New automated method for macrocontaminant analysis: Industrial applications

SYLVIE OSSARD, PATRICK HUBER, PASCAL BOREL, DAVY SOYSOUVANH, AND THIERRY DELAGOUTTE

ABSTRACT: In recycled paper processes, stickies are the origin of many production disturbances. In this paper, we present how the recently developed method for macrocontaminant analysis was used with industrial samples for process analysis. The new automated stickies measurement method allows (i) determination of the three-dimensional (3D) morphology of screened particles (without any deformation) and (ii) classification of the particles as stickies among contaminants. This is achieved by a combination of laser triangulation and local near-infrared spectroscopy (NIR).

Measurement of macrocontaminants in pulp samples and their classification allow meaningful evaluation of their specific removal. Chemical nature and amount of the macrocontaminants coming from different raw materials were studied and were shown to be very different in two different mills. In an Asian mill, a low removal of pressure-sensitive adhesives (PSAs) in the process (46%) was found in comparison with high removal of other stickies (99%). Applications of this device are shown, while the new on-line sensor for macrocontaminant analysis is being developed.

Application: This study demonstrates how mills can analyze stickies removal throughout the recycling process. As a consequence, they could control the process depending on the chemical nature of incoming stickies contamination.

In recycled paper processes, stickies are at the origin of many production disturbances, such as machine breaks, defects in paper, and converting problems. Monitoring of macrocontaminants is an essential task for most recycling mills. Currently, mills use tedious and laborious measurement methods. Macrocontaminants are generally separated from the pulp by screening, followed by various inspection and classification methods.

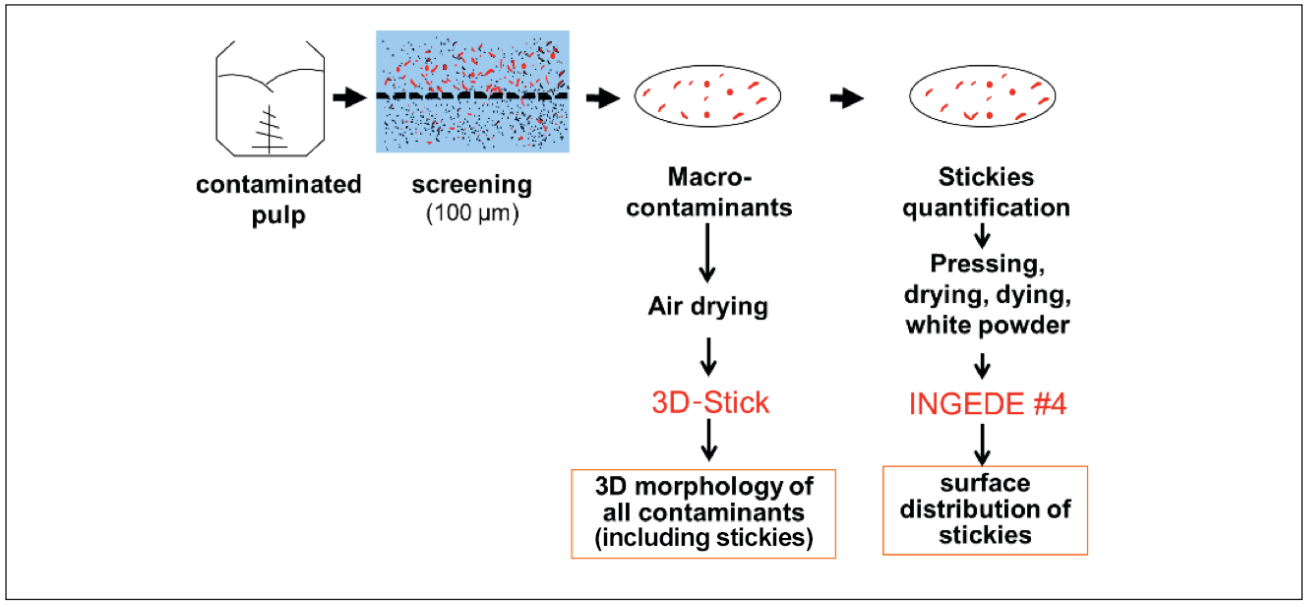
The most recognized methods in Europe are INGEDE #4 [1] and TAPPI Standard Test Method T 277 pm-99 "Macro stickies content in pulp: The 'pick-up' method." They give useful information about stickies particles count and size. Tacky particles (stickies) are specifically colored and quantified by image analysis.

The chemical nature of stickies makes them suitable for analysis by near infrared spectroscopy (NIRS). The technique is rapidly gaining acceptance in the industry, with applications in paper recycling such as sorting a recovered paper stream [2-4], raw material quality assessment [5-7], or quantification of stickies embedded in handsheets [8,9]. According to Doshi, a stickies measurement method should give access to quality of the furnish, efficiency removal processes, deposit control on paper machines, and performance improvement of recycled paper products [10]. In this context, the objective of this work is to describe the use of this new measurement method for stickies studies in recycled pulps in industrial applications for process analysis. The proposed 3D-Stick automated method for macrocontaminant analysis allows (i) determination of the 3-dimensional (3D) morphology of those screened particles (without any deformation) and (ii) classification of the particles as stickies among contaminants, meaning a tool to have both quantitative information (mm^2, or better yet mm^3, of stickies per kilogram of pulp, for example) and qualitative information such as the chemical nature of the stickies.

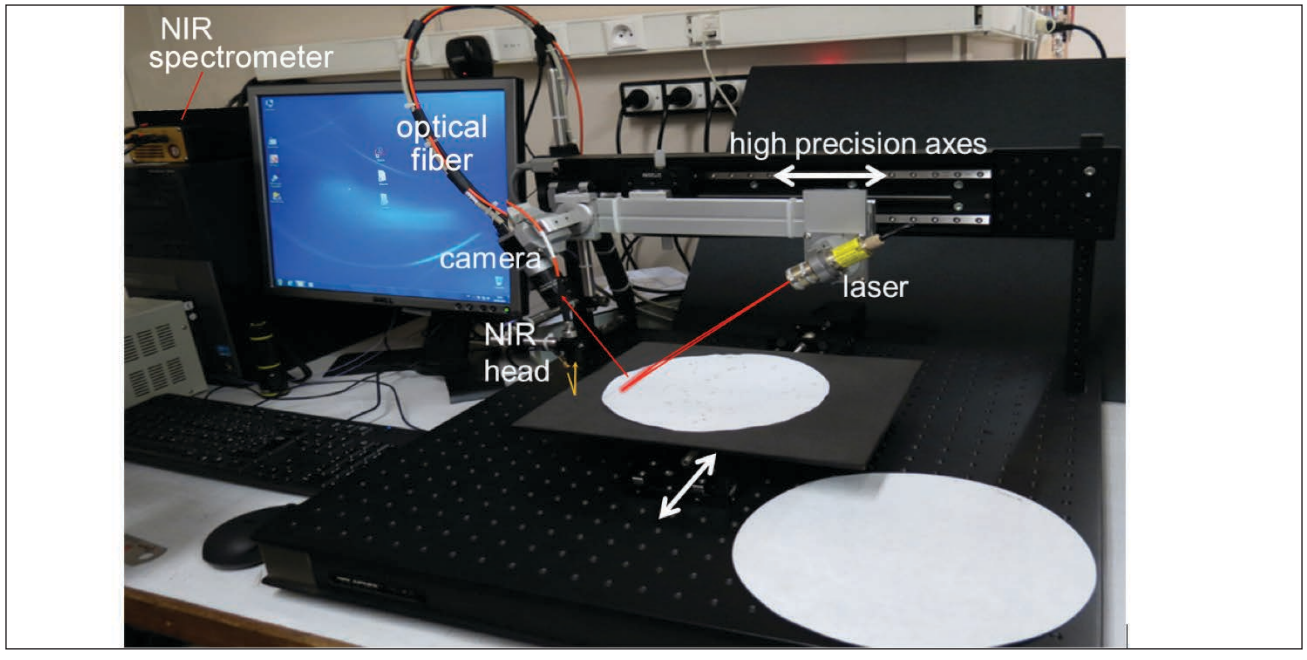
MATERIALS AND METHODS

Conventional characterization of stickies with INGEDE #4 and TAPPI T 277

Contaminants were previously separated from the fibers on a screening device with a 100 µm slots plate. The principles of the INGEDE #4 and the TAPPI T 277 methods are compared with the new proposed 3D-Stick method (**Fig. 1**). The INGEDE #4 and TAPPI T 277 methods are similar in principle, with only a difference in the dying step. The filter with the contaminants particles is covered with non-stick silicone paper and then dried on a Rapid-Köthen device (10 min, 94°C, pressure of 95 kPa). This step causes the sticky particles to strongly adhere to the filter paper. They are also subsequently pressed and deformed. The silicone paper is then released, and non-tacky contaminants are removed



1. Sample preparation for 3D-Stick analysis and for INGEDE #4 method.

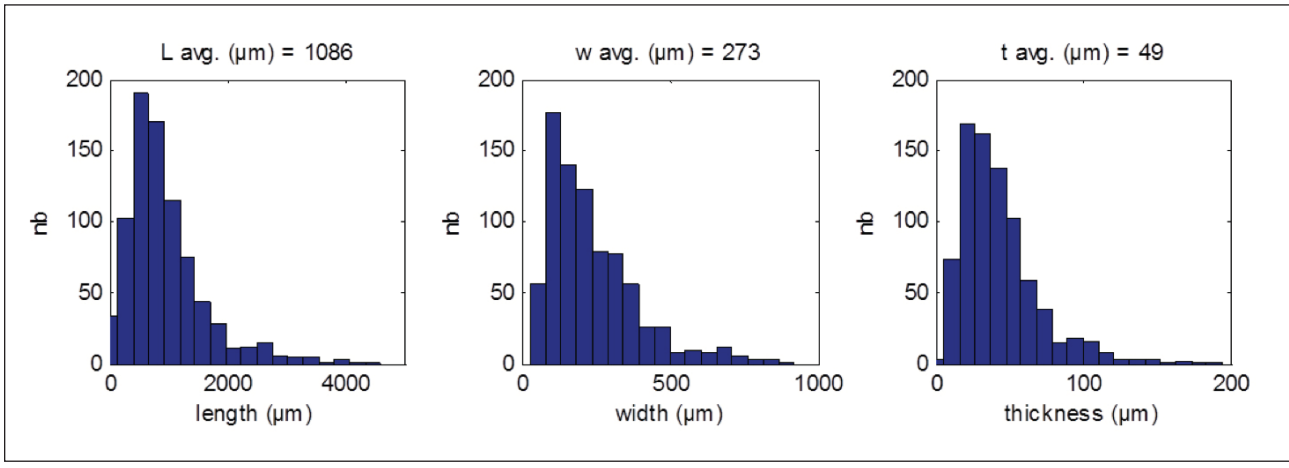


2. Three-dimensional (3D) stick device: A laser line and a camera perform the mapping by laser triangulation, and a near infrared (NIR) head analyzes chemical composition of contaminants.

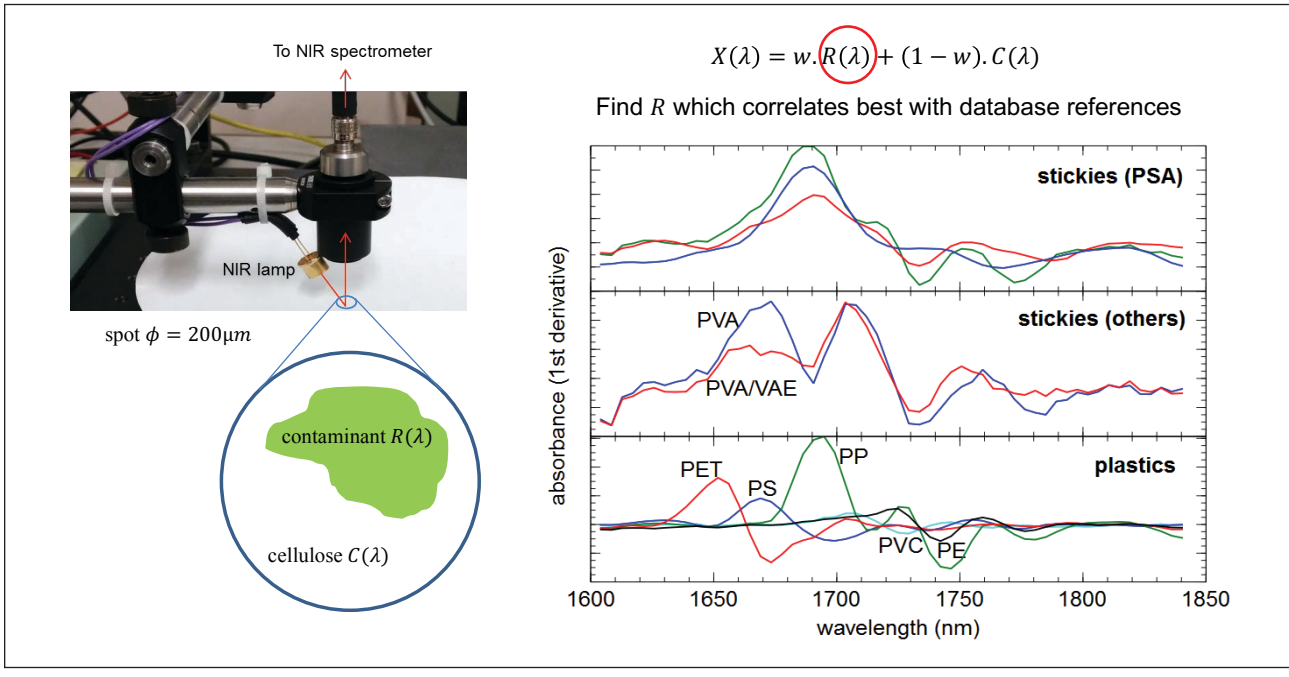
from the surface with a soft brush. The filter is then completely dyed through immersion in a water-soluble black ink bath. The dyed filter is dried on a Rapid Köthen device between 2 silicone papers under the same conditions described previously. Then, the stickies particles are revealed with a white alumina powder (INGEDE) or white coated paper (TAPPI) after pressing and drying again. This leaves only white stickies particles on a black background. Stickies are then counted and characterized on a desktop scanner through image analysis.

3D-Stick measurement

The principle of the 3D-Stick measurement has been previously described [11]. This method is briefly described as follows. The 3D-Stick analysis is achieved by a combination of laser triangulation and local NIRS (**Fig. 2**). First, the contaminants deposited on a filter paper are scanned by a moving laser line. The reconstruction of the altitude profiles gives access to the length, width, and thickness distributions of the macrocontaminants, with a resolution of 20 µm x 20 µmx 3 µm. An example of stickies size distribution



3. Example of industrial stickies size distribution (based on the analysis of 819 stickies).



4. NIR recognition of chemical nature of contaminants.

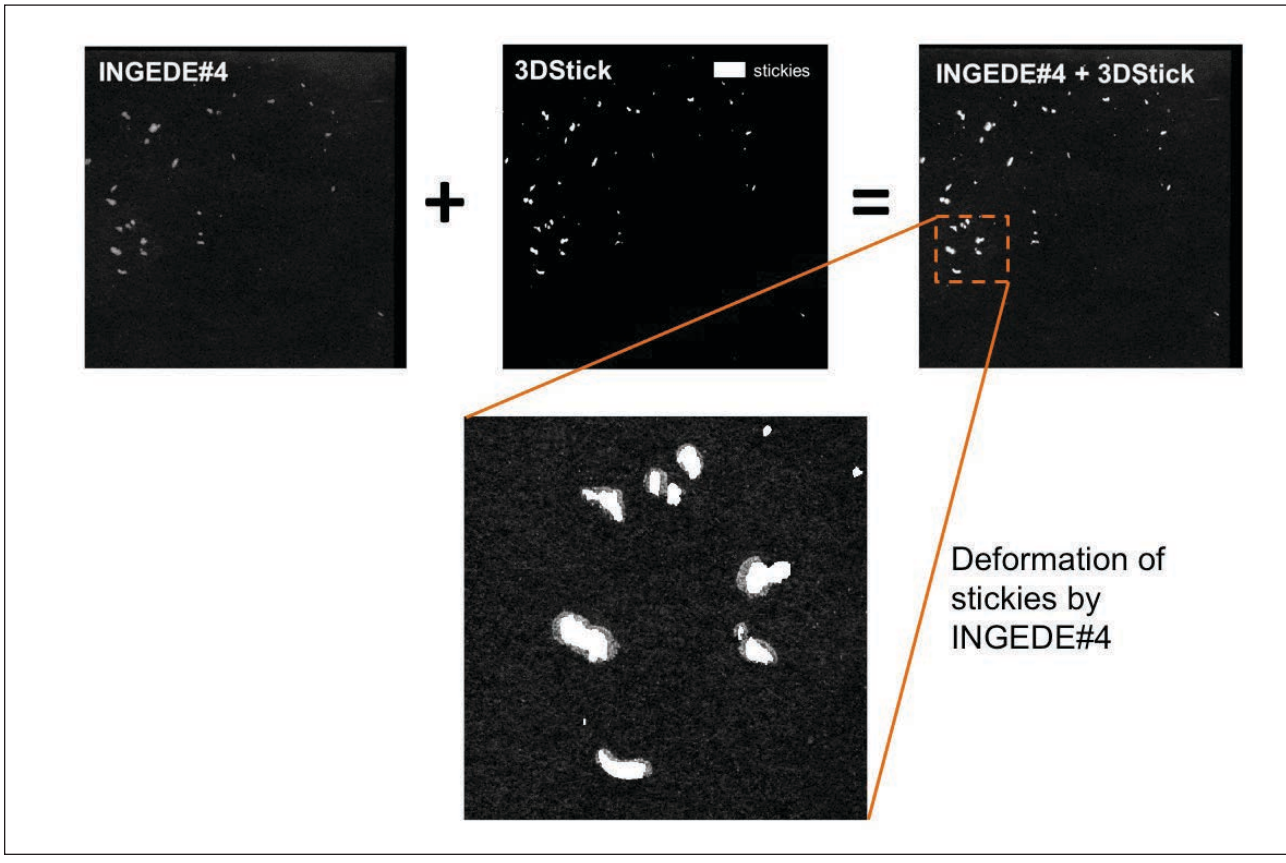
is illustrated in **Fig. 3**, where the average size is 1100 µm x 250 µm x 50 µm. Then, the chemical nature of the contaminants is assessed by NIRS. A database of expected compounds was built with model glues present on the market (**Fig. 4**). Several classes of contaminants were defined for classification according to previous mill experiences: stickies (pressure sensitive adhesives [PSAs]), stickies (others), and plastics. For the "stickies PSA" class, several acrylic model samples from glue suppliers were analyzed and added to the database. For the "stickies (others)" class, several model glues were included: among them several polyvinyl acetate (PVA)-based compounds (PVA homopolymers) and several mixtures of PVA and vinyl acetate ethylene (VAE) copolymer glues. Several glue formulations based on ethylene vinyl acetate (EVA) have been used as model hot melt stickies.

RESULTS AND DISCUSSION

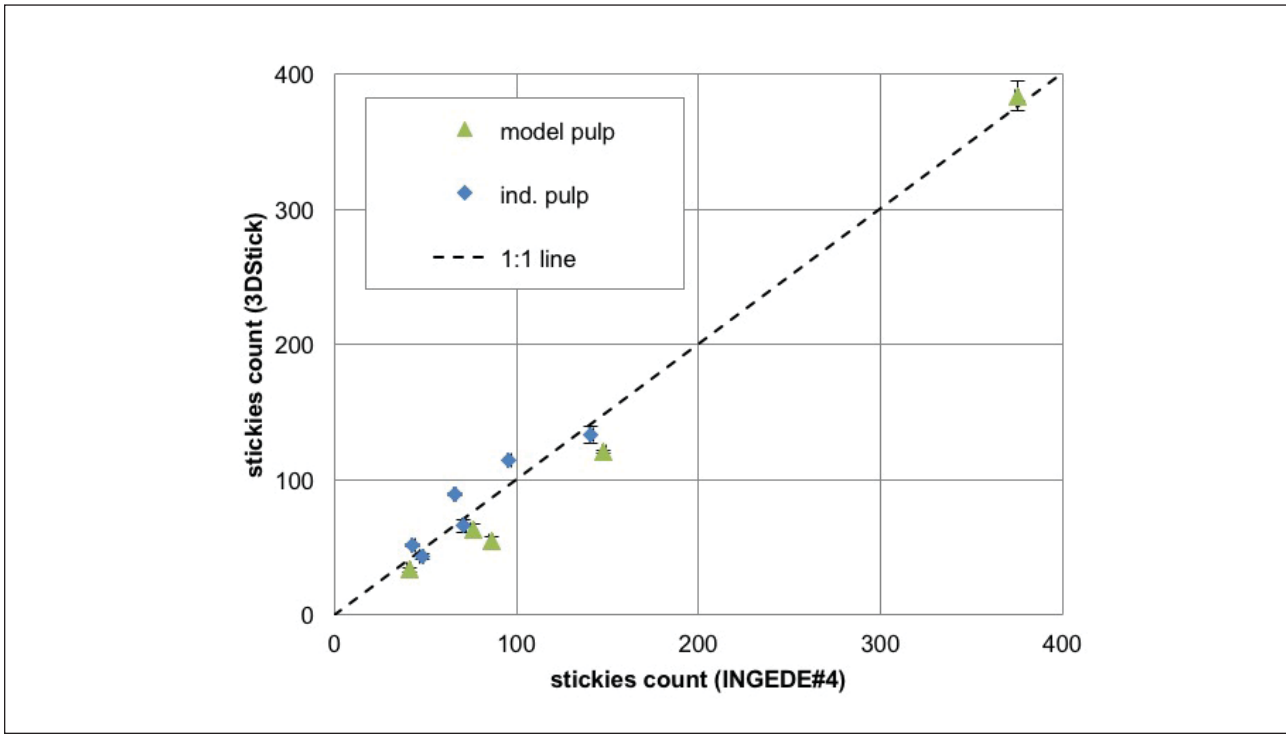
Method comparison

The comparison between the INGEDE #4 method and the 3D-Stick measurement has previously been published [11]. It was shown that contaminant morphologies are modified through pressing and heating steps in the INGEDE #4 method, as illustrated in **Fig. 5**. They are measured in their initial state in the proposed 3D-Stick method, avoiding this deformation. Also, the comparison of stickies count in number between the two methods was well correlated, as shown in **Fig. 6**.

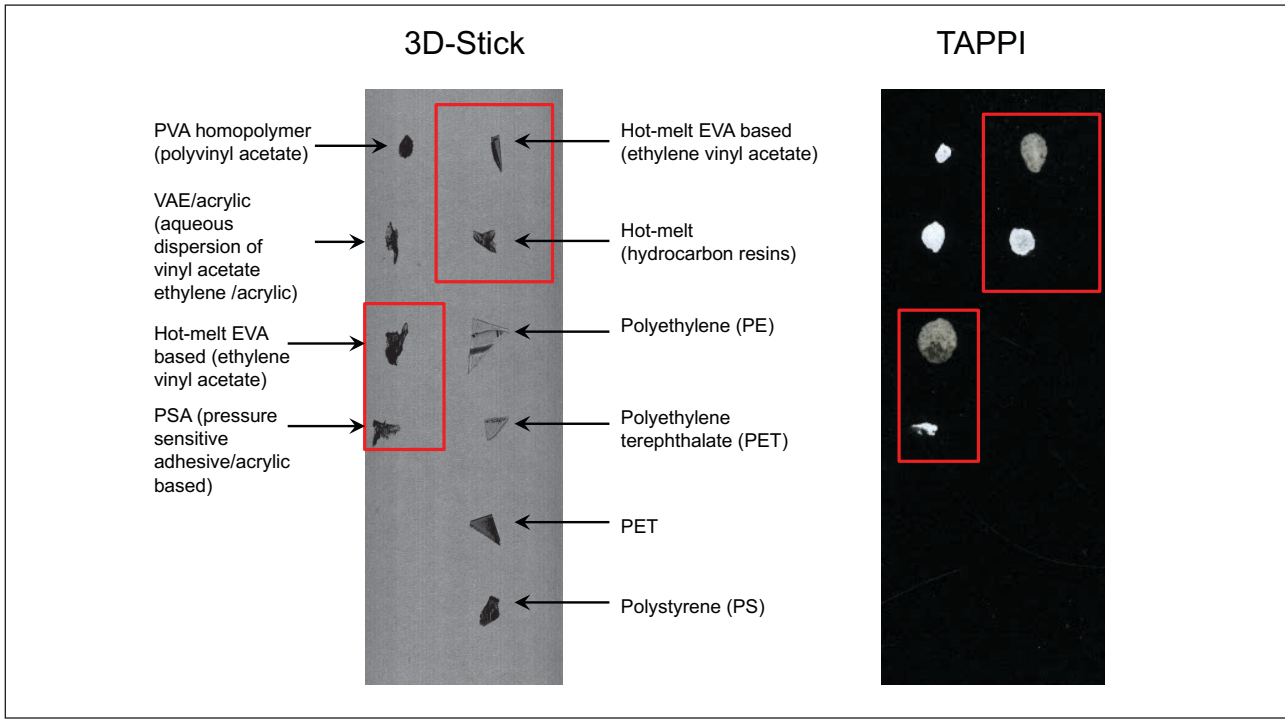
Besides those results, it was found that comparisons between 3D-stick measurements and the TAPPI T 277 method were not correlating in a specific industrial mill. This was not expected, as previous results indicate that INGEDE #4 and the TAPPI T 277 method were similar, with the main difference



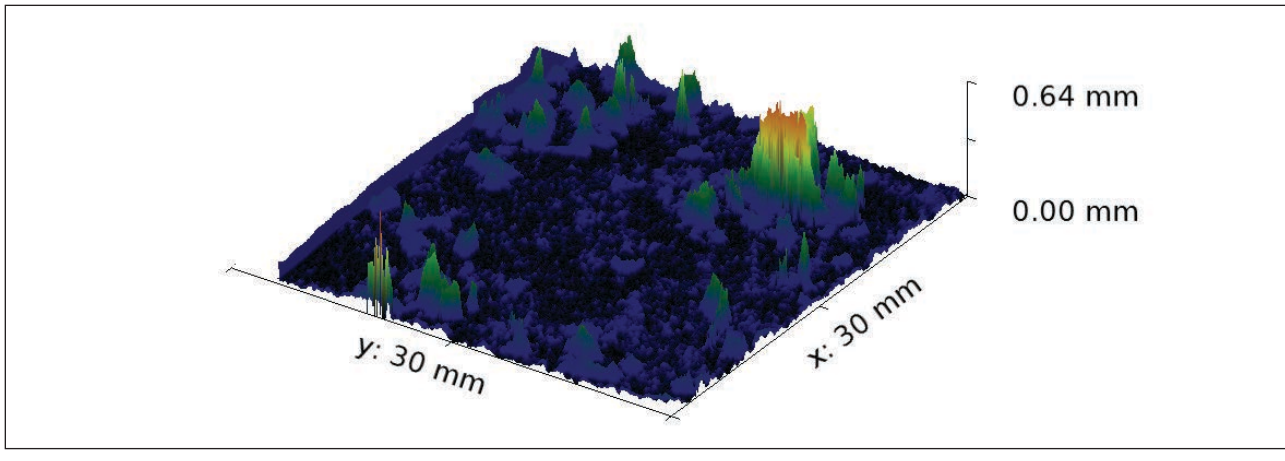
5. Illustration of deformation of the stickies due to pressing and heating.



6. Stickies count in number with 3D-Stick and INGEDE #4 methods.



7. Model contaminants deposited on a filter paper and identified with 3D-Stick (left) and with TAPPI Standard Test Method T 277 pm-99 (right).



8. A 3D image of part of sample 1 for illustration.

being that alumina powder is used to whiten sticky contaminant in the first, and coated paper in the second. The principles of the methods are similar.

The comparisons of the results on model contaminants analyzed with both methods are shown in **Fig. 7**. EVA-based hot melt, hydrocarbon hot melt, and PSA appear grey with the TAPPI T 277 method. They are not counted with the mill protocol and optical device. In this method, tacky particles are colored in white when they are pressed and heated in contact with a white coated paper. The grey color indicates that the coating has not been well detached and transferred to the stickies particles. On the other hand, the 3D-Stick perfectly identified the particles as EVA contaminants. These differences may be at the core of the differences between mill and 3D-Stick measurements and of the poor correlation between paper machine issues and stickies measurement performed in this mill. Those results indicate that 3D-Stick measurement gives more information than the mill method, with breakdown in contaminant classes (hot melt, total PSA and plastic contamination).

Process studies

The objectives of the process studies were to (i) evaluate contamination between two Asian mills, called production A and


Production A after pulping


Production B after pulping


Production B at the end of the process

9. Filter paper where stickies were deposited.

	Production A After Pulping	Production B After Pulping	Production B End of Process
Stickies (PSA)	3556	135	72
Stickies (others)	15227	7151	62
Plastic	101	49	9
Total stickies	18783	7286	134
PSA = pressure sensitive adhesive.			

I. Contamination in mm²/kg of the pulp in productions A and B (from 3D-Stick measurement).

production B, and (ii) evaluate contaminant removal in production B. A reconstructed 3D map is shown in **Fig. 8**, for example. Results of 3D-Stick measurements on the filter paper (**Fig. 9**) are shown in **Table I**.

The macrostickies contamination of production A after pulping is very high and much higher than that in production B (18800 versus 7300 mm²/kg). The raw materials are very different in the two mills, explaining those results. PSA stickies are found to represent about 19% of total stickies contamination in production A and 2% in production B. Those ratios are much lower than in European mills, where PSAs represent about 60% to 80% of the stickies contamination at the process inlet [12]. Those results indicate that the classification of stickies could be different in Asian mills (and probably in U.S. mills, which have not been tested up to now) than in European mills. In Europe, a differentiation of PSA and non-PSA stickies is adequate for process analysis; another stickies classification may be proposed with 3D-Stick analysis, and may be more adequate in the future for mills on other continents. However, PSAs represent 53% of the final pulp contamination in production B, indicating that their removal is lower than for other stickies, as found in Europe [13].

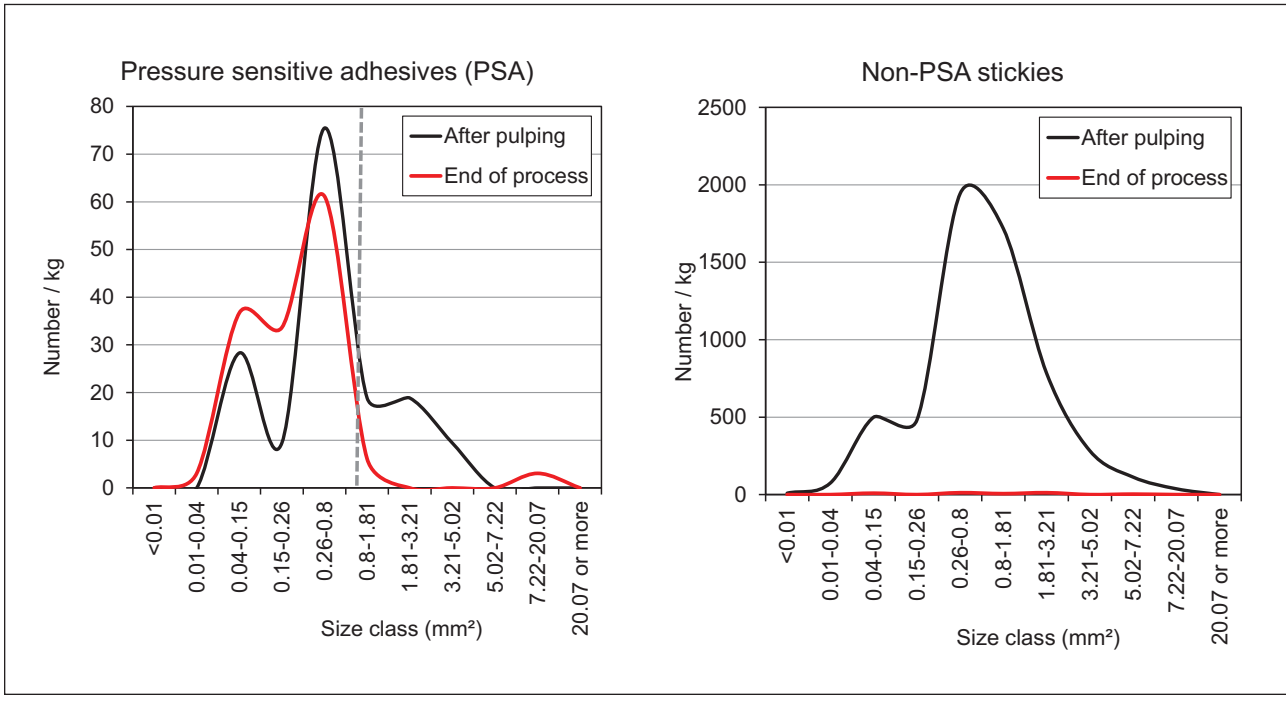
	Process Removal Efficiency (in % surface area)
PSA	46%
Other stickies	99%
PSA = pressure sensitive adhesive.	

II. Process removal efficiency, in percent surface area, for grade B, according to nature of stickies.

As presented in **Table II**, the process of removal is very different for PSA and non-PSA stickies. Overall, the removal of non-PSA stickies is very good and exceeds 99% as shown. On the contrary, PSA removal is lower, calculated as 46%. A similar result was already shown in a previous study through INGEDE #4 measurements [14] and a derived method for specific PSA count [13]. The proposed 3D-Stick method gives access to those results with a fast (20 min) and easy method (fully automated once the screen rejects have been deposited on a filter paper).

The morphological characteristics of stickies can be studied and their removal according to their size range evaluated. The frequencies by size classes of the stickies for production B samples are shown in **Fig. 10**. The decontamination of the stickies through the process is very good for non-PSA stickies for all the size ranges. This is consistent with the 99% removal of this stickies category.

On the contrary, for PSA particles, the size has a clear impact in their removal. The larger stickies with a size above 0.8 mm² (equivalent diameter of 1009 µm) are efficiently removed by the process. The PSAs smaller than 0.8 mm² are not effectively removed in the process. Those small stickies have been called mini-stickies in previous publications and



10. Stickies distribution according to size classes at the production B (i) inlet pulp and (ii) the end of the process.

have been shown to pass through the screening steps of most mills [12,14]. Hence, our results are consistent with previously shown PSA behaviors. It is noticeable that the size range of those mini-stickies seems to switch to smaller particles from the beginning to the end of process. This may be due either to:

(i) Fragmentation of PSA particles.

(ii) Better removal of stickies in the size range of 0.26–0.8 mm² and no or nearly no removal of smaller PSA particles. Hence, the smaller stickies are following the pulp flow. Some pulp and solids in suspension are removed from the process through flotation and screening steps, inducing weight losses. As a consequence, a small PSA number expressed per kilogram of pulp will be higher at the end of the process, and apparent contamination increases through the process.

Remarks

Please note that the results in this report are expressed in number/kg and in mm²/kg to be comparable with most methods used for stickies analysis (microscopic count, TAPPI, INGEDE methods), but the data obtained with the 3D-Stick will in the future be presented in mm³/kg to take into account the three dimensions of the stickies.

In the case of this Asian mill, the number of unidentified objects was quite high (up to 10% on the total objects, fibers, and contaminants) compared to amounts normally less than 5% in European mills. The update procedure of the database is straightforward (only one reference spectrum must be added), and this has already been performed in other mill cases.

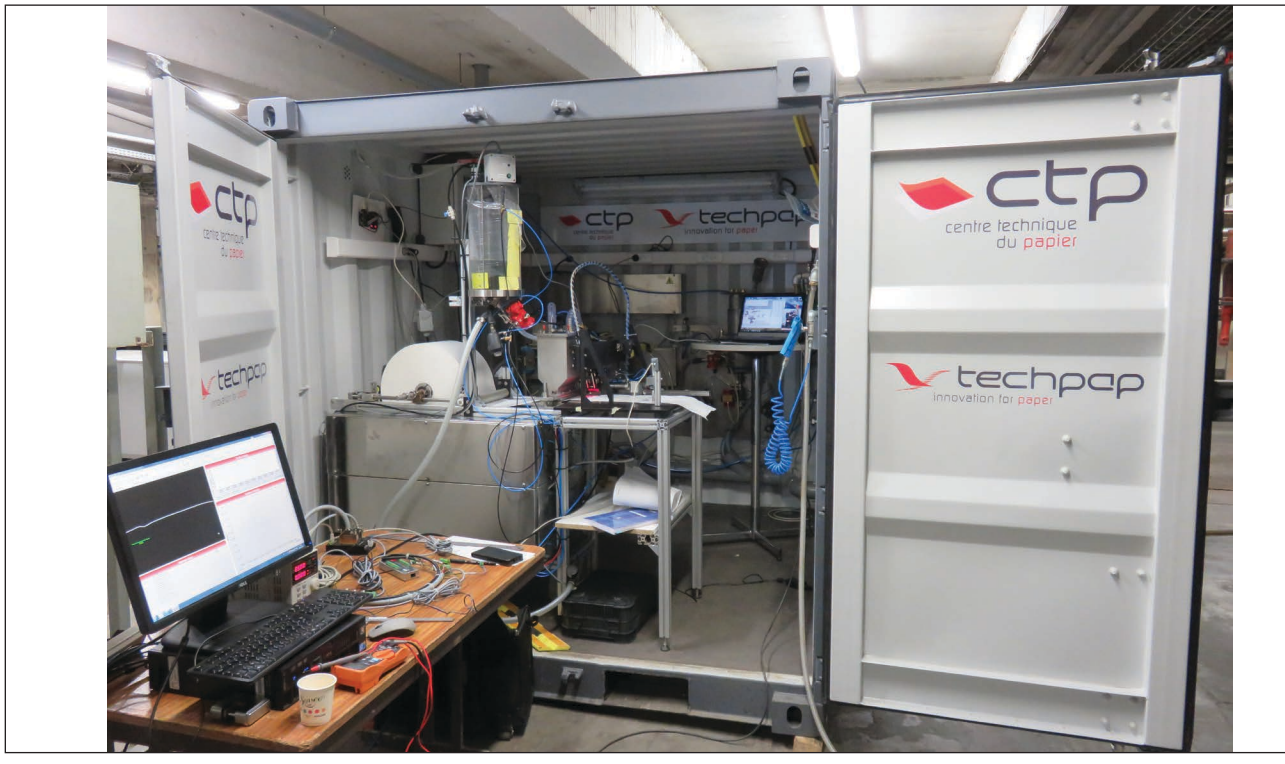
3D-Stick Class	Chemical Base
PSA	Acrylic-based PSA
PSA (hot melt)	SBR and styrene acrylic
Hot melt (non-PSA)	EVA-based hot melt and others
PVAE	PVA-VAE based
PVA	PVA based
Plastics	PE, PET, PP, PVC, PS
Stickies (other)	Non-classical glues as latex

PSA = pressure sensitive adhesive; SBR = styrene butadiene; EVA = ethylene vinyl acetate; PVA = polyvinyl acetate; VAE = vinyl acetate ethylene; PE = polyethylene; PET = polyethylene terephthalate; PP = polypropylene; PVC = polyvinyl chloride; PS = polystyrene.

III. Stickies classification performed by the 3D-Stick.

PERSPECTIVES

As described in the previous "3D-Stick measurement" section, many different model glues have been used to differentiate the stickies according to their chemical nature. Overall, a total of 45 different contaminants have been included into the database, and the main categories are presented in **Table III**. Those classes can be used for more precise analysis, depending



11. Photo of the first prototype of the PI-Stick for on-line macrocontaminant analysis.

on specific mill needs or issues. As previously cited in this research paper, PSAs and other stickies behave very differently in the process. Also, one can easily anticipate that PVA aqueous dispersion will behave very differently in the process compared to the hard hot melt glues. Those different behaviors within the process can now be tracked with the 3D-Stick.

Moreover, the interest around the results obtained with this new tool led the Centre Technique du Papier (CTP; Grenoble, France) and TechPap (Grenoble, France) to further develop an on-line measurement. A first prototype of such on-line sensor for stickies measurement has been built (**Fig. 11**), and initial industrial trials have been performed. This promising PI-Stick device allows following of macrocontaminant concentration and removal along the process for different stickies types.

CONCLUSION

The 3D-Stick is a new analytical tool for stickies characterization that will allow mills to:

1. Have quantitative, qualitative (nature), and morphological characterization of the macrocontaminant in pulp samples without any deformation.
2. Evaluate contamination modifications due to raw material changes.
3. Assess macrocontaminant removal efficiencies of each process step, per type of contaminant (PSA, hot melt, PVA, plastics, etc.).

The main advantages of this analytical tool are quantitative and qualitative measurements; fully automated, fast measurements (20 min for 300 objects); and a non-contact method (stickies not pressed).

As a perspective, CTP works closely with TechPap on an on-line measurement based on this 3D-Stick principle. This new, on-line fully-automated sensor would help mills follow up on stickies contamination at several points in their process, as well as address stickies removal. The final target is to perform a control-command of specific removal steps (screening rejects, flotation rejects) based on initial stickies contamination and final target. **TJ**

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ABOUT THE AUTHORS

Stickies have been an issue in the paper recycling process for decades, yet they remain a hot topic. However, until now, no clear differentiation between stickies by their chemical nature has been performed as part of the process. NIR analysis offers a very powerful tool for differentiating contaminants with close chemical composition, such as some stickies and plastics.

In this study, statistical analysis of the spectra was challenging, and gathering a team with the right competencies was critical to solving the issue. One interesting thing we discovered was that while the chemistry of glues seems similar all over the world, the breakdown of stickies from different countries can be drastically different, especially over different continents.

This study demonstrates how mills can analyze stickies removal throughout the recycling process and also control the process according to the chemical nature of incoming stickies contamination. Our next step is to develop on-line stickies measurements.

Ossard, Huber, Borel, and Delagoutte are project engineers and Soysouvanh is team leader at the Centre Technique du Papier (CTP), Grenoble, France. Email Huber at Patrick.Huber@webCTP.com.


Ossard


Huber


Borel


Delagoutte


Soysouvanh