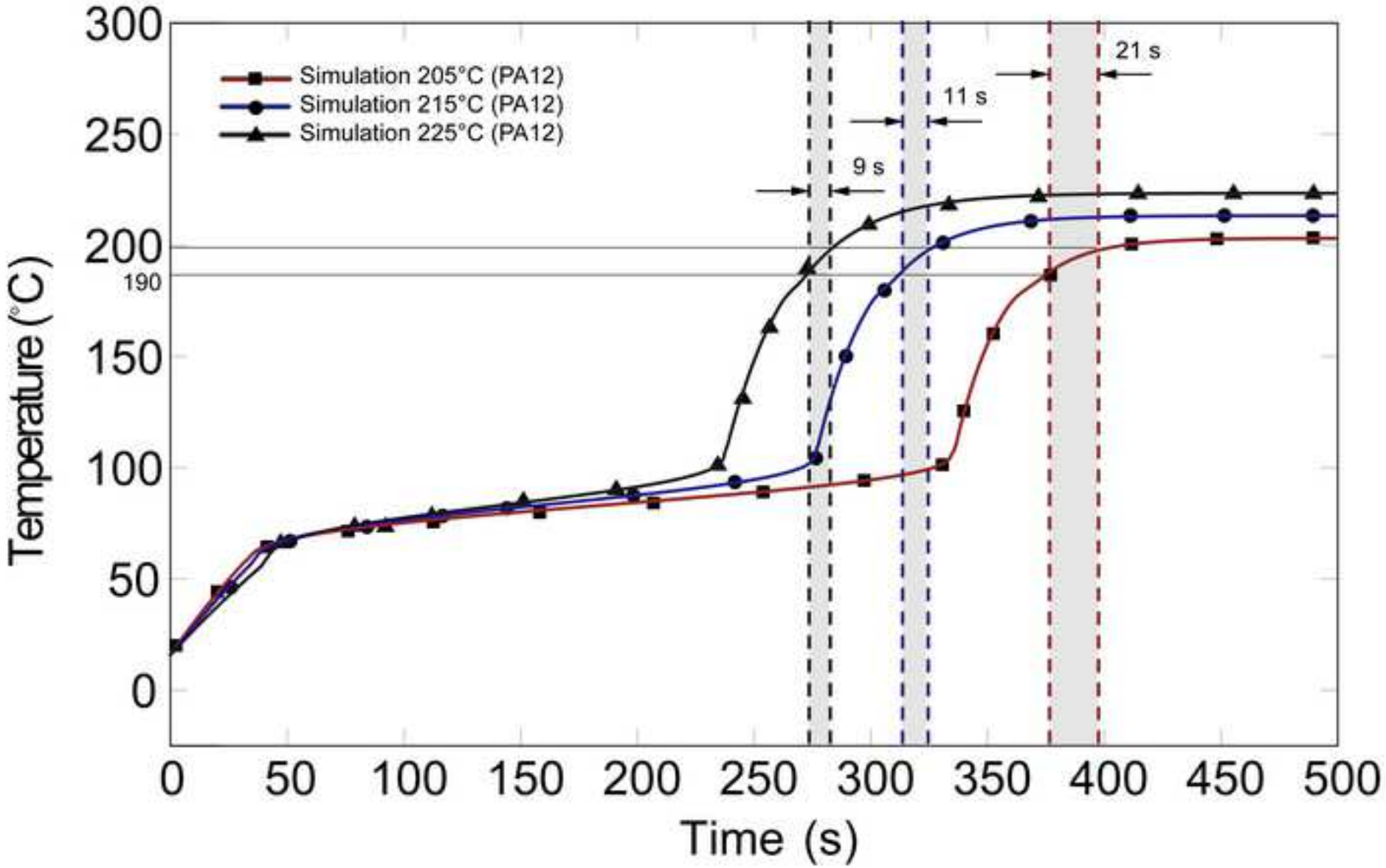


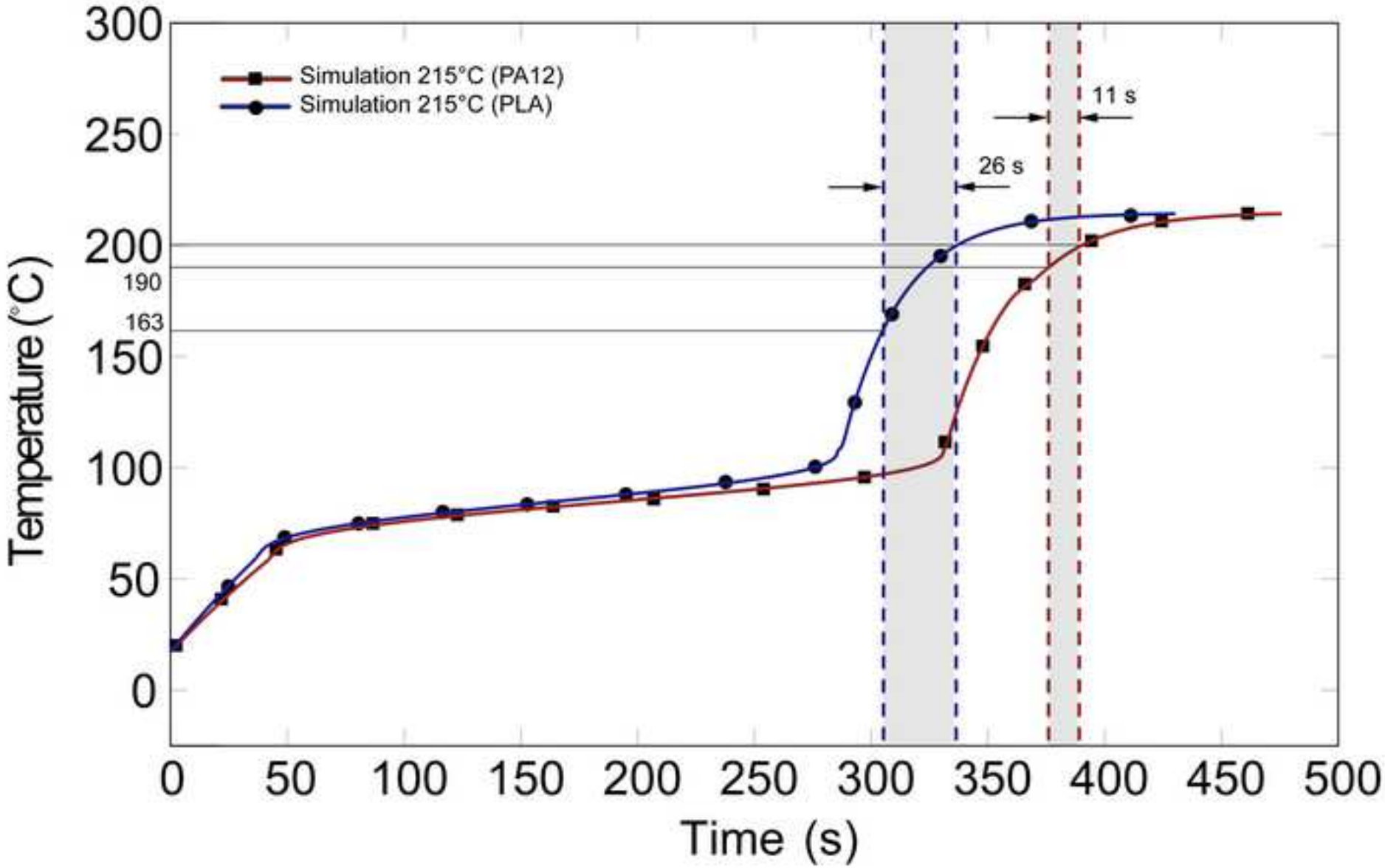


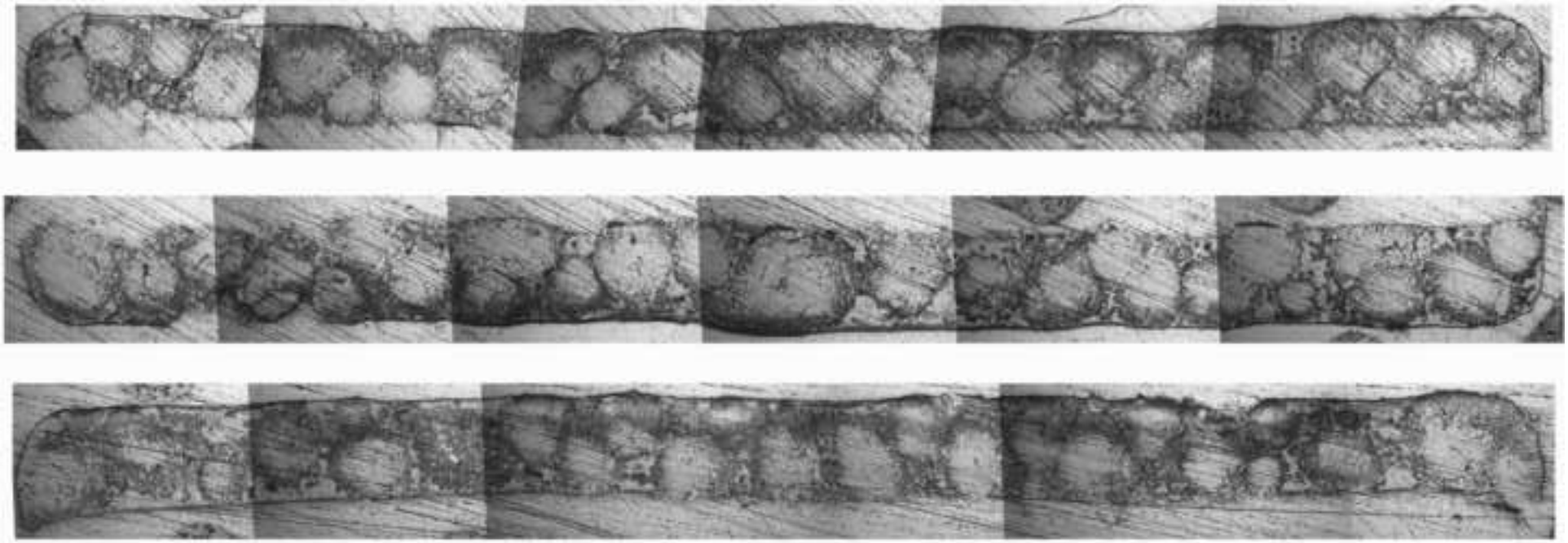


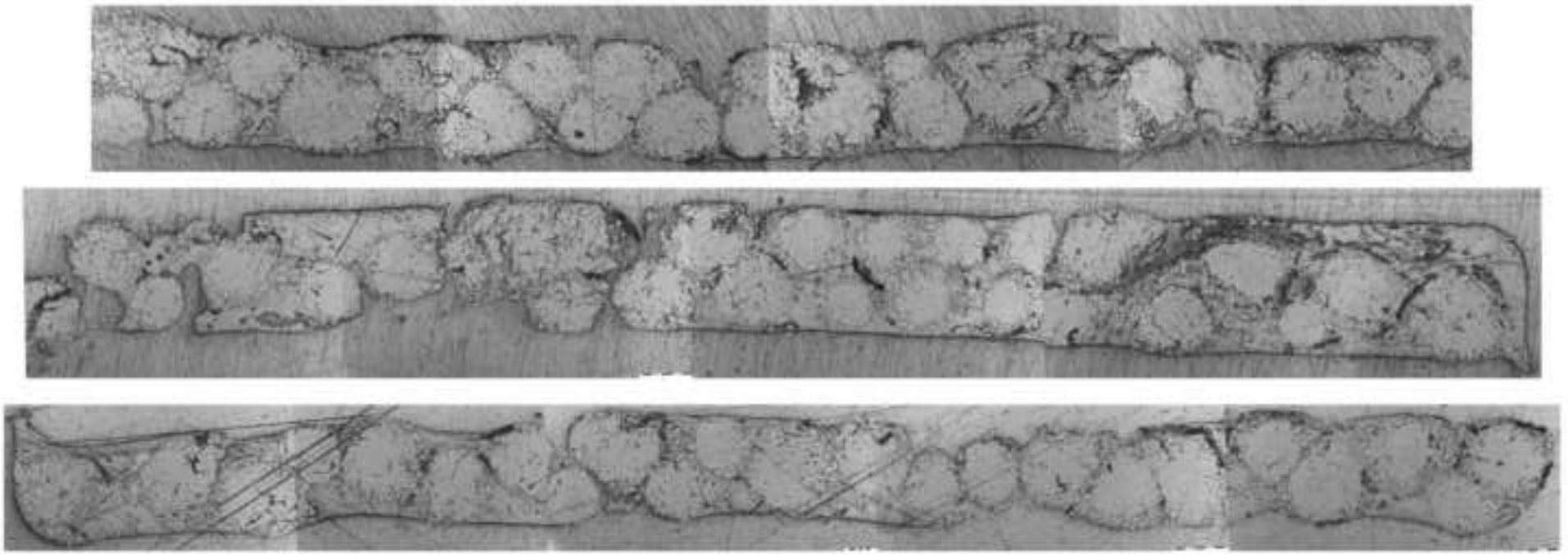


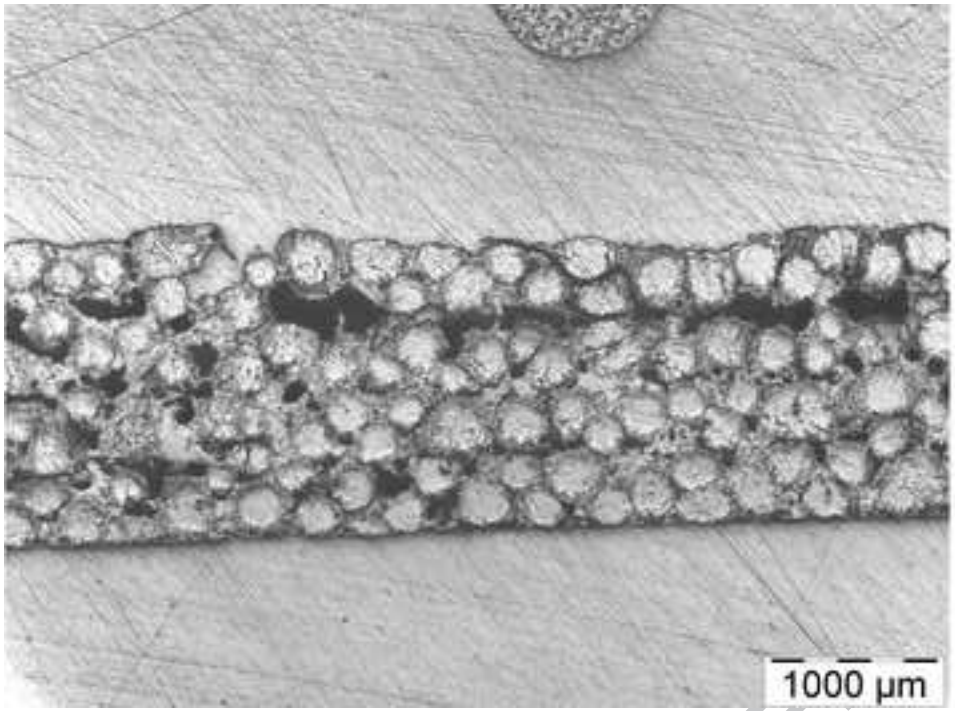


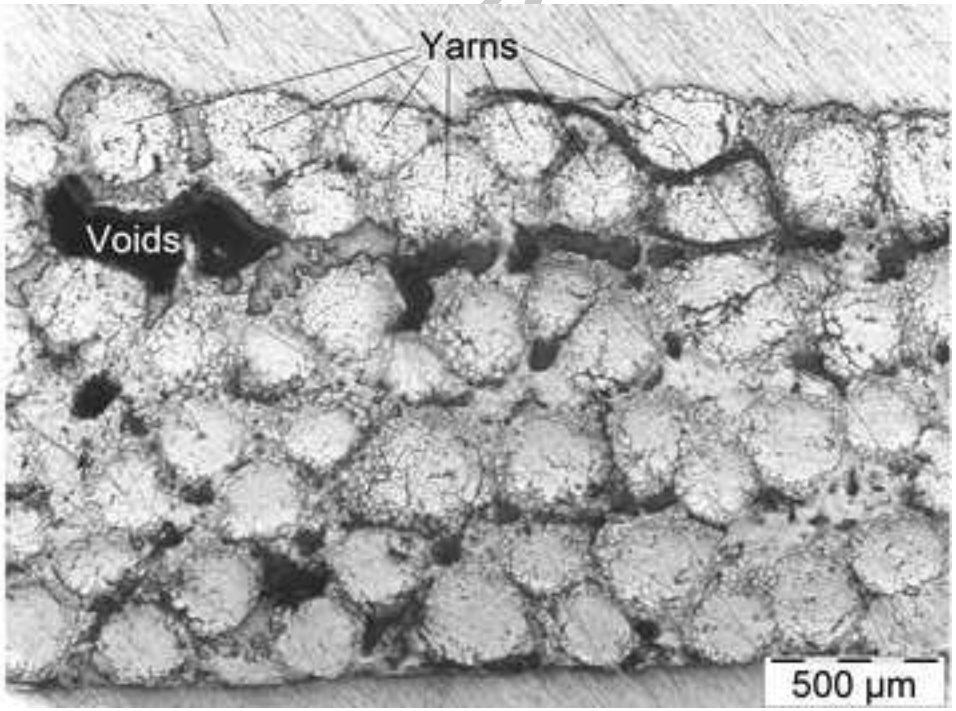












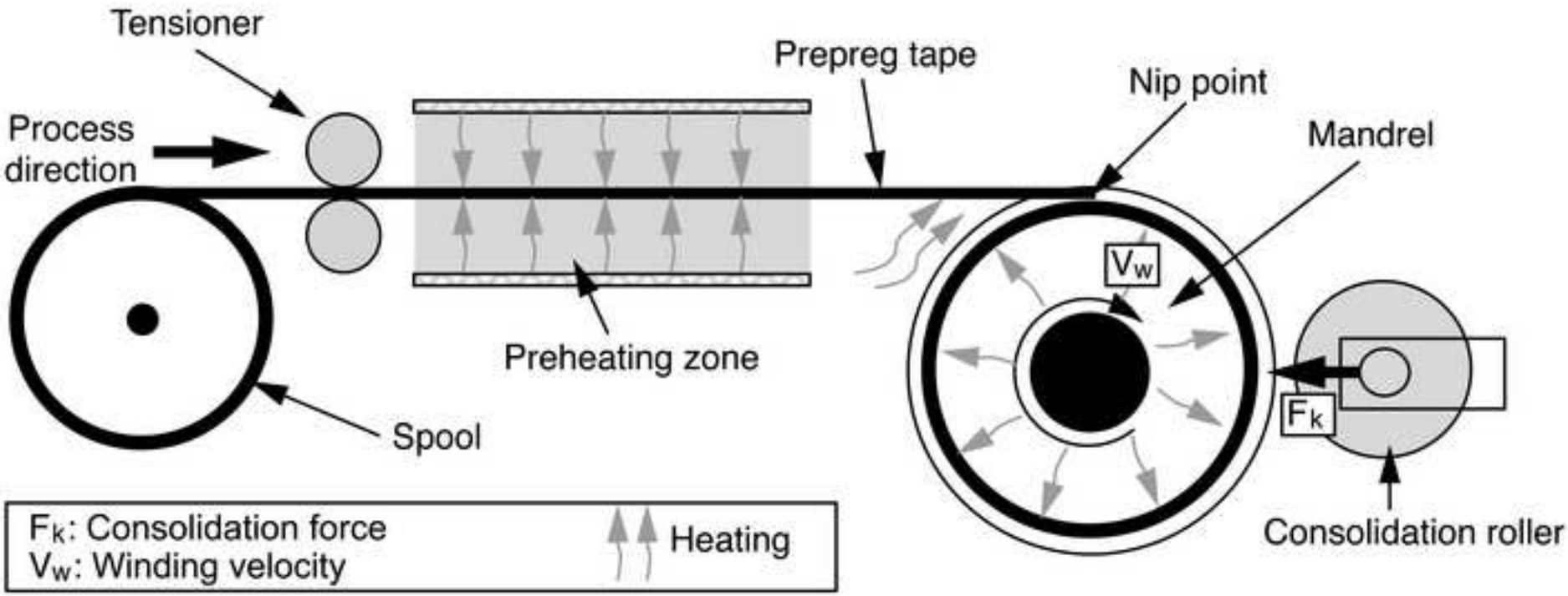


Table 1

Factors and levels used in the Taguchi designed experiment.

Number	Factor	Level			
		1	2	3	4
1	Ceramic heater 1 temperature (°C)	210	205	215	220
2	Ceramic heater 2 temperature (°C)	210	205	215	220
3	Roller oil temperature (°C)	125	120	130	135
4	Air heater temperature (°C)	400	300	500	600
5	Haul-off speed (m/min)	0.09	0.1	0.11	0.08

Table 2

Taguchi L-16 orthogonal array.

Trial Number	Ceramic Heater 1 (°C)	Ceramic Heater 2 (°C)	Roller Oil Temp (°C)	Air heater (°C)	Haul-off Speed (m/min)
1	210	210	125	400	0.09
2	210	205	120	300	0.10
3	210	215	130	500	0.11
4	210	220	135	600	0.08
5	205	210	120	500	0.08
6	205	205	125	600	0.11
7	205	215	135	400	0.10
8	205	220	130	300	0.09
9	215	210	130	600	0.11
10	215	205	135	500	0.09
11	215	215	125	300	0.08
12	215	220	120	400	0.11
13	220	210	135	300	0.11
14	220	205	130	400	0.08
15	220	215	120	600	0.09
16	220	220	125	500	0.10