EFFECTS ON FORMING WHEN USING ALIGNED MULTI WALL CARBON NANOTUBES IN MULTI-STACKED PREPREG

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ABSTRACT

Automated tape lay-up (ATL) combined with Hot Drape Forming (HDF) offer cost competitive manufacturing for large composite components. However, not carefully performed HDF of composite laminates could end up with out-of-plane fibre wrinkling. Previous studies with this technique has shown that the stacking sequence have a significant influence on wrinkle development during 3D-forming. One possible explanation to this might be the relatively high interply friction for the combination of [0] and [45] layers.

Prepregs containing thermoplastic toughener particles show a higher level of interply friction compared to prepregs which do not contains such particles. It is therefore likely that interfacial particles in general will increase the interply friction. Such particles could be thermoplastic toughener or aligned multiwall carbon nanotubes (MWCNT).

The aim of this study is to show how locally arranged MWCNTs in prepreg interlayers affect the global forming behaviour. An initial study of intraply shear and interply friction is performed with purpose to investigate how pregpreg with MWCNT interlayers on general influence the forming. Further an experimental forming study is performed with aligned MWCNT in the [-45] / [0] interlayers of a quasi-isotropic prepreg stacking sequence. A numerical study is also performed simulating the forming of the experimental spar geometry.

The results show that the intraply shear resistance of the MWCNT containing material is 100% higher than for the reference. Further the interply friction is 3 to 4 times higher for the MWCNT containing material compared to the reference. The experimental spar study shows increased out-of-plane wrinkling in the joggled spar flange when using MWCNT in the [-45]/[0] interlayers. The numerical study strengthen the experimental results by showing increased compression across the fibre direction in the [0] layer when adding contact surfaces in the [-45]/[0] interlayers.

1 INTRODUCTION

During the latest decade the use of structural composites has rapidly increased in commercial aircraft programs such as Boeing 787, Airbus 350 and Bombardier CSeries. The increased use of composites demands the manufacturing efficiency to heavily improve. This has driven the development of automated tape lay-up (ATL) and automated fibre placement (AFP). Both methods offer advantages for flat lay-ups in terms of high lay-up rates, capability to manufacture large parts and simplified offline machine programming [1]. The disadvantage with ATL is limited capability of producing complex geometries and the disadvantage with AFP is low lay-down rate for complex geometries. However, in addition a flat lay-up could be combined with multi-layer forming such as Hot Drape Forming (HDF), offering a cost competitive manufacturing route for large composite components.

With lack of knowledge of geometrical limitations of HDF, the laminates could easily end up with out-of-plane fibre wrinkles [2], [3]. Looking at a product involving doubly curved surfaces (such as a joggled spar) the case of wrinkle-free forming of a laminate having more than two unique fibre direction must invoke both interply slippage and intraply deformation [7]. Several studies [3], [4] have looked at wrinkling during forming of a recess area, such as a joggled spar flange area. It is concluded that out of plane wrinkling often occurs due to a combination of in-plane shear and ply compression [5]. Wrinkling is assumed to start in the ply with least stiffness in the load direction, i.e. the fibre direction perpendicular to the axial load [6].

Hallander et al. [3] showed a dependency between stacking sequence of UD prepreg laminate and wrinkle development when forming a spar geometry with a shallow recess area. This study touches on the dependency on interfacial coupling effects. An explanation to this might be the increased interply friction for the combination of [0] and [45] layers compared to other tested ply direction combinations [8]. The combination of [45] and [0] layer may also reduce the material´s ability to deform through shear [9].

Prepregs containing thermoplastic toughener particles shows a higher level of interply friction compared to prepregs which do not contains such particles [10]. It is therefore likely that interfacial particles in general will increase the interply friction. In [11] a study of forming prepreg composite parts with aligned multiwall carbon nanotubes (MWCNT) was performed. The MWCNT mat [12] used in this study was transferred to the prepreg surface to become MWCNT interlayers between the plies in the prepreg lay-up. The study showed that forming influences the MWCNTs. However, on the other hand, the MWCNTs probably also affects forming and the forming mechanisms, such as intraply shear and interply friction.

The aim of this study is to show how locally arranged MWCNTs in prepreg interlayers affect the global forming behavior.

2 EXPERIMENTAL

The presented work contains an initial study of intraply shear [13][14] and interply friction [8] [10] performed with purpose to investigate how prepreg with MWCNT interlayers generally influence the forming. Thereafter, an experimental study is performed where aligned MWCNT mats are transferred to prepreg and laid up in the [-45] / [0] interlayers of a quasi-isotropic stacking sequence. The MWCNT containing lay-up are thereafter formed using HDF to a spar geometry with a joggled flange area. Further an experimental study is performed where tool web friction is increased in order to study the influence of tool-part interaction on forming. An overview of all samples is presented in Table 1.

Sample ID	Lay-up	Geometry	Feature	Forming	Verification method
MWCNT 45/-45	[45, -45, 0, 90]s	Spar	MWCNT in recess flange [45, -45] interfaces in	HDF	Visual
Ref 45/-45	[45, -45, 0, 90]s	Spar	None	HDF	Visual
TF 8 [1)]	[45, 0, -45, 90]s	Spar	Increased tool friction web	HDF	Visual, micrograph
Ref 8 [2)]	[45, 0, -45, 90]s	Spar	None	HDF	Visual, micrograph
TF 32 [3)]	[(45,0,-45,90)4]s	Spar	Increased tool friction web	HDF	Visual, micrograph
Ref 32 [4)]	[(45,0,-45,90)4]s	Spar	None	HDF	Visual, micrograph
Bias Reference	[45,-45, -45, 45]	Flat	None	Bias Extension	Bias extension test
Bias MWCNT	[45,-45, -45, 45]	Flat	MWCNT in all interfaces	Bias Extension	Bias extension test
Friction Reference	[0]	Flat	None	-	Interply friction test
Friction MWCNT	[0]	Flat	MWCNT in all interfaces	-	Interply friction test

Table 1: Overview of experimental samples [1)] TF 8 = Tool Friction 8 layer, [2)] Ref 8 = Reference 8 layer, [3)] TF 32 = Tool Friction 32 layer, [4)] Ref 32 = Reference 32 layer

2.1 Material

A 180°C cure epoxy prepreg with unidirectional (UD) HT carbon fibre and approximately 57%

fibre volume content was used in the experiments. The prepreg had a CPT of 0.131 mm and did not contain any thermoplastic toughener particles in the matrix. Aligned MWCNTs grown on silicon wafers as described above, were used in the study. The MWCNTs had an approximately height of 20 microns [11], [12].

Further, a similar epoxy prepreg but containing thermoplastic toughener particles in the matrix was used in the tool web friction study. The interply friction was fairly equal for both prepreg systems.

2.2 MWCNT Transfer

Aligned MWCNTs were transferred from the silicon wafers to the prepreg surface where it was fixed to the matrix using a vacuum bag with controlled vacuum level and a heat source controlled by thermocouples. The samples were moderately heated during the MWCNT transfer. Transferred MWCNT area for spar forming is shown in Figure 1.

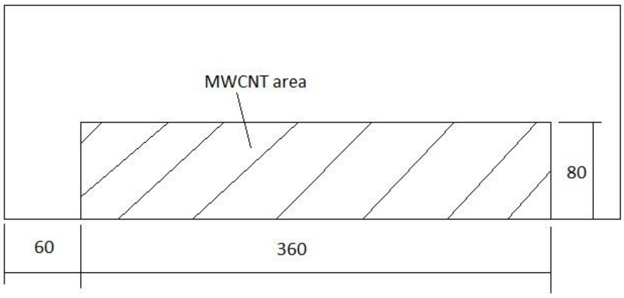

Figure 1 MWCNT area orientation in flat lay-up before HDF.

2.3 Intraply shear test

The intraply shear properties of cross-plied reference material (without MWCNT reinforcement) were compared to cross-plied material with interfacial MWCNT using bias extension testing [13], [14]. The sample lay-up was [45,-45,-45, 45] with a sample test area of 200 x 45 mm in-between grips. The MWCNT were prepared placing the MWCNT mats between all plies.

The bias extension testing was performed in an Instron machine with a dedicated oven to enable testing at elevated temperatures. Test speed 31 mm/min and test temperature 65 ˚C was chosen. Three reference material samples and four MWCNT containing material samples were tested with this parameter set up. Some more reference samples were also tested at temperature 55 ˚C

2.4 Interply friction test

The interply friction properties of the reference and the material with interfacial MWCNT were tested using the methods in [8], [10]. The test parameters are presented in table 2. Four samples of the reference and three samples of the MWCNT containing material were tested.

Property	Value
Temperature	65 ˚C
Crosshead speed	0.1 mm/min
Normal pressure	80 kPa
Relative fibre direction	[0]/[0]

Table 2: Interply friction test parameters

2.5 Forming of spars

The importance of [45]/[0] interface for the wrinkling behaviour was shown in [3] and the lay-up for the MWCNT containing spar forming study was therefore designed to isolate these combined layers to just two interfaces; see Table 1. Especially the interply friction seems to be important for wrinkle development. As the MWCNT was assumed to influence the interply friction, it was placed in the sensitive [45]/[0] interfaces. A joggled C-shape spar, sample *MWCNT 45/-45*, with aligned MWCNT in the [-45]/[0] interlayers was formed to evaluate the influence of MWCNT on the forming. To assure that the MWCNT areas would undergo different forming mechanism, a C-shaped spar with recess area was chosen. The spar geometry, also used in [3], is described in Figure 2 and Table 3. A sample, *Ref 45/-45*, without any MWCNT was also formed as a reference.

Further a tool friction study was performed on the same geometry as above. Joggled c-shaped spars were formed on a mould with increased surface roughness in the web; see Table 1. The mould web surface was grit blasted to Ra value 2.80 µm in order to increase the friction at the tool/part interface. Reference samples were formed to the reference mould with surface roughness Ra 0.39 µm.

Hot Drape Forming (HDF) was used to drape the pre-stacked material onto the mould. The lay-up was placed on top of the mould, where after the vacuum bag was loosely sealed on top of the lay-up. After heating up to 65°C, vacuum was applied forcing the material towards the mould. Vacuum was held until the lay-up temperature was at room temperature. Support walls were used during HDF and the forming temperature was controlled using a thermo-couple.

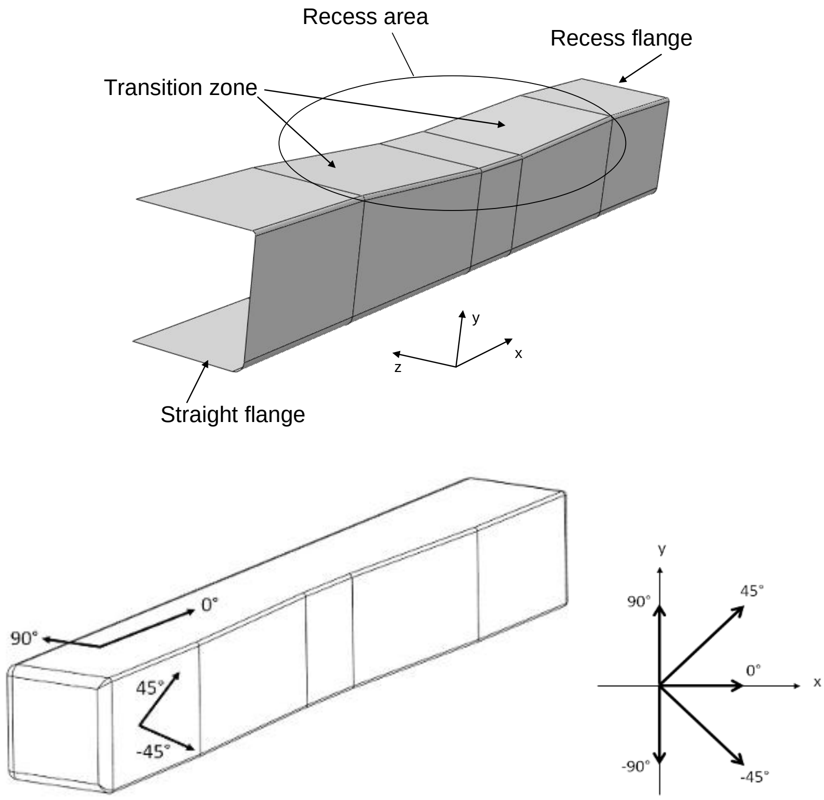

Figure 2: Spar geometry

Spar length [mm]	480
Web width [mm]	70
Flange length [mm]	55
Transition zone length [mm]	125
Recess depth [mm]	6.25
Nominal thickness [mm]	1.048 or 4.192
Radius recess flange [mm]	2
Radius Straight flange [mm]	6

Table 3: Spar geometry

2.6 Evaluation of C-Spar forming

All C-spars were visual inspected after forming and compared with the out-of-plane defect chart described in Figure 3. The C-spars in the tool-web friction study were cured in a standard aerospace autoclave process with a 6 bar pressure and a 2 h hold at 180 ˚C. The spars were thereafter cut in samples at the defects that visually differed from the references. The samples were cut perpendicular to the defect direction. The cut samples were polished and analysed by using micrographs.

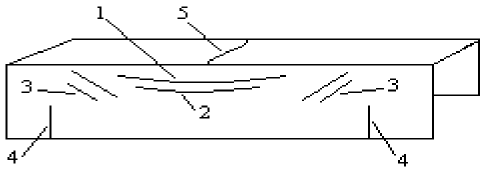

Figure 3: Definition of out of plane defect location

3 NUMERICAL STUDY

A numerical study was also performed simulating the forming of the experimental spar geometry using the methods in [15]. The software used for the study was Aniform, which is a Finite Element designed software for simulation of composite forming.

The in-plane behavior of the layer was modelled with linear elastic fibres. An elastic model and a viscous model were used in parallel to describe the matrix behavior. Fiber stiffness was implemented in one direction giving the laminate in-plane orthotropic behavior while the matrix was isotropic (see Table 4).

Linearly elastic fibre	
E[MPa]	1000
Viscoelastic matrix	
E[GPa]	0.0071
ν	0.33
η[GPa.s]	10.6

Table 4: Laminate in-plane properties

Considering out-of-plane properties, an orthotropic elastic model was used to simulate the difference in bending stiffness along and transverse the fibre direction (see Table 5).

Orthotropic bending	
E_1	100
E_2	0.95
ν	0.33
G_{12}	18.34

Table 5: Laminate out-of-plane properties

The friction behavior was modelled as a combination of viscous friction and Coulomb friction (see Table 6) working in parallel in the model. Friction models were calibrated for both friction in-between plies as well as for friction between ply and mould. The friction between the top layer and the diaphragm was assumed to be zero since a plastic film was placed between the diaphragm and the top ply. The friction in the MWCNT area of the joggled flange was modelled as an extra contact surface between the plies; see Figure 4. Based on the experimental observations, the viscoelastic friction at the contact surface was set to two times the viscoelastic friction in the plies. In total this added to three times as high viscoelastic friction for the MWCNT interfaces compared to the rest of the interfaces between the plies. The assumption of viscoelastic friction in the MWCNT surface was based on the experimental interply measurements made in this study.

Viscoelastic friction (HT)		Viscoelastic friction (Mould)		Viscoelastic friction (contact surface MWCNT area)	
μ	0.0116	μ	0.000579	μ	-
η	0.00710	η	0.0025	η	2 x 0.00710
t [mm]	0.01	t [mm]	0.01	t [mm]	0.01

Table 6: Friction behavior

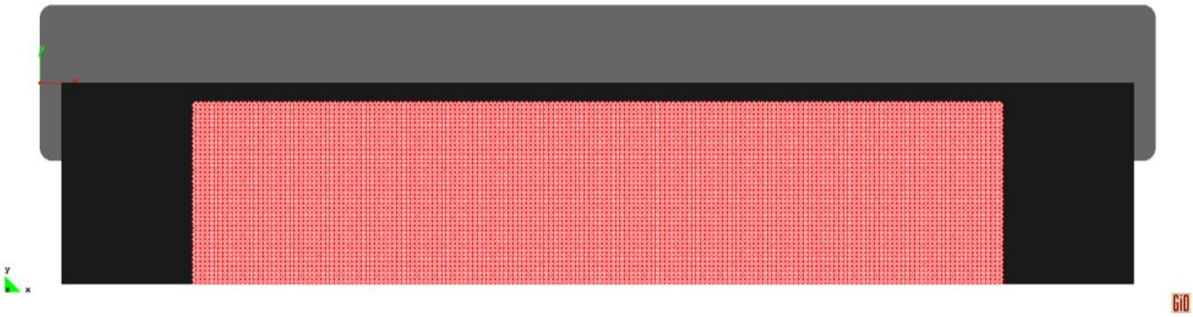

Figure 4: Added contact surface in the MWCNT area.

The rubber diaphragm was modelled using a Mooney-Rivlin material model with parameters in accordance with Table 7.

Mooney-Rivlin model	
C10	1.4265
C01	0.1953

Table 7: Rubber diaphragm properties

4 RESULTS

The results show that the intraply shear resistance of the MWCNT containing material is 100% higher than for the reference see; Figure 5. Further the interply friction is 3 to 4 times higher for the MWCNT containing material compared to the reference; see Figure 5.

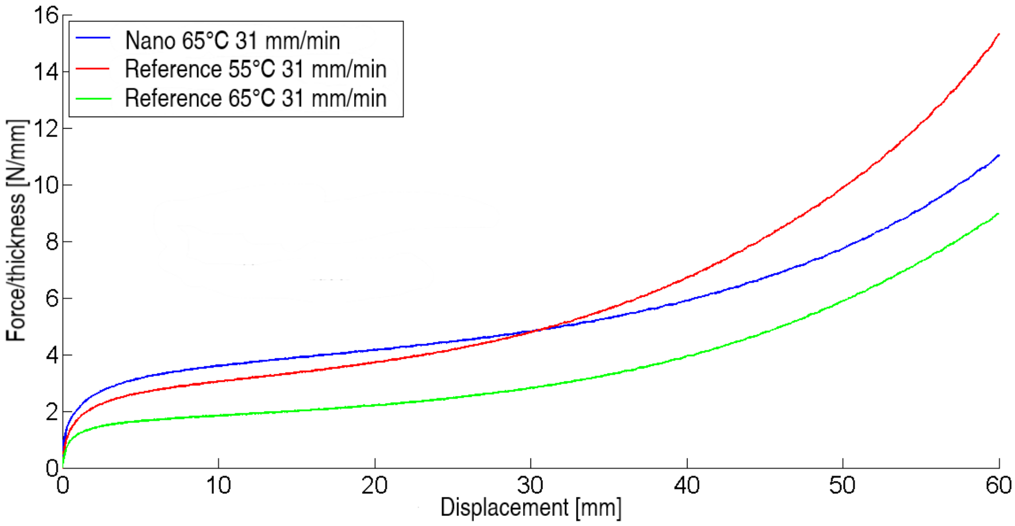

Figure 5a: Resulting load curves from intraply shear test.

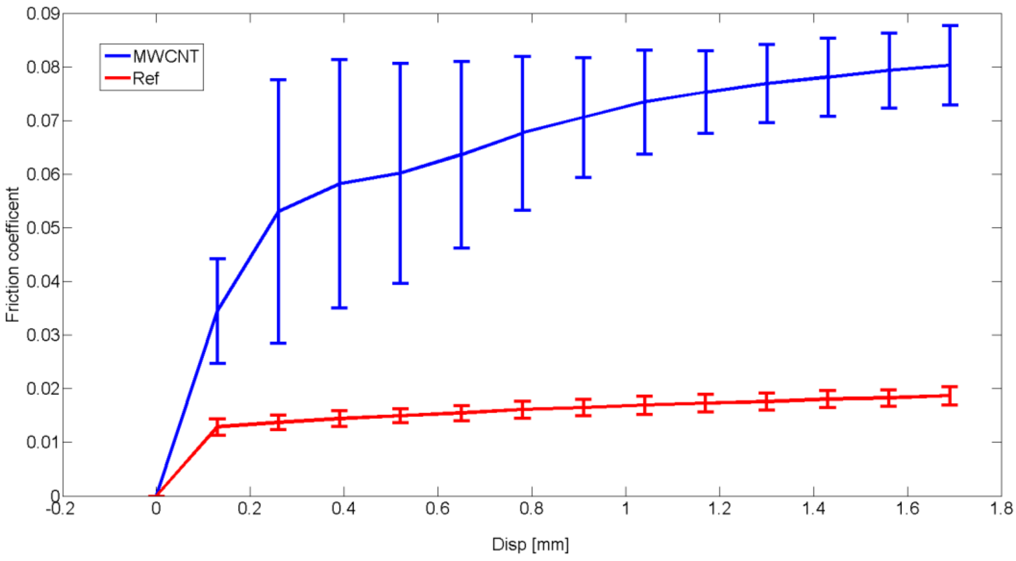

Figure 5b: Interply friction measurement graphs.

The results of all C-Spar forming experiments are summarised in Table 8. The experimental spar study, when using MWCNT containing [-45]/[0] interlayers, shows defect type 1 in the flange recess area and defect type 5 in the middle of the web for sample *MWCNT 45/-45*; see Figure 6. The reference sample *Ref 45/-45* did not show any visual defects.

Sample ID	Feature	Out of plane defect type
MWCNT 45/-45	MWCNT in recess flange [45, -45] interfaces in	1 and 5
Ref 45/-45	None	None
TF 8	Increased tool friction web	1 and 5
Ref 8	None	1 and limited 5
TF 32	Increased tool friction web	1,3, 4 and 5
Ref 32	None	1, 3 and 5

Table 8: Spar study observations

Figure 6: Out-of-plane wrinkling in the joggled area of a spar with a kvasi-isotropic lay-up with MWCNTs in the [-45] / [0] interlayers.

The numerical study shows a higher level of compression across the fibre direction in the flange recess area for layer 3 ([0] direction) when adding extra contact surfaces between the [0] and [45] layers; see Figure 7

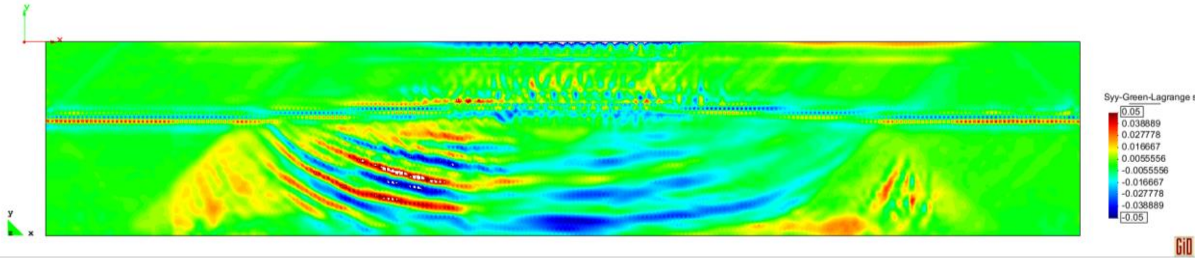

Figure 7a: Compression in terms of Syy in the joggle area of layer 3 [0] in the spar with kvasi-isotropic lay-up with added contact surfaces in the [-45] / [0] interlayers.

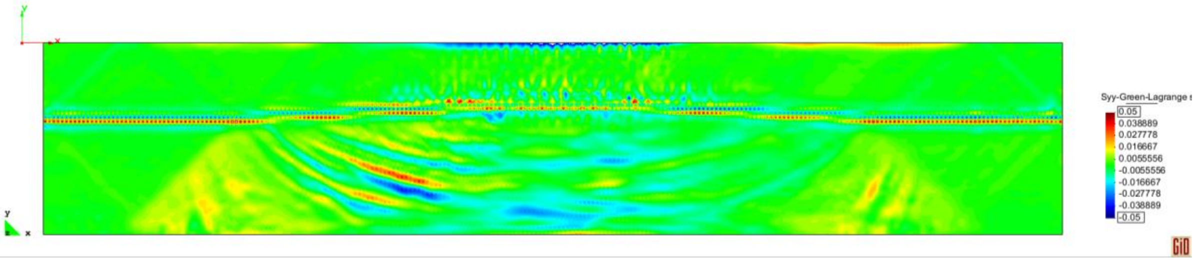

Figure 7b: Compression in terms of Syy in the joggle area of layer 3 [0] in the reference spar with kvasi-isotropic lay-up.

The 8 layer samples *TF 8* and *Ref 8* in the tool-web friction study shows the same type of defects; In this case only defect type 1 in the flange recess area and defect type 5 in the middle of the web occur. The level of defect type 1 is the same for both samples. However the web defect (type 5) is slightly decreased in *TF8* compared with *Ref 8*; see Figure 8. The 32 layer samples *TF 32* and *Ref 32* both show defect type 1 and 3 in the flange recess area and defect type 5 in the middle of the web. There are no remarkable differences in defect levels for these samples. However defect type 4 just outside the flange recess area did occur in the *TF 32* sample, but not in *Ref 32*.

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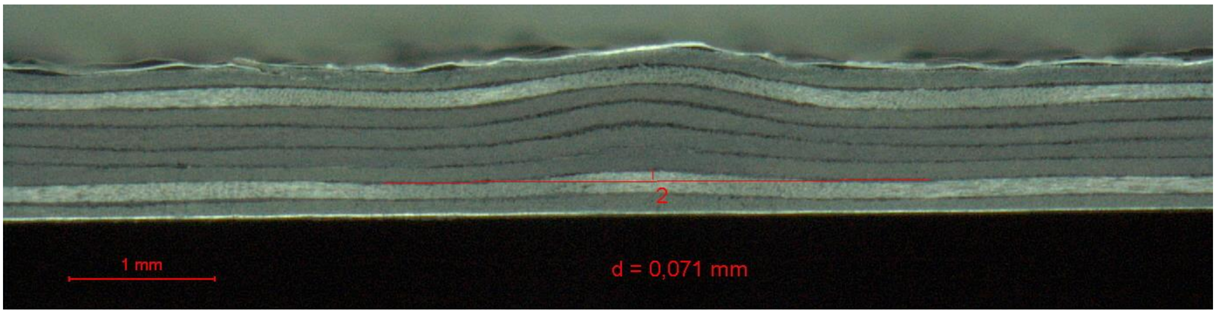

Figure 8a: Defect type 5 in sample *TF 8*

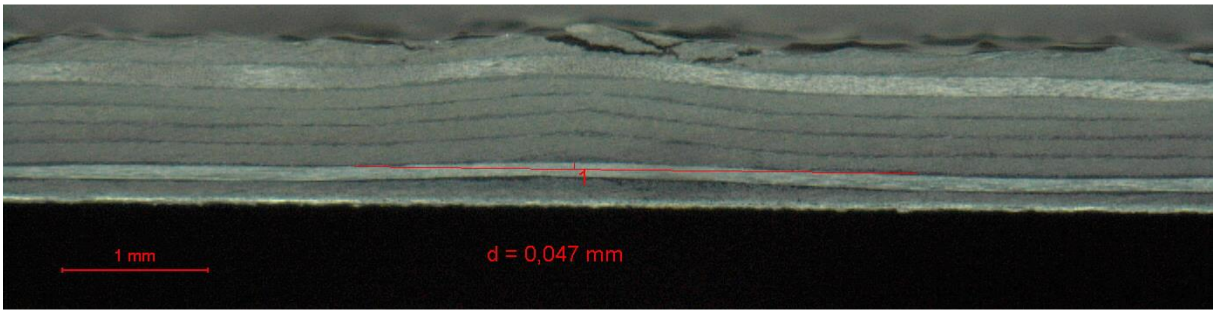

Figure 8b Defect type 5 in sample *Ref 8*

5 DISCUSSION

The experimental study shows that MWCNT containing interlayers clearly affects the interply friction and intraply shear of the prepreg. It is therefore likely that the MWCNT also affects the global forming behavior of the pre-stacked material.

In the experimental set-up of the MWCNT containing spar forming study it was assumed that the interply friction of the [0]/[45] interface was of great importance for the wrinkling behavior and that the interply friction was influenced by the MWCNT presence. The latter was also shown above. Since defect 1 and 5 did appear in the *MWCNT 45/-45* sample and not in the reference this proves that locally placed MWCNTs in the [0] / [45] prepreg interfaces will promote the development of out-of-plane defects. This also strengthens the theory that both interply friction between [0] and [45] layers and the lay-up sequence itself will have a strong impact on the out-of-plane defect development. The numerical study also supports this theory.

In general the tool-web friction study does not show any great difference in forming induced out-of-plane defect type and level. Especially not when increasing the number of layers. This indicates that other forming mechanisms [3] than the tool-part friction is more important for the out-of-plane defect development during forming. Such forming mechanisms could be the interply friction and intraply shear. However defect type 4 did appear in the *TF 32* sample just outside the recess area in the flange which was not the case for the reference. This indicates that the ability to move excess material just outside the transition zone towards the center [3] still might be effected by the tool/part friction.

6 CONCLUSIONS

The aim of this study was to show how locally arranged MWCNTs in prepreg interlayers affected the global forming behaviour.

The study shows that MWCNT containing interfaces will increase the interply friction and intraply shear which will affect the global forming behavior. Especially if the MWCNT are locally arranged in the [0]/[45] interfaces since these interfaces are of great importance for the wrinkling development. Further the study shows that tool-part friction is less important for the wrinkle development during forming.

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