

High-speed computed tomography to visualize the 3D microstructural dynamics of oil uptake in deep-fried foods

Corresponding Author: Dr Pieter Verboven

This file contains all reviewer reports in order by version, followed by all author rebuttals in order by version.

Version 0:

Reviewer comments:

Reviewer #1

(Remarks to the Author)

The paper titled "High-speed 4D computed tomography for uncovering the dynamics of oil uptake in deep-fried food products" presents interesting and novel work that helps advance knowledge on deep-frying and microstructural studies. In order to consider this work for publication, the authors must carefully address some major concerns I have.

Significant remarks I have are:

RESULTS

1. A significant omission in the entire paper is water. Frying is a dehydration process, and therefore, water is key. There is no attempt to distinguish water within the structure nor to determine the water evaporation rate and so forth. This is a major drawback and must be addressed.
2. Related to this, frying is a moving front evaporating process. Therefore, there must be a link between this process and structural changes. How do you link this fact with the description of the structure within the samples? For instance, you refer in Lines 179–182 to the existence of big pores within the core at 32 seconds. How would this happen? Would the moving front arrive at the core in 32 seconds to induce the formation of big pores? It would be valuable to measure the temperature within the samples to assess this aspect (in an equivalent benchtop setup). If this is not possible, it should at least be estimated. This is also relevant with respect to the reasoning about phase transitions in the different regions presented in Lines 191–203, and how this relates to evaporation kinetics (moving front), and therefore to temperature and the presence (or absence) of liquid water. This must also be referred to when cooling takes place, to give an idea of the time frame that must elapse before oil absorption begins, the kinetics of oil penetration, and so forth (section starting in Line 254).
3. Related to this, significant differences will certainly appear when comparing the structure for the same frying time at 120°C, 150°C, and 180°C, since there are completely different moisture loss kinetics. So, in order to make pertinent comparisons, the structure at a certain moisture loss obtained after frying at 120°C, 150°C, and 180°C should be compared, rather than what happens at a fixed frying time. I mean, it can be done, but conclusions will be partial.
4. How reproducible are the results? I ask this because major statements are derived from the figures shown, which correspond to limited samples. A good effort is made in Table 1, and it should be better exploited. On the other hand, when discussing the inner microstructure, which is very relevant in the paper, an equivalent effort must be made. Interesting results are presented in Table S1 for structure thickness. I would recommend analyzing this information, differentiating the crust and the core, to provide numerical data to support the statements (paragraph starting in Line 153), and give them statistical support (other parameters can also be analyzed).

DISCUSSION

1. Overall, the discussion should be almost completely rewritten. There is too much redundancy with the introduction and results sections, and it is hard to see a real discussion in this section.
 - o Lines 295–338: Redundant, very extensive, and no real discussion is presented.
 - o Lines 352–258: "The results from this study suggest that low-temperature deep-frying operations can be employed as a pre-processing technique in a two-stage deep-frying process. In the first stage, deep-frying at lower temperatures forms a porous microstructure with limited connectivity at the product surface, thereby hindering oil uptake and limiting the formation of harmful by-products. This is followed by deep-frying at a higher temperature to tailor microstructural properties and achieve the desired textural quality in a crispy exterior with a rigid crust." This cannot be derived at all from this work; it has nothing to do with a two-stage frying process, which is a completely different experimental setup.

o Line 374: The rate of moisture loss is not addressed in this study, so it cannot be referred to as such in the discussion (and actually should be, as I pointed out earlier).

MATERIALS AND METHODS

1. The main concern I have is related to the thresholds used to differentiate the different phases (Solid, Oil, and Gas). This is key, as it is the basis for the work presented. How did you verify that the protocol you defined was accurate? For instance, did you examine the different components (e.g. oil, solids) on their own to verify the protocol defined? What happened with water? You lumped it into the solids. Is this correct? Is there any chance to segment it? In this section, it is very important to develop the importance of X-ray attenuation, and elaborate on this, with respect to your components (not really density!) and how you can justify it accordingly, with much more depth than what is actually described. This must be developed as it is critical (Lines 145–152 and 486–491 in M&M).

2. Lines 393–394: “with perforations (0.7 mm diameter) along the surface to allow easy exchange of fluids during deep-frying.”

o Explain the perforation setup (number of holes, frequency/pattern, % of open surface, etc.). This is critical.

3. Lines 459–464: If I understood correctly, it took 1 second to acquire each tomograph, didn't it?

FIGURES

• Figure 2c: I cannot really see: “(2) localized oil ingress through ruptured walls of open pores at the surface during the late stages of deep-frying” (Lines 208–209: “Even when oil was detected in pores near the surface, it was unable to penetrate further into the sample indicating a lack of connectivity”).

• Figure 2c: Numbers 1 and 2 cannot be read properly in the figure. Differentiate them better.

• Figure 3: What is the scale? 500 microns?

• Figure 3: Images are every 4 seconds, not every 3 seconds. Figure 3b is every 11 seconds, not 10.

• Figure 3: Lines 174–178: I am not sure this can be stated so definitively. Also, I cannot really judge whether it is a phenomenon that starts from the core or not, as I do not know where this tiny region of the sample is located.

• Figure S2: Lines 250–252: “The minimal pore connectivity between the crust and the core due to a strong wall network prevented large amounts of oil from penetrating deeper into the structure, with most of the oil identified in small, compressed pores near the surface.” I do not really identify the strong wall network.

• Figure 7: In order to compare the effect of different frying temperatures, it would be suitable to compare the structure for the same moisture loss. Is this possible? Or at least report the moisture loss at each time (physically determined in a parallel experiment), so this can be discussed? According to the images, I would suggest that they could be similar, that is, 23 seconds at 150°C could be closer to 56 seconds at 120°C, and so forth.

Reviewer #2

(Remarks to the Author)

The topic of the manuscript is not very novel, as the dynamics of oil uptake in deep-fried food products have been studied by many authors, also using X-ray microcomputed tomography. However, the use of time-resolved 3D X-ray computed tomography makes it interesting. The design of the experiment and description of methods are not well-prepared, and this part needs to be improved. Additionally, some changes should be made in the next part - results and discussion.

1. Abstract: What is the definition of a “two-stage deep-frying operation”? Actually, the samples in this study were fried at temperatures of 120, 150, or 180 °C on a single occasion.

2. In this study, why was wheat-flour dough selected as the study object? Does it have universal applicability?

3. How does the 4D imaging system improve our understanding of the theories related to the deep-frying process?

4. How does the pore network integrity affect oil uptake in fried foods?

5. As you know, frying is a rapid process, especially during the initial seconds of immersing the sample in the frying oil. Conducting 3D X-ray computed tomography testing is a lengthy process, involving the exposure of samples over 360° and the acquisition of over 1000 radiographs. The precision of the time-resolved data appears to be lacking.

6. In order to further confirm the precision and reproducibility of the analysis of image processing for reconstructed images, additional methodologies for quantitative analysis of pore structure (nitrogen adsorption methods and mercury intrusion porosimetry) should be included.

7. Can this 4D computed tomography be applied to other types of cooking methods?

Reviewer #3

(Remarks to the Author)

1. Core Content: The paper “High-speed 4D computed tomography for uncovering the dynamics of oil uptake in deep-fried food products” explores the use of a 4D imaging system, specifically fast synchrotron radiation tomography, to visualize the highly dynamic processes occurring during the deep-frying and post-frying cooling of wheat flour dough (WFD) based food products. The study provides in-situ, time-resolved 3D imaging to understand the changes in the porous microstructure and

oil uptake dynamics. Key findings suggest that the pore network integrity significantly impacts oil absorption rates and distribution, and that a two-stage deep-frying process at different temperatures can reduce oil content while achieving the desired crust.

2. Limitations:

- a. Spatial and Temporal Resolution: The study utilizes high-speed synchrotron radiation tomography, but the resolution may still be insufficient to capture the smallest microstructural changes, particularly during the rapid phase transitions in the early stages of deep-frying.
- b. Sample Size and Representativeness: The research is based on WFD samples, which may not fully represent the diversity of deep-fried food products. The generalizability of findings to other types of food matrices is limited.
- c. Experimental Conditions: The controlled laboratory conditions, including the use of specific temperatures and sample holders, may not perfectly replicate real-world deep-frying scenarios. The influence of variations in oil type, frying duration, and food composition on the results was not extensively studied.
- d. Machine Learning Segmentation: The segmentation process, while advanced, introduces potential inaccuracies due to classification uncertainty, particularly in regions of phase transition and boundary determination.

3. Significance of Limitations:

- a. Spatial and Temporal Resolution: Importance: High - Accurate resolution is critical for understanding the detailed microstructural changes and their impact on oil uptake. Insufficient resolution can lead to incomplete or misleading interpretations of the dynamics involved. Explanation: Detailed imaging is essential to capture the nuances of pore formation, expansion, and oil ingress. Any loss in resolution could obscure critical transitional phases, affecting the study's conclusions about oil absorption mechanisms.
- b. Sample Size and Representativeness: Importance: Medium - While WFD provides a useful model, deep-fried foods vary widely in composition and structure. Explanation: Broader applicability requires testing on various food types. Limited sample diversity restricts the ability to generalize findings and apply them to different culinary practices or industrial applications.
- c. Experimental Conditions: Importance: Medium - Laboratory conditions provide control but may not fully mimic commercial frying environments. Explanation: Real-world variability in frying conditions, such as oil degradation, varying moisture content, and different frying techniques, could influence the microstructural changes and oil uptake dynamics, potentially altering the study's practical relevance.
- d. Machine Learning Segmentation: Importance: Medium - Segmentation accuracy is vital for quantitative analysis. Explanation: Errors in segmentation can lead to incorrect quantification of oil and gas phases, affecting the reliability of the data on oil uptake and microstructural changes. This limitation impacts the precision of the study's findings and their subsequent applications.

Conclusion: The study significantly advances the understanding of oil uptake dynamics in deep-fried foods through innovative 4D imaging techniques. However, limitations in resolution, sample diversity, experimental conditions, and segmentation accuracy must be addressed in future research to enhance the applicability and precision of these findings across a broader range of food products and real-world conditions.

Details:

- 1、 Abstract needs to be rewritten again. Lack data results
- 2、 Keywords failed to reflect the main aspects of the MS
- 3、 Which kind of deep frying products? Why choose wheat dough not French fries?
- 4、 What are the frying oils? Why choose sunflower frying oil not palm oil or others?
- 5、 The sample holder is not clear in Figure 1, please provide a clearer picture and mark the location where the sample is placed. In addition, the position of the thermocouple should also be marked.
- 6、 How are gas, oil and solid precisely defined in Figure 1D? Is there a basis for this definition?
- 7、 What do the different colors represent in Figure S2.
- 8、 In lines 252-253, no pore filling was observed in the core area? You can see pore in the central regions in Fig. S3.
- 9、 In lines 271-272, where is the crust microstructure of WFD deep-fried at 180°C ?
- 10、 In lines 276-279, the paper stated that the sample shrink to less than half of their initial gas porosity during the cooling period, which does not suitable for all fried samples, such as the samples fried in 180°C. What is structure thickness? In Table 1, the oil content and porosity of fried WFD are obtained from image analysis, which is somewhat subjective and can be added objective test indicators, such as the use of Soxhlet extraction to measure oil content and mercury meter to measure the porosity.
- 11、 In lines 287-290 and 338-340, there is a contradiction in the explanations of these two parts. The former states that structural deformation hinders oil absorption, while the latter states that deformation of pore space contributes to oil uptake. The structure should contain the pore.

Version 1:

Reviewer comments:

Reviewer #1

(Remarks to the Author)

Dear Authors,

I sincerely congratulate you on the excellent revision you have provided. The additional experiments, descriptions, and discussions have significantly improved the manuscript.

However, I have a few additional minor (but important) aspects that need to be addressed:

1) To carry out appropriate comparisons, the moisture content must be calculated on an oil-free and water-free basis, that is, mH₂O/m defatted-dried-solids), that is, the mass of solids exclude oil and moisture, to ensure a constant basis. Accordingly, the associated figure and discussion must be modified.

2) When presenting results on moisture loss, describe the possible mechanisms of water loss (would it be diffusion? Convection? Capillary flow? etc.?), and discuss them, according to the literature. Would there be any differences in the mechanisms and driving forces involved when working below or above the boiling point of water?

3) You specify that "In the case of WFD samples deep-fried at 120 °C and 150 °C, the difference between the temperature profiles in the crust and core were less pronounced." Why might this occur?

Reviewer #3

(Remarks to the Author)

The objectives and the rationale of the study have been clearly stated.

The limitations of current work should be elaborated upon in the text.

Version 2:

Reviewer comments:

Reviewer #1

(Remarks to the Author)

The authors have adequately addressed all queries, and thus, I consider the paper acceptable for publication.

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Response to reviewer's comments (NCOMMS-24-35988)

Title: High-speed 4D computed tomography for uncovering the dynamics of oil uptake in deep-fried food products

Authors: U. Verma, I. M. Riley, B. Lukić, L. Broche, P. Verboven*, J. A. Delcour, B. M. Nicolai

We thank the reviewers for taking their time to evaluate the manuscript and providing constructive comments. We have addressed all the comments and revised the manuscript accordingly. Below, we provide our responses to the referees' comments in blue, with major changes in the main text and supplementary information highlighted in red.

We made the following notable changes to the manuscript and supplementary information, which were not directly requested by the referees:

1. The order of the Figures has been updated after the addition of **Figure 4** (moisture content and temperature measurement plots). This change is reflected throughout the manuscript where applicable.
2. **Figure 5** and **Figure 8** in previous version of the manuscript have been combined into a single **Figure 6** in the updated manuscript for better comparison of the 3D volume renders across the different temperatures.

Reviewer #1 (Remarks to the Author):

The paper titled “High-speed 4D computed tomography for uncovering the dynamics of oil uptake in deep-fried food products” presents interesting and novel work that helps advance knowledge on deep-frying and microstructural studies. In order to consider this work for publication, the authors must carefully address some major concerns I have.

A: We thank the reviewer for their positive feedback on the relevance of this work. We sincerely appreciate the valuable suggestions, which have significantly contributed to the enhancement of this manuscript. We have responded to the individual comments and suggestions below.

RESULTS

1. A significant omission in the entire paper is water. Frying is a dehydration process, and therefore, water is key. There is no attempt to distinguish water within the structure nor to determine the water evaporation rate and so forth. This is a major drawback and must be addressed.

A: We agree that moisture loss from the food during deep-frying is a critical aspect of this process and was not discussed in detail within the manuscript. It is, however, not possible to distinguish between water and the solid matrix in the CT images. To address this comment, additional experiments were, therefore, conducted to evaluate the moisture loss from the sample at various stages of the deep-frying process. Using identical experimental conditions and setup as during the 4D μ CT, the moisture content was measured for the WFD samples deep-fried at different time points (for 5, 10, 15, 20, 30, 40, 50 and 60 s) and different oil temperatures (180 °C, 150 °C, 120 °C). While the time intervals do not follow the exact time intervals as 4D μ CT, these still cover the total duration of the deep-frying process. The results are presented in **Figure 4a** in the updated manuscript and discussed in the results and discussion sections in the manuscript.

Additions have been made to the following lines in the updated manuscript:

Lines 654-659: The moisture content of the WFD and deep-fried samples at 180 °C, 150 °C or 120 °C for 5, 10, 15, 20, 30, 40, 50 and 60 s was evaluated using the AACC method 44-15.02⁷⁹. Samples were deep-fried using the same experimental setup and cooled for 60 s before moisture content determination. The measurements were repeated for a total of six replicates for each of the time points. The moisture content was calculated on an oil free basis $m_{H_2O} (m_{H_2O} + m_{dough\ DM})^{-1}$, where mass (m) is expressed in grams and DM indicates dry matter.

Lines 184-192 The structure expanded during the early stages of deep-frying (i.e., between 1 s and 13 s) and was transformed into a highly porous solid matrix (**Fig. 3a**). Parts of the starch-gluten network were observed to rupture under continued expansion, thus forming large pores as smaller adjacent gas pores coalesced. **At this stage, the moisture content was approximately 35.2 % ($MC_{180^\circ C, 15s}$, **Fig. 4a**) indicating that the sample had lost a small portion of water. This thus substantiates that** persistent volumetric expansion induced by water evaporation and consequent enlargement of steam-filled pores inside the WFD solid structure increasingly exerted more forces on the internal walls resulting in thinning of the solid matrix.

Lines 201-202: Prolonged moisture loss ($MC_{180^\circ C, 30s}$ approximately 26.5 %, **Fig. 4a**) prompted the formation of a distinct region representing a crust-like structure after 34 s of deep-frying (**Fig. 2c**).

Line 206-209: Expansion inside the highly porous structure successively slowed down between 45 s and 56 s of deep-frying, which was attributed (in part) to the substantial loss of moisture (**final $MC_{180^\circ C, 60s}$**

approximately 15 %, Fig. 4a) and the increased structural rigidity of the biopolymers in the WFD system³³.

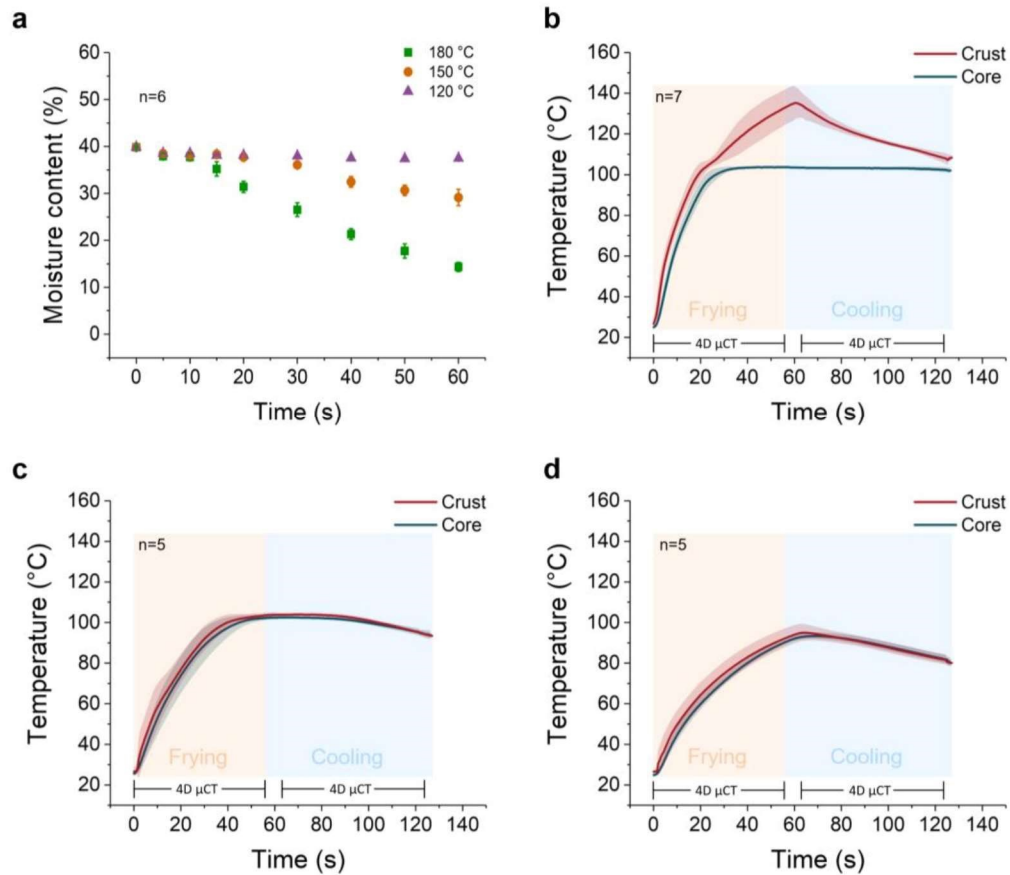


Figure 4. Moisture content and temperature measurements of WFD samples. (a) Moisture content of non-fried (0 s) and deep-fried (5 – 60 s) WFD samples with initial hydration levels of 40 % deep-fried at different temperatures (180 °C, 150 °C and 120 °C). Means and standard deviations (error bars) of six replicates are displayed; MC, % is calculated on an oil free basis $m_{H_2O} (m_{H_2O} + m_{dough\ DM})^{-1}$, where mass (m) is expressed in grams and DM indicates dry matter; Local temperature measurements inside the WFD samples during deep-frying and post-frying cooling at different temperatures (b) 180 °C, (c) 150 °C, and (d) 120 °C. Each plot presents mean temperature profiles in the crust and core regions inside the samples (shadows plot the standard deviation across five to seven replicates). The temperature measurements shown here corresponded to the deep-frying phase (orange shaded background) and post-frying cooling phase (blue shaded background), aligned with periods of 4D μ CT observations.

Line 310-315: As the internal temperatures of samples deep-fried at 120 °C did not reach the boiling point of water, the structure did not show signs of extensive deformation including thin wall rupture (Fig. S4a). The higher residual moisture content after deep-frying (final $MC_{120^\circ C, 60s}$ approximately 37.5 %, Fig. 4a) suggests that most internal regions still exist in a rubbery state, preventing any apparent rupture of the thin-walled structures in the core.

Lines 322-332: Oil temperature significantly affected the moisture loss kinetics in WFD samples due to differences in the rate of heat conduction (**Fig. 4**). Gradual internal structure expansion was observed at temperatures approaching the boiling point of water due to slow vapour generation, similar microstructural features were seen among samples deep-fried at 180 °C after 12 s (**Fig. 3b**), 150 °C after 23 s (**Fig. S4b**) and 120 °C after 34 s (**Fig. S4a**) with moisture content measuring at approximately 37 % in all samples (**Fig. 4a**). At temperatures around 100 °C, more violent structure deformations such as formation of large pores and thin wall rupture were induced by continuous evaporation and increasing vapour pressure inside the structure. This was apparent from the reconstructed images for samples deep-fried at 180 °C after 23 s (**Fig. 3b**) and at 150 °C after 45 s (**Fig. S4b**), which corresponded to similar moisture content values of 31.4 % ($MC_{180^{\circ}\text{C},20\text{s}}$) for 180 °C and 32.5 % ($MC_{150^{\circ}\text{C},40\text{s}}$) for 150 °C (**Fig. 4a**).

Line 333-341: After removal from the hot oil (i.e., 8 s of cooling), the WFD samples deep-fried at 120 °C and 150 °C preserved their porous microstructures (**Fig. 6**), thus resembling the structures observed at the final time point of deep-frying (**Fig. 8**). However, these samples progressively collapsed during the cooling period, shrinking to less than half of their initial gas porosity (**Table 1**) as the internal core collapsed (see Supplementary **Fig. S5** and **Table S1**). This phenomenon was likely related to the still rubbery state of biopolymers due to higher residual moisture (final $MC_{120^{\circ}\text{C},60\text{s}}$ approximately 37.5 %, $MC_{150^{\circ}\text{C},60\text{s}}$ approximately 29.1 % **Fig. 4a**) resulting from steam accumulation inside the core regions of the structures during deep-frying.

2. Related to this, frying is a moving front evaporating process. Therefore, there must be a link between this process and structural changes. How do you link this fact with the description of the structure within the samples? For instance, you refer in Lines 179–182 to the existence of big pores within the core at 32 seconds. How would this happen? Would the moving front arrive at the core in 32 seconds to induce the formation of big pores? It would be valuable to measure the temperature within the samples to assess this aspect (in an equivalent benchtop setup). If this is not possible, it should at least be estimated. This is also relevant with respect to the reasoning about phase transitions in the different regions presented in Lines 191–203, and how this relates to evaporation kinetics (moving front), and therefore to temperature and the presence (or absence) of liquid water. This must also be referred to when cooling takes place, to give an idea of the time frame that must elapse before oil absorption begins, the kinetics of oil penetration, and so forth (section starting in Line 254).

A: In response to this suggestion, additional experiments were conducted to measure the temperature inside the samples. The measurements were logged continuously during deep-frying (56 s) and post-frying (63 s) to match the 4D μCT imaging. Measurements were carried out separately for the ‘crust’ and the ‘core’ regions of the sample in the outermost regions of the structure (i.e., below the surface) and in the central region of the structure, respectively (**Figure S9** in Supplementary materials).

The results presented in **Figure 4b** (see reply to comment 1 in RESULTS – [page 3](#)) highlight a clear distinction between temperature profiles measured in the crust and core regions for samples deep-fried at 180 °C. This provides new insights into the changing temperature gradients within the structure during deep-frying, shedding light on the moving evaporation front and its effect on structure deformation.

Heat transfer by conduction inside the crust region causes temperatures to rise to the boiling point of water. In the WFD sample that was deep-fried at 180 °C, the effects of this temperature increase and consequent vapor formation corresponded to structural changes. The core experienced a delayed and slower heat conduction, reaching 100 °C after approximately 26 s of deep-frying. This coincided with the formation of large pores inside the core (around 34 s) due to rising vapour pressure inside the core from

continued steam generation (**Figure 4a** in main text). **Lines 201–206** (previously lines 179–182) are therefore substantiated in light of this new information.

Additions have been made to the following lines in the updated manuscript:

Lines 661-669: Temperature profiles inside the WFD samples during and after deep-frying were measured in the crust and core separately using a K-type thermocouple (0.075 mm probe diameter, RS-pro, London, UK) connected to a digital acquisition system (34970A, Agilent Technologies, Santa Clara, CA, USA) recording data at a frequency of 2 Hz. The measurements in the crust and core regions were conducted near the surface for the crust (within 1 mm) and at the centre of the sample for the core (see Supplementary **Figure S9**). Temperatures were logged continuously during deep-frying (56 s) and the post-frying (63 s) period, same as the 4D μ CT imaging. At least five replicates were obtained for each deep-frying temperature 180 °C, 150 °C or 120 °C to account for any variation in the placement of the thermocouple.

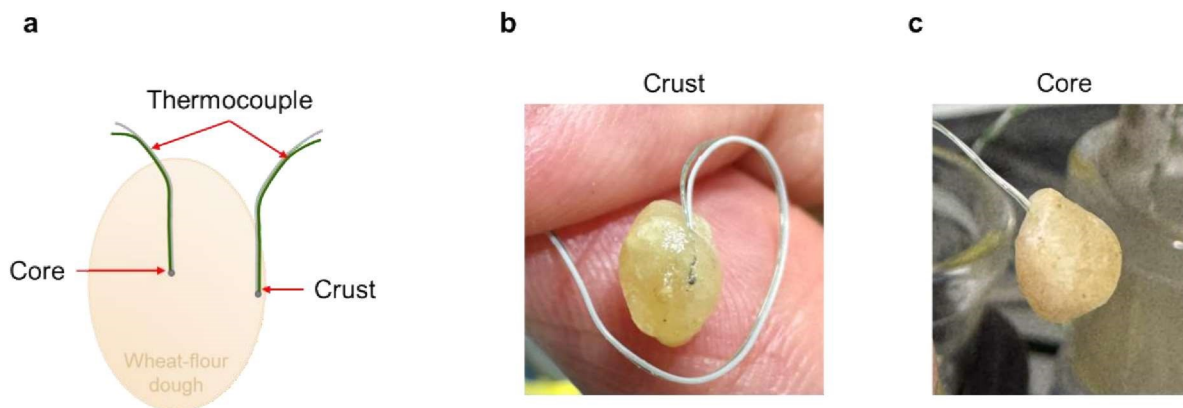


Figure S9. Thermocouple placement inside the WFD samples. **a** Schematic drawing of probe placement within the samples during frying; **b** Probe inserted in the crust region; **c** Probe inserted in the core region.

Lines 180-185: Through continued heat conduction inside the sample, the temperatures below the surface rapidly approached the boiling point of water, while heat conduction progressed more slowly into the centre (**Fig. 4b**). As a consequence of the increased steam generation and rising gas pressures, deformation was initiated by expansion of pre-existing pores, essentially serving as nucleation sites. The structure expanded during the early stages of deep-frying (i.e., between 1 s and 13 s) and was transformed into a highly porous solid matrix (**Fig. 3a**).

Lines 193-204: Between 19 s and 23 s, the temperatures in the crust plateaued at around 100 °C for a brief period before increasing at a slower rate thereafter (**Fig. 4b**). This brief period where temperatures in the crust plateaued suggests that the evaporation front advanced to the core regions once sufficient moisture was removed from the crust. Expansion in the core regions continued as temperatures level to 100 °C (**Fig. 4b**), simultaneously pushing the surface layers against the inner walls of sample holder (observed from 23 s onwards in **Fig. 3b** and **Fig. 2c**) indicated a high flexibility of the biopolymer constituents hampering vapour escape at the surface during the early stages of deep-frying^{11,55}. Prolonged moisture loss ($MC_{180^{\circ}\text{C},30\text{s}}$ approximately 26.5 %, **Fig. 4a**) prompted the formation of a distinct region representing a crust-like structure after 34 s of deep-frying (**Fig. 2c**). Temperatures steadily

increased above 100 °C in the crust, while stabilizing around the boiling point of water in the core (**Fig. 4b**).

Lines 223-229: Through heating, the temperature increased to the boiling point and water evaporated. Temperatures in the crust increased from 100 °C (25 s) and reached ~135 °C by the end of deep-frying. Continued loss of moisture and increasing temperatures below the surface led to a rigid crust after transitioning into a glassy state^{29,33}. In the core, temperatures stabilized at 100 °C with steam trapped inside the structure, suggesting that the amount of moisture in this region was sufficient for the biopolymers to remain in a rubbery state, thereby supporting structure expansion induced by steam generation.

In the case of WFD samples deep-fried at 120 °C and 150 °C, the difference between the temperature profiles in the crust and core were less pronounced. While the rate of change of temperature was higher in the crust compared to the core regions, due to the small sample size, heat conduction inside the core progressed at a similar rate.

Additions have been made to the following lines in the updated manuscript:

Lines 300-315: Similar to what had been shown when deep-frying at 180 °C, structure expansion initiated from the pre-existing gas cells present in the non-fried dough. However, gas cell sizes increased at a notably slower rate in these samples compared to those deep-fried at 180 °C (see Supplementary **Fig. S4**). This delayed expansion is attributed to the slower steam formation at lower deep-frying temperatures since heat conduction inside these samples progressed at a slower rate. Temperatures within the samples did not exceed 95 °C (**Fig. 4d**) and 100 °C (**Fig. 4c**) when deep-fried at 120 °C and 150 °C respectively. Internal thinning of the solid matrix continued from sustained expansion in the core regions. Comparing the microstructures of deep-fried (56 s) WFD samples at 180 and 120 °C, those deep-fried at 120 °C presented a more uniform pore morphology without a distinct crust (see Supplementary **Fig. S4a**). As the internal temperatures of samples deep-fried at 120 °C did not reach the boiling point of water, the structure did not show signs of extensive deformation including thin wall rupture (**Fig. S4a**). The higher residual moisture content after deep-frying (final $MC_{120^{\circ}\text{C},60\text{s}}$ approximately 37.5 %, **Fig. 4a**) suggests that most internal regions still existed in a rubbery state, preventing any apparent rupture of the thin-walled structures in the core.

Lines 322-332: See reply to comment 1 in RESULTS – [page 4](#)

Although these experiments indeed provide additional insights into the conditions related to structural transitions in the crust and core of the WFD samples, it is more difficult to individually distinguish between the state transitions for the starch and gluten constituents. Nonetheless, these conditions help substantiate the starch and gluten phase transitions of the WFD samples proposed in the first manuscript.

Additions have been made to the following lines in the updated manuscript:

Lines 214-232: Starch and gluten in the samples progressively undergo structural transformations during deep-frying⁵⁶. In the WFD system, despite starch being a major component, the structure is primarily supported by the viscoelastic gluten network due to the extensive structural changes that occur during deep-frying^{33,57}. However, it was recently shown that the transition of potato starch (in potato starch-water model systems) from the rubbery state, after gelatinization, to the glassy state, is necessary to maintain the expanded structure due to frying-induced water evaporation¹⁴. Changes in the moisture content of the sample at different stages of deep-frying, together with temperature measurements in the crust and core, provide additional insights into the conditions underlying

structural transitions inside the WFD system. Through heating, the temperature increased to the boiling point and water evaporated. Temperatures in the crust increased from 100 °C (25 s) and reached ~135 °C by the end of deep-frying. Continued loss of moisture and increasing temperatures below the surface led to a rigid crust after transitioning into a glassy state^{29,33}. In the core, temperatures stabilized at 100 °C with steam trapped inside the structure, suggesting that the amount of moisture in this region was sufficient for the biopolymers to remain in a rubbery state, thereby supporting structure expansion induced by steam generation. Image data depicting structure expansion during deep-frying reveals minimal structure alteration of hardened surface regions while the central regions continually deform (Fig. 3), indicating a difference between the physical states of crust and core regions at the end of 56 s of deep-frying.

Lines 310-315: As the internal temperatures of samples deep-fried at 120 °C did not reach the boiling point of water, the structure did not show signs of extensive deformation including thin wall rupture (Fig. S4a). The higher residual moisture content after deep-frying (final $MC_{120^{\circ}\text{C},60\text{s}}$ approximately 37.5 %, Fig. 4a) suggests that most internal regions still exist in a rubbery state, preventing any apparent rupture of the thin-walled structures in the core.

Line 333-343: After removal from the hot oil (i.e., 8 s of cooling), the WFD samples deep-fried at 120 °C and 150 °C preserved their porous microstructures (Fig. 6), thus resembling the structures observed at the final time point of deep-frying (Fig. 8). However, these samples progressively collapsed during the cooling period, shrinking to less than half of their initial gas porosity (Table 1) as the internal core collapsed (see Supplementary Fig. S5 and Table S1). This phenomenon was likely related to the still rubbery state of biopolymers due to higher residual moisture (final $MC_{120^{\circ}\text{C},60\text{s}}$ approximately 37.5 %, $MC_{150^{\circ}\text{C},60\text{s}}$ approximately 29.1 % Fig. 4a) resulting from steam accumulation inside the core regions of the structures during deep-frying. Core regions lacking sufficient rigidity were unable to maintain the expanded shape as steam condensation induces negative vapour pressures due to reducing temperatures during cooling^{2,11,60}.

Another outcome from the separate temperature measurements inside the crust and the core regions was observed with respect to the differences in rate of temperature drop during the post-frying cooling period. For WFD samples deep-fried at 180 °C, the temperature inside the crust rapidly dropped during cooling; however, the temperature remained above 100 °C at the end of 63 s. This suggests that the observed oil uptake was primarily driven by capillary action in the small crust pores as the vapour pressures inside these pores could not go below the atmospheric pressure. In light of this finding, the subsection 'Post-frying oil uptake' has been modified to reflect the same.

Additions have been made to the relevant sections in the manuscript.

Lines 260-264: After removing the samples from the frying oil, the internal temperatures began to drop. The rate of temperature decrease varied significantly between the crust and core, with the crust cooling faster than the core (Fig. 4b). It is of note that the core temperatures did not fall below 100 °C during the post-frying cooling phase assessed here (i.e., 63 s), as the crust insulated the core regions.

Lines 272-282: It has been reported that, as temperatures drop, gas expulsion from the surface stops due to negative gauge pressures induced by condensing vapours^{12,59}, which subsequently pulls oil into open pores at the surface. However, in the WFD samples the temperatures stayed above 100 °C in both the crust and core regions at the end of 63 s during cooling. Although the oil uptake associated with the vacuum effect from condensing vapours cannot be completely ruled out, it is more likely that the observed oil uptake was primarily driven by capillary action in the small crust pores, as the vapour pressures inside these pores could not drop below the atmospheric pressure. In tortilla chips, oil

content was also found to be primarily influenced by small pores exerting great capillary action before condensation occurs²⁶. The crust comprising of mostly small, connected pores (pore size distribution shown in **Fig. S7a**) was saturated with oil²⁵.

In the case of WFD samples deep-fried at 150 °C and 120 °C, temperature for the two positions reduced at the same rate. After 150 °C frying, a plateau of 100 °C was observed before temperature decline indicating continued evaporation during cooling. The quick temperature drop after deep-frying at 120 °C coincided with the observed structure collapse (see **Figure 6b** and **Table 1** in the main text).

Additions have been made to the following lines in the updated manuscript:

Lines 359-362: Similar to the samples deep-fried at 180 °C, most of the oil absorption in samples deep-fried at lower temperatures occurred during cooling. Changing capillary pressures in crust pores and vapour condensation from temperatures decreasing below 100 °C inside the sample during cooling are both expected to have contributed to oil uptake.

These new results refine our previous conclusions on oil uptake dynamics across the samples deep-fried at different temperatures. We would like to thank the reviewer for this suggestion as this has led to an overall improvement of the manuscript.

3. Related to this, significant differences will certainly appear when comparing the structure for the same frying time at 120°C, 150°C, and 180°C, since there are completely different moisture loss kinetics. So, in order to make pertinent comparisons, the structure at a certain moisture loss obtained after frying at 120°C, 150°C, and 180°C should be compared, rather than what happens at a fixed frying time. I mean, it can be done, but conclusions will be partial.

A: We appreciate the reviewer's insightful suggestion to compare structural deformation based on equivalent moisture loss across different frying temperatures. The moisture loss kinetics were indeed different at 120 °C, 150 °C, and 180 °C; this was also evident from **Figure 4a** (see reply to comment 1 in RESULTS – [page 3](#)).

To address this comment, in addition to **Figure 4a**, where possible, we have reported the moisture content at each timepoint together with structural observations to draw any associated comparisons (see reply to comment 1 in RESULTS – [page 2](#)). We hope this added detail would allow readers with additional information in the manuscript to interpret structural changes alongside the moisture content trends across different temperatures.

Comparison of structural features are made to the following lines in the results section of the manuscript:

Lines 322-332: See reply to comment 1 in RESULTS – [page 4](#)

4. How reproducible are the results? I ask this because major statements are derived from the figures shown, which correspond to limited samples. A good effort is made in Table 1, and it should be better exploited. On the other hand, when discussing the inner microstructure, which is very relevant in the paper, an equivalent effort must be made. Interesting results are presented in Table S1 for structure thickness. I would recommend analyzing this information, differentiating the crust and the core, to provide numerical data to support the statements (paragraph starting in Line 153), and give them statistical support (other parameters can also be analyzed).

A: The results presented in the manuscript are based on observations and quantification across three repetitions performed for each of the different temperatures (120 °C, 150 °C, and 180 °C) and timepoints. We have also mentioned instances where failed reconstructions prevented segmentation (denoted by ^tn=2 in **Table 1** and **Table S1**). All numerical data has been presented with the mean values and their standard deviation.

We agree with the reviewer that statistical support can be added to the presented data, therefore, we have performed statistical analysis using multiple comparisons to determine the combined effect of interactions between the temperature and frying time for the presented numerical data, **Table 1** and **Table S1** have been updated to reflect the same. The new temperature and moisture data also indicate good replicability of the results.

Lines 671-676: The results are expressed as mean and standard deviation of replicates. To determine whether the means were significantly different, a mixed model with one-way ANOVA was performed to determine the combined effect of frying temperature and time. A multiple comparisons test was performed using Tukey's honest significant difference for all pairwise comparisons with the level of significance set at $p < 0.05$. All statistical analyses were performed using JMP pro 17.2 (SAS Institute, Cary, NC, USA).

Table 1. Oil content and porosity of fried wheat flour dough (WFD) formulation with 40 % moisture content evaluated during post-frying cooling.

Oil temperature	Cooling time (s)	Volume fraction of oil (% oil/sample volume)	Volume fraction of oil (% oil/solid)	Gas porosity (%)	Total porosity (%)
180 °C	8	4.47 (2.72) ^{cd}	7.90 (4.67) ^d	44.20 (1.90) ^a	48.67 (1.77) ^{ab}
	19	4.32 (1.65) ^{cd}	7.76 (2.79) ^{de}	45.01 (2.18) ^a	49.33 (0.79) ^{ab}
	30	9.28 (0.75) ^b	16.39 (1.05) ^{bc}	43.43 (1.55) ^a	52.71 (1.11) ^a
	41	8.55 (0.08) ^b	14.97 (0.40) ^c	42.87 (1.12) ^a	51.42 (1.18) ^a
	52	12.33 (0.40) ^{ab}	22.30 (0.78) ^{ab}	44.68 (1.23) ^a	57.02 (1.21) ^a
	63	14.42 (1.34) ^a	24.04 (1.76) ^a	40.09 (1.25) ^{ab}	54.51 (0.17) ^a
150 °C	8	8.12 (1.13) ^{+bc}	11.66 (1.82) ^{+cd}	30.16 (1.22) ^{+bc}	38.28 (2.35) ^{+bcc}
	19	10.66 (0.79) ^{+ab}	14.70 (1.72) ^{+cd}	27.09 (3.15) ^{+cd}	37.75 (3.94) ^{+bcc}
	30	10.73 (1.12) ^{ab}	13.06 (1.87) ^{cd}	17.40 (3.37) ^{def}	28.12 (4.43) ^{cd}
	41	11.88 (1.02) ^{ab}	13.87 (0.30) ^{cd}	14.42 (5.94) ^{ef}	26.30 (4.95) ^{cd}
	52	12.22 (0.43) ^{ab}	14.24 (0.63) ^c	13.97 (5.39) ^{ef}	26.20 (5.09) ^{cd}
	63	12.20 (0.90) ^{ab}	14.29 (1.31) ^c	14.36 (4.60) ^{ef}	26.56 (4.75) ^{cd}
120 °C	8	1.15 (0.34) ^d	1.53 (0.51) ^{ef}	24.13 (3.71) ^{cde}	25.28 (3.93) ^{de}
	19	1.08 (0.34) ^d	1.30 (0.41) ^f	17.29 (2.36) ^{def}	18.36 (2.37) ^{def}
	30	1.09 (0.45) ^d	1.25 (0.51) ^f	13.35 (1.74) ^{ef}	14.44 (1.51) ^{ef}
	41	1.18 (0.57) ^d	1.32 (0.62) ^f	11.39 (1.65) ^f	12.58 (1.21) ^f
	52	1.29 (0.65) ^d	1.43 (0.70) ^f	10.68 (1.48) ^f	11.96 (0.96) ^f
	63	1.28 (0.71) ^d	1.41 (0.76) ^f	10.21 (1.48) ^f	11.49 (0.84) ^f

Values represent the mean ($n = 3$, ^t $n = 2$) with standard deviation presented in brackets. Values in the same column are significantly different ($p < 0.05$) when not sharing the same lower-case alphabetic character.

Table S1. Average structure thickness (evaluated for the sample and the separated crust and core regions) of the solid matrix for WFD samples deep-fried at 180, 150 and 120 °C during post-frying cooling.

Oil temperature	Cooling time (s)	Average structure thickness - Sample (μm)	Average structure thickness - Crust (μm)	Average structure thickness - Core (μm)
180 °C	8	110.20(4.52) ^a	98.72(1.23) ^{def}	116.35(7.19) ^{bcde}
	19	102.16(3.95) ^a	93.95(1.70) ^{efg}	106.54(5.77) ^{cdef}
	30	95.83(3.85) ^a	88.45(2.17) ^{fg}	100.08(5.54) ^{def}
	41	86.21(3.52) ^a	80.03(2.85) ^g	89.56(4.44) ^f
	52	88.72(3.66) ^a	83.91(0.45) ^{fg}	92.85(4.64) ^{ef}
	63	96.50(5.00) ^a	87.00(6.31) ^{fg}	100.16(5.99) ^{def}
150 °C	8	119.66(6.00) ^{†a}	102.02(5.75) ^{†def}	125.48(7.39) ^{†abcd}
	19	122.55(6.65) ^{†a}	111.95(5.31) ^{†cde}	126.50(6.29) ^{†abcd}
	30	127.96(6.16) ^a	114.01(4.21) ^{cd}	133.68(6.06) ^{ab}
	41	128.69(10.03) ^a	111.48(6.51) ^{cd}	136.36(10.80) ^{ab}
	52	131.07(10.29) ^a	112.01(5.81) ^{cd}	139.74(11.42) ^{ab}
	63	131.15(10.06) ^a	111.61(5.33) ^{cd}	140.18(11.36) ^{ab}
120 °C	8	129.20(1.97) ^a	124.22(1.81) ^{bc}	128.08(2.13) ^{abc}
	19	136.99(1.90) ^a	132.80(3.47) ^{ab}	134.71(1.10) ^{ab}
	30	142.91(3.67) ^a	138.21(5.09) ^{ab}	140.54(2.38) ^{ab}
	41	146.77(3.93) ^a	141.31(5.13) ^{ab}	144.74(2.64) ^a
	52	146.87(3.69) ^a	141.18(4.67) ^{ab}	145.02(2.59) ^a
	63	147.38(5.50) ^a	141.55(6.47) ^a	145.64(4.28) ^a

Average structure thickness evaluated on a crust thickness of 0.5 mm and the remaining core regions. Values represent the mean ($n = 3$, $^†n = 2$) with standard deviation presented in brackets. Values in the same column are significantly different ($p < 0.05$) when not sharing the same lower-case alphabetic character.

Lines 360-365: Changing capillary pressures in crust pores and vapour condensation from temperatures decreasing below 100 °C inside the sample during cooling are both expected to have contributed to oil uptake. The samples deep-fried at 120 °C experienced significantly lower ($p < 0.05$) oil uptake across all samples due to the limited interconnectivity of pores at the surface due to lack of a distinct crust formation during deep-frying (Fig. 7c).

Based on the reviewer's suggestion, the microstructural differences between the crust and core regions of the deep-fried samples were investigated. Considering a constant thickness of 0.5 mm, the crust region was separated from the core to compare structure thickness and pore size distributions (evaluated at 8 s and 63 s). The results are presented in Figure S6 and Figure S7 in Supplementary information and Table S1. It should be noted that the quantitative analysis was only possible on the segmented dataset during post-frying cooling, therefore, the results have been added to the relevant sections in results.

Additions have been made to the following lines in the updated manuscript:

Lines 256-259: The morphological properties of the crust (considered as a 0.5 mm thickness subsurface layer) and core (remaining internal regions) was evaluated by using local structure thickness of the solid gas cell walls (presented in Supplementary Fig. S6 and Table S1) and pore size distributions (Fig. S7a).

Lines 276-282: Although the oil uptake associated with the vacuum effect from to condensing vapours cannot be completely ruled out, it is more likely that the observed oil uptake was primarily driven by capillary action in the small crust pores, as the vapour pressures inside these pores could not drop below the atmospheric pressure. In tortilla chips, oil content was also found to be primarily influenced by small pores exerting great capillary action before condensation occurs²⁶. The crust comprising of mostly small, connected pores (pore size distribution shown in **Fig. S7a**) was saturated with oil²⁵.

Lines 290-292: During deep-frying the pore-space continually expanded in the core region and compressed pores in the outer regions of the structure, resulting in a higher frequency of smaller pores in the crust (see Supplementary **Fig. S7a**).

Lines 344-358: Analysis of the morphology in the separated crust and core regions in comparison to WFD samples deep-fried at 180 °C reveals significant differences ($p < 0.05$) in the structure collapse behaviour of WFD samples deep-fried at 120 °C and 150 °C (**Table S1**). The frequency of smaller pores (0 – 50 µm) increased significantly after collapse in both crust and core regions (**Fig. S7b, c**). In samples deep-fried at 120 °C, the distributions of local cell wall thickness in the ‘crust’ and ‘core’ regions were virtually indistinguishable (**Fig. S6**). The average structure thickness in the evaluated ‘crust’ regions was significantly different ($p < 0.05$) compared to the crust regions of the samples deep-fried at 150 °C and 180 °C (**Table S1**), further supporting the observation that no distinct crust was formed in these samples during deep-frying. In contrast, in the samples deep-fried at 150 °C, the average structure thickness of the crust at 8 s was not significantly different ($p > 0.05$) from the crust in samples deep-fried at 180 °C at 8 s (**Table S1**), confirming the presence of a rigid crust. Furthermore, during collapse, the shift in the distribution of local structure thickness towards thicker pore walls was notably slower in the crust than in the core, indicating that portions of the crust were more rigid than the core regions.

Lines 643-652: To investigate the microstructural differences between the crust and core regions of the deep-fried samples, an outer layer with a constant thickness of 0.5 mm was separated at the surface using the chamfer distance map in Avizo. The crust and core regions were masked from the segmented dataset and pore size distribution was evaluated at 8 s and 63 s during the post-frying cooling following a watershed separation. Local structure thickness distribution (expressed in µm) of the starch-gluten matrix was evaluated for the sample and the separated crust and core regions using CTAn (Version 1.18.8.0, Bruker SkyScan, Kontich, Belgium). The sphere-fitting algorithm in the 3D analysis module provided the local thickness of the structure as represented by the diameter of the largest enclosed sphere⁷⁸.

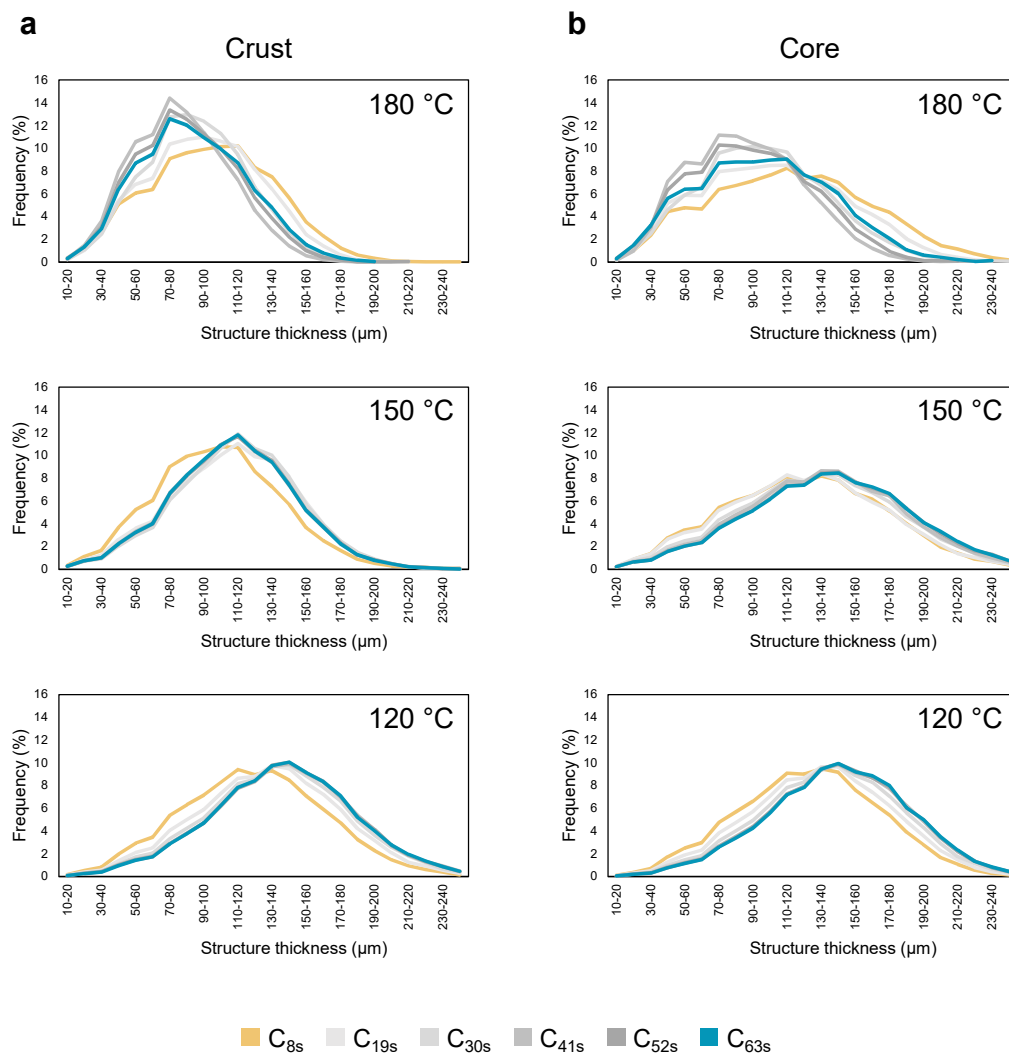
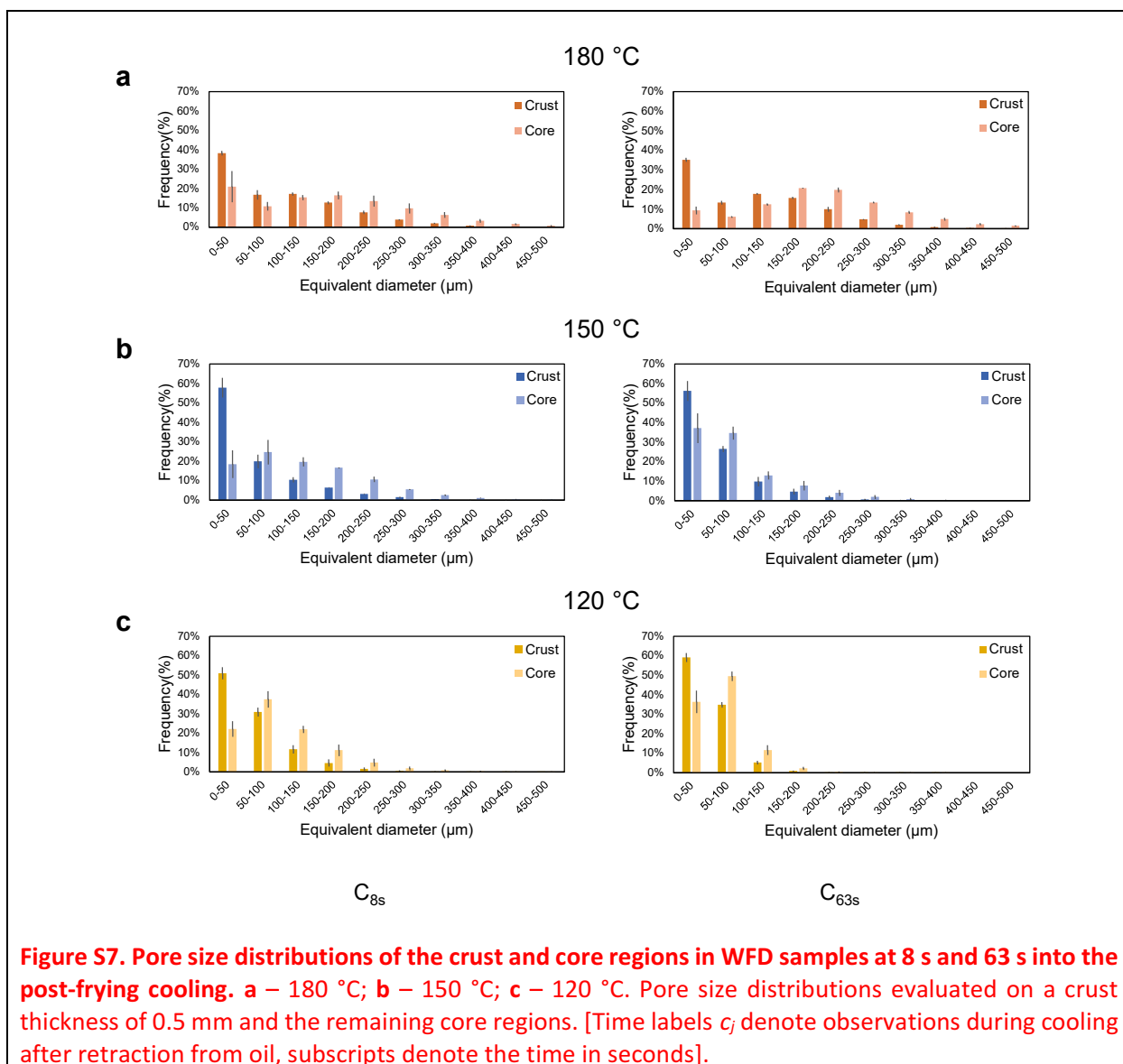


Figure S6. Evolution of regional structure thickness distribution of the solid gas cell wall matrix for WFD samples deep-fried at 180, 150 and 120 °C during post-frying cooling. a Local structure thickness distribution in a crust region of 0.5 mm thickness; **b** structure thickness distribution in the remaining core regions. [Time labels c_j denote observations during cooling after retraction from oil, subscripts denote the time in seconds].



DISCUSSION

1. Overall, the discussion should be almost completely rewritten. There is too much redundancy with the introduction and results sections, and it is hard to see a real discussion in this section.

o Lines 295–338: Redundant, very extensive, and no real discussion is presented.

A: We appreciate the reviewer's suggestion to rework the discussion section and agree that there was some overlap with other sections; however, we feel it is important to discuss the results presented in the manuscript with respect to pre-established theories related to the deep-frying process and oil uptake. Nevertheless, we recognize that the section can be restructured to convey a clear message based on our findings and their implications. The discussion has been largely modified to reflect the same, new discussion related to moisture loss and temperature measurements has also been added together with additional discussion related to the presented technique based on comments from the other reviewers.

Lines 373-439: Despite growing health consciousness among consumers, deep-fried foods remain popular in the global snack market. Their high caloric density, resulting from oil uptake during the deep-frying process, has significant implications for both consumers with preferences for low-fat products and formulation strategies within the food industry. Innovations in healthier frying techniques and the formulation of new deep-fried products with reduced fat content require fundamental knowledge of the mechanisms governing oil absorption dynamics. However, the mechanisms resulting in deep-frying-induced oil uptake have been difficult to substantiate (much less visualize) due to the highly complex and intense nature of the process².

We demonstrated that high speed time-resolved X-ray tomography can be a useful technique for dynamic in-situ visualization of the deep-frying process and presented a comprehensive study exploring the spatial (in three dimensions) and temporal changes in microstructure during and after deep-frying and its impact on oil absorption dynamics. The effect of different frying temperatures was used to study differences in the structure deformation, oil uptake, and the development of crust and core regions in a model wheat flour dough (WFD) system.

The current study addresses the limitations of previously used imaging methods in this research domain (e.g., CLSM, SEM) that lack the combined spatial and temporal resolution necessary to capture this highly dynamic process. Using a bespoke, compact experimental setup with adjustable temperature control and remote operation in a synchrotron setting, we eliminated the need for sample manipulation during and after deep-frying. Additionally, optimized spatial and temporal resolutions enabled fast acquisition at 1 tomograph/s, allowing us to resolve critical features of the evolving internal microstructure and capture previously unobservable events, such as the influence of pore connectivity on oil uptake.

This technique facilitates relatively easy adjustments to the acquisition protocol in terms of frying conditions (duration, temperature), spatial and temporal resolution, allowing for the investigation of specific phenomena related to the deep-frying process. However, as presented above, certain limitations remain: (1) the water phase was not resolved and is lumped with the solid matrix due to complex interactions between the sample constituents; (2) motion artifacts prevented phase segmentation during frying; and (3) oil uptake, that may be occurring in the small nano-pores, remains undetectable at current voxel resolutions. Balancing spatial and temporal resolution, alongside data processing demands, remains a key consideration. Furthermore, while reliance on synchrotron light sources currently limits accessibility, rapid advances in lab-based systems and iterative reconstruction

algorithms hold significant promise for broader application of this technique for similar applications in food processing^{46,47}.

Given the large amounts of time series data generated during imaging, machine learning assisted segmentation has proven to be indispensable for the processing and segmentation of the different phases for quantification of important parameters during post-frying cooling. Although undesirable uncertainty in prediction maps is unavoidable. Validation steps, including visual cross-checks between segmented datasets and corresponding grey-scale images, with a human-in-the-loop during training, were applied to minimize inaccuracies. Furthermore, μ CT image analysis offers distinct advantages over traditional methods such as nitrogen adsorption (NAM) and mercury intrusion porosimetry (MIP) for evaluating pore characteristics. These techniques require extensive sample preparation, including degassing and removal of oil and moisture, which can induce irreversible changes to the sample microstructure (e.g., shrinkage), potentially leading to discrepancies in measured values. NAM is effective for detecting nanopores but is not suited for characterizing larger pores (> 500 nm)⁶¹, while MIP, although useful for analysing larger pores ($0.2 - 250$ μ m), requires high pressures for effective mercury intrusion, that can potentially damage the samples⁶². In contrast, several studies have shown that image analysis (μ CT) yields results comparable to traditional analytical methods, with no significant differences in oil content and porosity measurements^{45,61,63}, confirming the reliability and accuracy of image processing for characterizing these porous structures.

We were able to confirm a number of pre-existing theories related to the process, (1) continually escaping gases significantly impede oil uptake during deep-frying^{4,31}; (2) higher oil temperature, and, consequently more extensive moisture loss, during deep-frying results in higher oil uptake^{2,30}; and (3) the rising trend of oil uptake during post-frying cooling^{23,64}. Combined with complementary tests for moisture content and temperature measurements, the present investigation provides additional insights into these underlying theories. Capillary action within the small crust pores was the primary driver of oil absorption during cooling, with vapour condensation further contributing to oil uptake as temperatures drop below 100 °C. The formation of a distinct crust in samples deep-fried at 180 °C and 150 °C was more conducive for oil uptake due to surface openings in a deformed microstructure. Differences in structural transformation (of starch and protein constituents) due to differing rates of moisture loss from heat conduction within the samples contributed to the distinctly different oil uptake rates and oil distribution inside the microstructure. For samples deep-fried at 120 °C, limited deformation during deep-frying resulted in a significantly lower final oil content due to reduced connectivity of the collapsed pore-space during cooling.

o Lines 352–258: “The results from this study suggest that low-temperature deep-frying operations can be employed as a pre-processing technique in a two-stage deep-frying process. In the first stage, deep-frying at lower temperatures forms a porous microstructure with limited connectivity at the product surface, thereby hindering oil uptake and limiting the formation of harmful by-products. This is followed by deep-frying at a higher temperature to tailor microstructural properties and achieve the desired textural quality in a crispy exterior with a rigid crust.” This cannot be derived at all from this work; it has nothing to do with a two-stage frying process, which is a completely different experimental setup.

A: We have removed the discussion related to a two-stage deep frying from the manuscript. We agree that there was not enough evidence to support the associated conclusion based on our experiments at single temperature deep-frying.

The paragraph has been updated as below:

Lines 452-462: Deep-frying not only induces physicochemical changes in food components but also in the frying medium (oil) as oil degradation has been linked to frying duration and temperature⁶⁶. Moreover, it is well established that the formation of acrylamide, a compound classified as a probable human carcinogen by the International Agency for Research on Cancer, accelerates at temperatures above 120 °C^{7,67}. While this has prompted the adoption of several pretreatment techniques (e.g., pulsed electric field, ultrasound, radiofrequency, etc.)⁶⁸ and modified frying processes at lower temperatures (e.g., par-frying and vacuum frying)^{21,69}, the exact influence on the product microstructure and dynamics of oil uptake remains unclear. 4D μ CT imaging can be employed to assess the effectiveness of the different pre-processing techniques in products undergoing deep-frying operations to assist with the interpretation and control of oil uptake.

o Line 374: The rate of moisture loss is not addressed in this study, so it cannot be referred to as such in the discussion (and actually should be, as I pointed out earlier).

A: As recommended by the reviewer, new data related to moisture loss during deep-frying has been added in the updated manuscript (see reply to comment 1 in RESULTS – [page 2](#)). **Line 463-465** (previously line 374) remains unaffected from the new results and has not been modified. Some additional discussion related to moisture loss and structure deformation has also been added.

MATERIALS AND METHODS

1. The main concern I have is related to the thresholds used to differentiate the different phases (Solid, Oil, and Gas). This is key, as it is the basis for the work presented. How did you verify that the protocol you defined was accurate? For instance, did you examine the different components (e.g. oil, solids) on their own to verify the protocol defined? What happened with water? You lumped it into the solids. Is this correct? Is there any chance to segment it? In this section, it is very important to develop the importance of X-ray attenuation, and elaborate on this, with respect to your components (not really density!) and how you can justify it accordingly, with much more depth than what is actually described. This must be developed as it is critical (Lines 145–152 and 486–491 in M&M).

A: Identifying the different phases is indeed critical to how we draw inferences from the observations during the 4D μ CT.

With respect to water, in **lines 586–587** (previously lines 489–491) it was stated: “Here, the solid structure represents the (deep-fried) WFD including starch-gluten-water as the interaction between the components changes during the process.”, to clarify that water is indeed lumped together with the solids. It was not possible to identify and segment water from the solid.

In relation to the differentiation of the different phases, the transmitted beam detected by the detector assembly was attenuated not just by the individual sample constituents, but also by the borosilicate glass (flask) and a significant volume of hot frying oil. Therefore, imaging the individual constituents outside the experimental setup would not have been comparable to verify the defined protocol. However, using lab based μ CT, we have previously been able to verify the order of greyscale intensities in similar deep-fried model systems (Contardo & Bouchon, 2018; Riley et al., 2023). Thus, the greyscale intensity order of the components described in **line 585** (previously line 489), combined with their spatial context, provides a reliable basis for confidently identifying these phases. Additions have been made to the following lines in the updated manuscript:

Lines 156–166: 3D volume images were obtained that render the porous microstructure contrasting the solid matrix, gas or oil filled pores and frying oil in addition to the Teflon sample container (TSC) (**Fig. 1d**). Here, the highly attenuating solid phase (representing the deep-fried WFD) appears brighter in the tomograms, thus enabling the separation from the lowly attenuating gas, whereas the oil phase lies in-between these two phases due to differences in X-ray attenuation and phase effects. **The phases were not differentiated based solely on their greyscale intensity thresholds, but together with spatial and temporal context at different stages during acquisition.** As the sample expanded during deep-frying, the space it occupied increased due to increasing gas volume (representing increased histogram fraction) thus displacing the oil volume (representing a decreasing histogram fraction).

Lines 563–572: The 3D topographies for each acquisition time point were reconstructed taking into account parameters such as centre-of-rotation alignment, single material complex refraction index ratio [$\Delta/\beta = 1000$ (frying), 6000 (post-frying)] and ring artefact removal to obtain cross-section image stacks of 3D distribution of material phases. **The transmitted beam detected by the detector assembly was attenuated not just by the individual sample constituents, but also by the borosilicate glass (flask) and a significant volume of hot frying oil.** Owing to propagation-based phase contrast contribution thanks to (partial)spatial coherence of the impinging radiation, sufficient sensitivity was obtained to distinguish between four different phases within the 3D reconstructed phase fields: gas, oil, sample, TSC.

Lines 578–595: **As individual phases affected the absorption characteristics of X-rays, phases were characterized using their respective intensity values based on their distinct attenuation-characteristics**

arising from differences in atomic number and density along with spatial and temporal context from time-series data at different stages during acquisition^{73,74}. For example, from the images and grey intensity histograms in **Figure 1d** we identified oil in contact with the sample inside the TSC at f_{15} followed by subsequent expulsion of oil in later timepoints f_{345} and used the corresponding attenuation coefficient to identify oil in the sample. The order of greyscale intensities (from bright to dark) followed TSC > Solid > Oil > Gas. Here, the solid structure corresponds to the (deep-fried) WFD including starch-gluten-water as the interaction between the components changed during the process. It was previously demonstrated using lab-based μ CT that the starch-water solid structure indeed exhibited higher X-ray attenuation than oil and air¹⁴. As most of the water was bound by the starch and gluten components in the hydrated WFD sample, only a small proportion of free water would have been available^{33,75}. During deep-frying, the phase conversion of water to vapour would further render the identification of water difficult as the moisture content continuously reduced during the process (**Figure 4a**). Therefore, it was not possible to identify and segment water from the solid. Other authors confirmed the challenge of separating water from the solid structure due to their similarity of effective atomic number (Z_{eff})⁶³.

Lines 606-612: Conversely, sample deformation was limited for the post-frying process and did not influence the segmentation. This was performed with the help of machine learning assisted pixel classification in Ilastik v1.4.0⁵⁸. Sparse manual annotations were marked **based on spatial context and greyscale intensities** across 10 % of the reconstructed images (150 cross-sectional slices) for each timestep and for each sample group from the different deep-frying temperatures (180, 150 and 120 °C) as training data for the random forest classifier to label pixels into four classes: oil, gas, solid and TSC.

The effectiveness of spatial encoding during segmentation is best demonstrated in the images acquired during the post-frying cooling, where there is no interference of the flask filled with the frying oil. Using purely greyscale intensity-based thresholding leads to misclassification of the different phases, including the extremes of TSC (highest grey value) and air (lowest grey value) (**Figure R1**). In contrast, with the help of machine learning (ML) assisted pixel classification in ilastik, the resulting probability maps considers spatial correlations in neighbouring regions (presence of oil in perforations of TSC, oil between the sample and TSC) defined during model training. This was also evident from the images in **Figure S3** in Supplementary information.

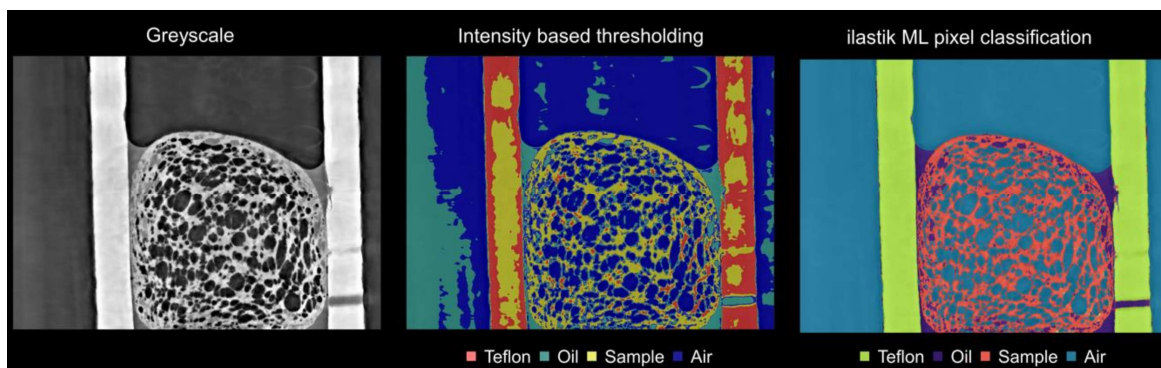


Figure R1. Comparison between segmented images using intensity-based thresholding and ML-assisted pixel classification in ilastik.

The advanced image processing pipeline we have applied thus ensures the complex spatial context is taken into account. ilastik uses a random forest classifier, a type of ensemble learning method, to classify pixels based on the provided training data. The classifier learns to associate specific features with the user-defined classes.

2. Lines 393–394: “with perforations (0.7 mm diameter) along the surface to allow easy exchange of fluids during deep-frying.”

o Explain the perforation setup (number of holes, frequency/pattern, % of open surface, etc.). This is critical.

A: The TSCs comprised of 115-120 perforations with 0.7 mm diameter in a circular pattern along the surface and ~15 perforations with 0.3 mm diameter at the bottom. This amounts to ~20% of open surface area of the tube. Based on our observations and analysis in previous studies, the perforations were adequate in permitting access to hot oil and expulsion of gases.

Based on the suggestion, we have provided the additional description in the materials section and added **Figure S8** in Supplementary information:

Lines 482-486: Cylindrical TSCs were machined from Polytetrafluoroethylene (PTFE) rods in a standard configuration of 5.5 mm internal diameter and 18 mm high, with ~20% open surface area through 115-120 perforations with a diameter of 0.7 mm along the surface and 17 perforations with a diameter of 0.3 mm at the bottom to allow easy exchange of fluids during deep-frying (see Supplementary **Figure S8a, b**).

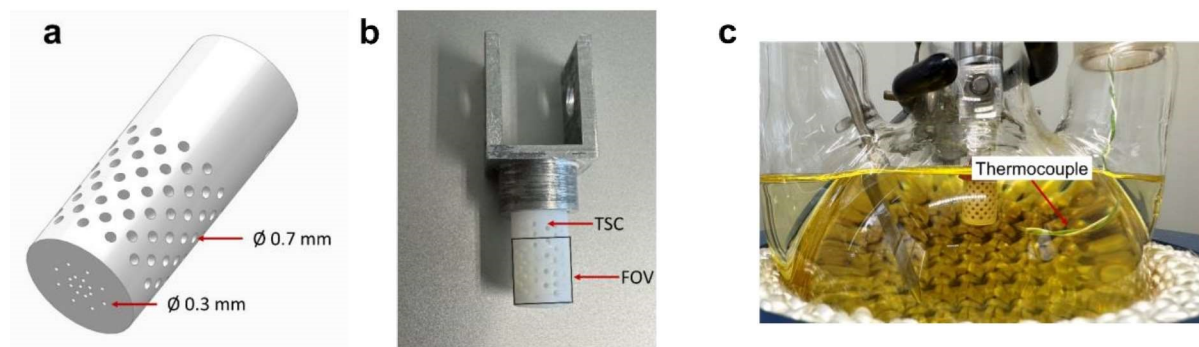


Figure S8. Sample holder and thermocouple location. **a** Perforation pattern along the TSC; **b** TSC inserted into the coupler for the linear actuator, the perforations align with the location of the sample enabling adequate exchange of fluids. FOV during imaging is highlighted with a black box; **c** Location of thermocouple to measure the temperature of oil was kept at the same depth as the sample holder.

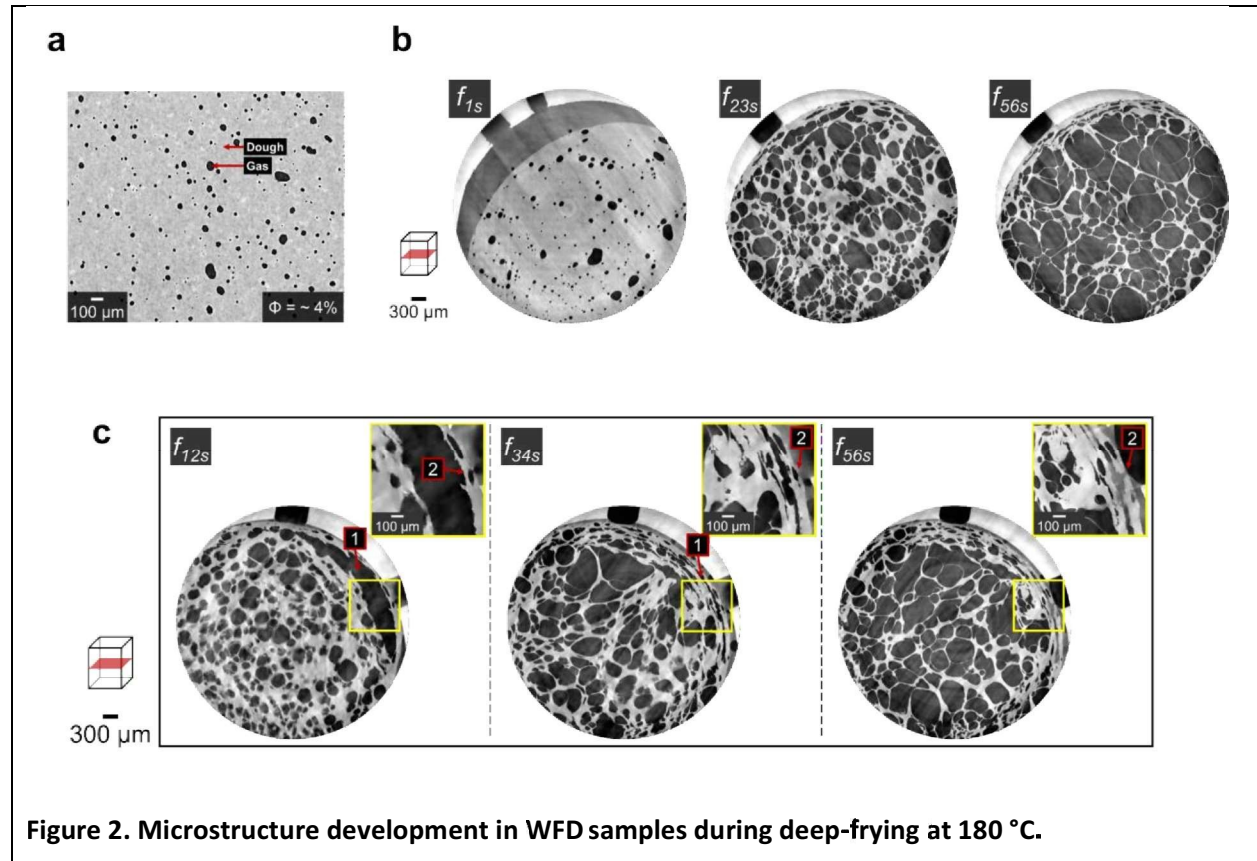
3. Lines 459–464: If I understood correctly, it took 1 second to acquire each tomograph, didn't it?

A: That is correct, each tomograph was acquired in 1 s capturing 1000 radiographs at 1 ms exposure. Each acquisition in turn, comprised six tomographs during deep-frying or post-frying cooling.

FIGURES

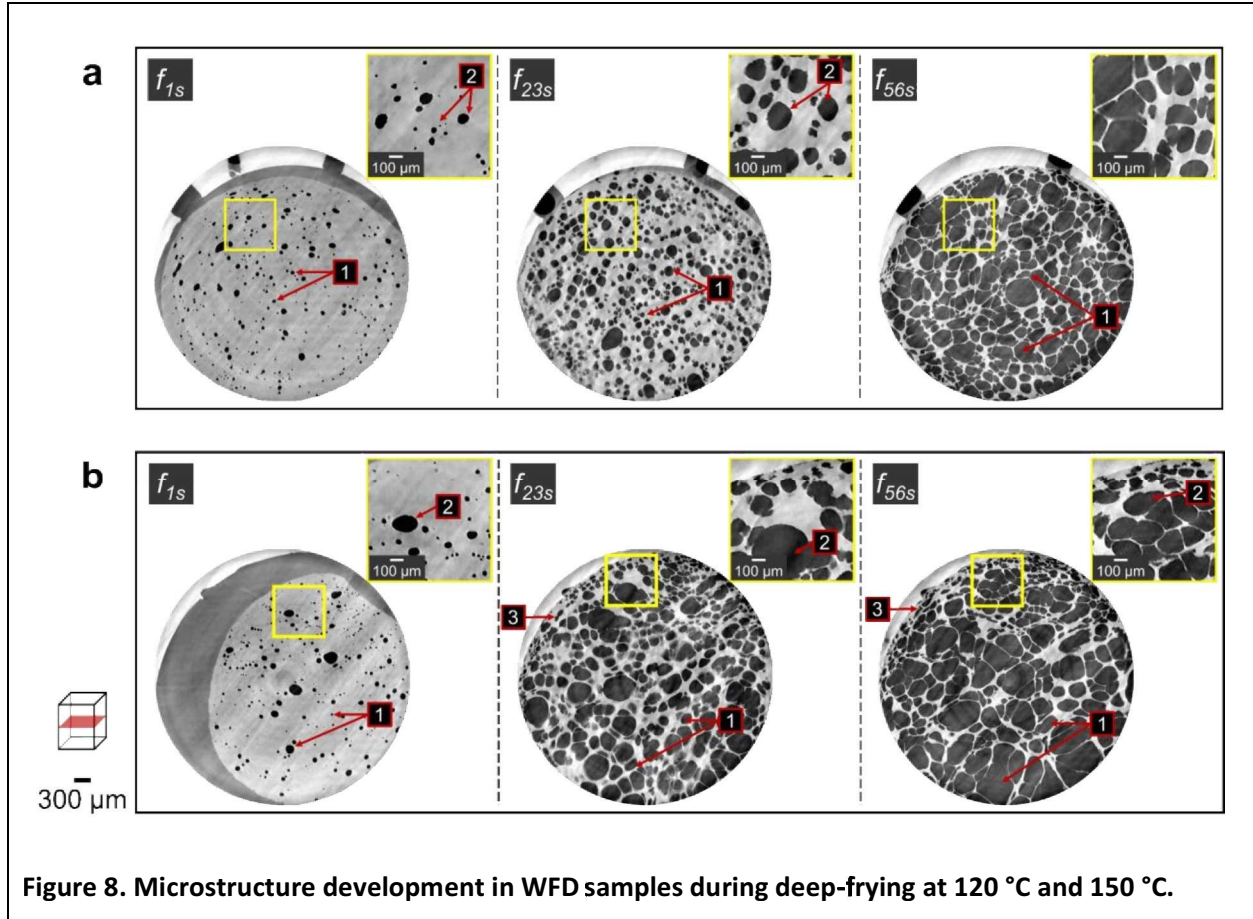
- Figure 2c: I cannot really see: “(2) localized oil ingress through ruptured walls of open pores at the surface during the late stages of deep-frying” (Lines 208–209: “Even when oil was detected in pores near the surface, it was unable to penetrate further into the sample indicating a lack of connectivity”).

A: We have updated **Figure 2**, the number label ‘2’ has been repositioned to better highlight the location of the rupturing pore wall at the surface and oil ingress in subsequent timepoints. With regards to **Lines 238–240** (previously lines 208–209), the series of images in **Figure 2c** and their insets show that the oil ingress was restricted to the pore that was ruptured during frying and did not invade any neighbouring pores. This statement is further supported by **Figure 6a (C_s at 180 °C)** in the updated manuscript where the segmented and labelled pore space confirmed this lack of connectivity.



- Figure 2c: Numbers 1 and 2 cannot be read properly in the figure. Differentiate them better.

A: The font size for the numbers has been increased, in addition a border has been applied to these numbers for better differentiation in **Figure 2c** (see previous comment) and **Figure 8** (previously **Figure 7**).



3. Figure 3: What is the scale? 500 microns?

A: Indeed, the scale bars in all figures is in micrometres. The same was mentioned in the figure captions with “Scale bars indicated in μm ...”. We identify this could be better expressed; therefore, in the updated manuscript we have added the units to the scalebars in all figures.

4. Figure 3: Images are every 4 s, not every 3 s. Figure 3b is every 11 s, not 10.

A: That is correct we thank the reviewer for pointing this out, we identify the mistake and have updated the figure caption with 4 s and 11 s respectively.

Figure 3. Vertical cross-sections of reconstructed images highlighting microstructure development in WFD samples during in-situ deep-frying at 180 °C. a Reconstructed images follow the expansion of WFD structure during the first 21 s (observations every 4 s) after immersion in hot oil; **b** Reconstructed images follow the development of WFD microstructure during 56 s of deep-frying (observations every 11 s). [Scale bar indicated in μm ; time labels f_j denote observations during deep-frying period, subscripts denote the time in seconds].

5. Figure 3: Lines 174–178: I am not sure this can be stated so definitively. Also, I cannot really judge whether it is a phenomenon that starts from the core or not, as I do not know where this tiny region of the sample is located.

A: The images presented in **Figure 3** are located in the middle of the sample, as denoted by the small cube representation  in the figure. The field of view (FOV) for image acquisition is fixed, therefore the sample expands from what is observed at f_1 . The inner wall of the TSC can be seen on the right edge of the image in **Figure 3b**. These sentences are thus supported by the observations in **Figure 3b** and in **Figure 2c**. Furthermore, as was confirmed by the temperature measurements (**Figure 4b**), temperatures inside the core climb to 100 °C before the evaporation before 23 s (see reply to comment 2 in RESULTS - [page 5](#)). This results in continuous evaporation, leading to the observed structure deformation in the core. However, we recognize this message was not easily conveyed from **Figure 3b** alone, therefore we have updated the **lines 196-200** (previously lines 174-178) as:

Lines 196-200: Expansion in the core regions **continues as temperatures level to 100 °C (Fig. 4b), simultaneously pushing the** surface layers against the **inner walls of the** sample holder (observed from 23 s onwards in **Fig. 3b and Fig. 2c**) indicated a high flexibility of the biopolymer constituents hampering vapour escape at the surface during the early stages of deep-frying^{11,52}.

6. Figure S2: Lines 250–252: “The minimal pore connectivity between the crust and the core due to a strong wall network prevented large amounts of oil from penetrating deeper into the structure, with most of the oil identified in small, compressed pores near the surface.” I do not really identify the strong wall network.

A: We agree and have replaced the phrase ‘strong wall network’ with ‘intact wall network’ in **lines 292-295** (previously lines 250-252):

Lines 292-295: The minimal pore connectivity between the crust and the core due to **an intact** wall network prevented large amounts of oil from penetrating deeper into the structure, with most of the oil identified in small, compressed pores near the surface.

7. Figure 7: In order to compare the effect of different frying temperatures, it would be suitable to compare the structure for the same moisture loss. Is this possible? Or at least report the moisture loss at each time (physically determined in a parallel experiment), so this can be discussed? According to the images, I would suggest that they could be similar, that is, 23 seconds at 150°C could be closer to 56 seconds at 120°C, and so forth.

A: This comment has been addressed in our reply to comment 3 in the RESULTS section – [page 8](#). We have added new moisture content data as **Figure 4a** in the updated manuscript and discussed the rate and extent of moisture loss between temperatures in relevant sections. **Figure 8** (previously **Figure 7**) illustrates specific differences in structure deformation and pore morphology for the samples deep fried at different temperatures. **Figure S4** provides a more comprehensive comparison of structure across different timepoints together with the new moisture content data.

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Reviewer #2 (Remarks to the Author):

The topic of the manuscript is not very novel, as the dynamics of oil uptake in deep-fried food products have been studied by many authors, also using X-ray microcomputed tomography. However, the use of time-resolved 3D X-ray computed tomography makes it interesting. The design of the experiment and description of methods are not well-prepared, and this part needs to be improved. Additionally, some changes should be made in the next part - results and discussion.

A: We greatly appreciate the constructive suggestions, which have been instrumental in enhancing the manuscript. To begin we would like to humbly emphasize the novelty and difference of our work compared with others as follows:

- This study presents a first of its kind use of time-resolved synchrotron tomography to capture immediate structural changes and oil uptake during the challenging conditions of the deep-frying process.
- Most previous studies using X-ray microcomputed tomography have relied on static imaging taken before or after deep-frying, rarely achieving this level of combined spatial and temporal resolution especially under these conditions. In contrast, our approach allows dynamic visualization of rapid structural deformation and oil uptake in a 3D heterogeneous structure throughout both frying and cooling phases at various temperatures using a singular setup.
- The ability to image the cooling phase immediately after deep-frying allowed us to observe and quantify events such as oil uptake and structure collapse, previously not possible with other imaging setups. This has enabled new insights into the structural factors (e.g., pore connectivity) that govern oil uptake in fried foods.
- Furthermore, new pretreatment techniques (pulsed electric field, ultrasound, microwave, radiofrequency, or their combination) are being adopted to modify microstructure of products to reduce oil uptake. This method offers an important tool for examining the effects of these different pretreatment techniques during the deep-frying process to determine their applicability in reducing oil uptake.

We believe that this work highlights the importance of high speed 4D imaging technique in the context of fried food processing. This also has potential in studying and enhancing the understanding of previously unobservable dynamic changes in other processes, thus opening new possibilities for product improvement.

Based on the comments from another reviewer, additions have been made to the materials section with respect to new temperature and moisture content measurements (see reply to comment 1 and 2 by Reviewer #1 in RESULTS – [pages 2-8](#)). We have reviewed the methods section thoroughly to ensure all procedures, and data analysis steps are clearly described. If there are specific areas where more detail or clarification is needed, we would be grateful for guidance to revise the same. All other comments have been addressed below.

1. Abstract: What is the definition of a "two-stage deep-frying operation"? Actually, the samples in this study were fried at temperatures of 120, 150, or 180 °C on a single occasion.

A: We have removed the mention of 'two-stage deep-frying operation' from the manuscript (both from the abstract and the discussion sections). We agree that there was not enough evidence to support the associated conclusion based on our experiments at single temperature deep-frying.

The abstract and discussion have been updated as below:

Abstract:

Oil serves as both the high-temperature heating medium during deep-frying unit operations and a contributor to the organoleptic properties of deep-fried foods. Its absorption is linked to structural deformations during deep-frying that create pathways for oil to enter the food microstructure. This study proposes a 4D imaging system (three spatial dimensions and time) using fast synchrotron radiation tomography for in-situ visualization during deep-frying and post-frying cooling to understand the mechanisms behind oil absorption. Using wheat flour dough as a model food system, we investigate the impact of frying oil temperature on structural deformation and pore formation in the crust and core, pore structure integrity, and oil uptake and distribution. The results show that higher temperatures lead to the formation of a distinguished crust with surface openings, facilitating greater oil absorption in small crust pores through capillary action. Comparing 3D microstructures attained at different frying oil temperatures, final oil content was measured as 14.4 % at 180 °C, 12.2 % at 150 °C, and 1.3 % at 120 °C, with temperature-dependent structural changes in pore connectivity and network integrity significantly impacting the rate of oil uptake and its distribution.

Lines 452-462: Deep-frying not only induces physicochemical changes in food components but also in the frying medium (oil) as oil degradation has been linked to frying duration and temperature⁶⁶. Moreover, it is well established that the formation of acrylamide, a compound classified as a probable human carcinogen by the International Agency for Research on Cancer, accelerates at temperatures above 120 °C^{7,67}. While this has prompted the adoption of several pretreatment techniques (e.g., pulsed electric field, ultrasound, radiofrequency, etc.)⁶⁸ and modified frying processes at lower temperatures (e.g., par-frying and vacuum frying)^{21,69}, the exact influence on the product microstructure and dynamics of oil uptake remains unclear. 4D μ CT imaging can be employed to assess the effectiveness of the different pre-processing techniques in products undergoing deep-frying operations to assist with the interpretation and control of oil uptake.

2. In this study, why was wheat-flour dough selected as the study object? Does it have universal applicability?

A: Wheat flour dough was selected to showcase the versatility of the technique, the samples exhibit features of distinct crust and core analogous to various fried products. Products such as doughnuts, noodles, and breaded snacks are all based on wheat-flour, and these undergo similar frying processes with distinct microstructural behaviours. Unlike potato-based products, which have a more homogenous cellular structure, wheat flour dough enables exploring a wider range of structure deformations and interactions due to its complexity and variability across different formulations. To clarify the use of selected samples in the presented work, we have made additions to the Introduction (**lines 39-43**) to highlight the versatility of wheat flour dough.

Lines 39-43: Deep-fried products based on wheat flour (ca. 70-75 % starch and 10-12 % protein) are popular across several product categories (e.g., donuts, noodles, breaded products) and culture-specific product variations within each category (e.g., the donut-style beignet in France, churros in Spain, youtiao in China) have made deep-fried foods based on wheat flour ubiquitous worldwide.

3. How does the 4D imaging system improve our understanding of the theories related to the deep-frying process?

A: 4D imaging (three spatial dimensions and time) provides a unique capability to observe the deep-frying process in real time. This approach revealed how continuous structure deformation during deep-frying alters the pore connectivity thus influencing oil uptake during cooling. By visualizing these phenomena in-situ and in high resolution, it has been possible to link these structural changes to key process parameters, such as temperature and frying time, with increased accuracy. As the internal microstructure is heterogeneous, having 3D spatial information has proven crucial in understanding the location and extent of oil ingress into the food with respect to crust and core regions. Our findings based on the 4D imaging offer new insights that refine and expand existing theories related to the deep-frying process, especially on how the deforming microstructure of the food during deep-frying governs oil uptake during cooling. This has been addressed in the discussion:

Lines 425-439: We were able to confirm a number of pre-existing theories related to the process, (1) continually escaping gases significantly impede oil uptake during deep-frying^{4,31}; (2) higher oil temperature, and, consequently more extensive moisture loss, during deep-frying results in higher oil uptake^{2,30}; and (3) the rising trend of oil uptake during post-frying cooling^{23,64}. Combined with complementary tests for moisture content and temperature measurements, the present investigation provides additional insights into these underlying theories. Capillary action within the small crust pores was the primary driver of oil absorption during cooling, with vapour condensation further contributing to oil uptake as temperatures drop below 100 °C. The formation of a distinct crust in samples deep-fried at 180 °C and 150 °C was more conducive for oil uptake due to surface openings in a deformed microstructure. Differences in structural transformation (of starch and protein constituents) due to differing rates of moisture loss from heat conduction within the samples contributed to the distinctly different oil uptake rates and oil distribution inside the microstructure. For samples deep-fried at 120 °C, limited deformation during deep-frying resulted in a significantly lower final oil content due to reduced connectivity of the collapsed pore-space during cooling.

4. How does the pore network integrity affect oil uptake in fried foods?

A: Pore network integrity dictates the pathways for oil ingress from the surface. Intact networks limit the oil ingress in the outer layers of the sample. This was also confirmed from the results presented in the 'Pore connectivity' subsection in results. And also partially highlighted in lines 238-240 wherein oil ingress during frying was limited to ruptured pores through the altered pore connectivity during deep-frying. Furthermore, the importance of an intact pore network was emphasised in Figure 6 and Figure 7 (in the main text) where, oil was observed entering far deeper into the collapsed microstructure of samples deep-fried at 150 °C compared to 180 °C.

5. As you know, frying is a rapid process, especially during the initial seconds of immersing the sample in the frying oil. Conducting 3D X-ray computed tomography testing is a lengthy process, involving the exposure of samples over 360° and the acquisition of over 1000 radiographs. The precision of the time-resolved data appears to be lacking.

A: For this study 1000 radiographs were captured over 360° for each tomograph in only 1 s. This was found sufficient to see the microstructural changes in-situ. If this resolution were insufficient, we would have very blurred image reconstructions that could not be interpreted at all. We were able to image these samples immediately after immersion into a sufficiently large oil volume and throughout frying and

cooling without interruption of the processes. The ability to obtain a tomograph in only 1 s was in fact the main motivation to carry out the experiments at a synchrotron facility.

We have also showcased the flexibility that this technique offers in selecting different time intervals for acquisition (3 s and 10 s in this study). Although localized motion artifacts were present in the images captured during deep-frying, the protocol presented here was optimized to fit the scope of this study, i.e., capturing a comprehensive account of important events during the complete duration of the deep-frying process while also managing storage constraints. This acquisition protocol can be further adjusted depending on the intended application for future studies. For example, shortening time intervals between successive tomographs would allow for a greater precision in capturing rapid early-stage processes, such as the initial bubbling stage and deformation. Similarly, with adequate storage infrastructure, the whole process can be visualized through continuous acquisition, however, this could lead to large volumes of data to process.

Similarly, the ongoing advancements in iterative reconstruction methods could help in further reducing the number of radiographs required for reconstruction, thus shortening the duration of acquisition to increase temporal resolution (Withers et al., 2021). A new discussion (**lines 394-405**) has been added with regards to the temporal resolution.

Lines 395-406: This technique facilitates relatively easy adjustments to the acquisition protocol in terms of frying conditions (duration, temperature), spatial and temporal resolution, allowing for the investigation of specific phenomena related to the deep-frying process. However, as presented above, certain limitations remain: (1) the water phase was not resolved and is lumped with the solid matrix due to complex interactions between the sample constituents; (2) motion artifacts prevented phase segmentation during frying; and (3) oil uptake in the small nano-pores remains undetectable at current voxel resolutions. Balancing spatial and temporal resolution, alongside data processing demands, remains a key consideration. Furthermore, while reliance on synchrotron light sources currently limits accessibility, rapid advances in lab-based systems and iterative reconstruction algorithms hold significant promise for broader application of this technique for similar applications in food processing^{46,47}.

6. In order to further confirm the precision and reproducibility of the analysis of image processing for reconstructed images, additional methodologies for quantitative analysis of pore structure (nitrogen adsorption methods and mercury intrusion porosimetry) should be included.

A: In the manuscript, all quantitative analyses have been presented with means and their respective standard deviations. We agree with the reviewer that the reproducibility could have been better highlighted, therefore in the updated manuscript, we have added statistical basis to all reported data.

Lines 671-676: see reply to comment 4 from Reviewer #1 in RESULTS – [page 9](#)

Lines 344-358: see reply to comment 4 from Reviewer #1 in RESULTS – [page 11](#)

Lines 360-365: see reply to comment 4 from Reviewer #1 in RESULTS – [page 9](#)

A: A number of previous studies have confirmed that image analysis (μ CT) yields results comparable to traditional analytical methods, with no significant differences in oil content and porosity measurements (Contardo et al., 2020; Contardo & Bouchon, 2018; Zhang et al., 2024), so we believe that additional confirmative methods are not necessary. Further, correlating μ CT results with additional methods such as

nitrogen adsorption (NAM) and mercury intrusion porosimetry (MIP) would be challenging because of the following reasons:

1. Isolated and closed pores cannot be characterized (Farber et al., 2003) while μ CT does allow this.
2. Extensive sample preparation is required for techniques like nitrogen adsorption and mercury intrusion porosimetry, including de-gassing, moisture removal (via hot air drying or freeze-drying), and oil extraction (using solvents). These processes can cause irreversible changes to the sample and pore structure (e.g., shrinkage), potentially leading to discrepancies between measured and actual values (Zhang et al., 2020).
3. NAM while inherently non-destructive requires removal of moisture and oil from the structure before analysis, even so, only pores sizes in the nanometre range are characterized (Ho & Hutmacher, 2006). This would lead to under reporting of larger pores in these samples. It has also been reported in a previous study that pores in the range of 0.03-10 μ m limit oil absorption which would reduce the applicability of this technique for current application (Zhang et al., 2020).
4. Porosity measurements using MIP are highly dependent on the applied pressure, with characterized pores typically ranging between 0.2 – 250 μ m. Higher pressures required for adequate mercury intrusion [to prevent ink-bottle effect (Xue et al., 2020)] would lead to a possibility of damaging the sample structure due to compression (Ho & Hutmacher, 2006).

We acknowledge that quantitative analysis evaluated from the μ CT analysis is dependent on the voxel resolution (Cnudde et al., 2009). However, for the samples evaluated, the micrometre scale was sufficient to describe the observed porosity changes and oil uptake behaviour.

We have now addressed these issues in the updated manuscript:

Lines 413-424: Furthermore, μ CT image analysis offers distinct advantages over traditional methods such as nitrogen adsorption (NAM) and mercury intrusion porosimetry (MIP) for evaluating pore characteristics. These techniques require extensive sample preparation, including degassing and removal of oil and moisture, which can induce irreversible changes to the sample microstructure (e.g., shrinkage), potentially leading to discrepancies in measured values. NAM is effective for detecting nanopores but is not suited for characterizing larger pores (> 500 nm)⁶¹, while MIP, although useful for analysing larger pores (0.2 – 250 μ m), requires high pressures for effective mercury intrusion, that can potentially damage the samples⁶². In contrast, several studies have shown that image analysis (μ CT) yields results comparable to traditional analytical methods, with no significant differences in oil content and porosity measurements^{45,61,63}, confirming the reliability and accuracy of image processing for characterizing these porous structures.

Lines 397-403: However, as presented above, certain limitations remain: (1) the water phase was not resolved and is lumped with the solid matrix due to complex interactions between the sample constituents; (2) motion artifacts prevented phase segmentation during frying; and (3) oil uptake in the small nano-pores remains undetectable at current voxel resolutions. Balancing spatial and temporal resolution, alongside data processing demands, remains a key consideration.

Lines 632-634: Oil uptake in the small nano-pores present in the crust region was not investigated in order to achieve a good balance for the acquisition protocol across acquisition rate, voxel resolution and FOV considerations.

7. Can this 4D computed tomography be applied to other types of cooking methods?

A: Real-time, dynamic imaging capabilities of 4D μ CT make it applicable for studying other complex, fast-evolving cooking processes where structural changes are important. This process has already been used at a lower temporal resolution for bread baking [to image fermentation (Turbin-Orger et al., 2015) and baking (Schott et al., 2023)], freeze drying (Gruber et al., 2024) and microstructural evolution in ice cream (Guo et al., 2017) among others. The work presented in this manuscript broadens the methodological toolkit available for investigating processes where real-time high resolution imaging of dynamic processes is valuable.

We have made additions to the Introduction to highlight the versatility of 4D computed tomography for other cooking applications:

Lines 109-115: The brilliance of synchrotron light sources enable μ CT acquisitions at spatial and temporal resolutions that are magnitudes higher than those currently achievable in lab based μ CT settings⁴⁷. 4D μ CT imaging has been used at lower temporal resolutions to gain valuable insights into processes such as bread dough fermentation⁴⁸, bread baking⁴⁹, and freeze-drying kinetics⁵⁰. Given the added flexibility in terms of experimental setups, these are attractive for investigating and uncovering theories related to the deep-frying process.

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Reviewer #3 (Remarks to the Author):

1. Core Content: The paper "High-speed 4D computed tomography for uncovering the dynamics of oil uptake in deep-fried food products" explores the use of a 4D imaging system, specifically fast synchrotron radiation tomography, to visualize the highly dynamic processes occurring during the deep-frying and post-frying cooling of wheat flour dough (WFD) based food products. The study provides in-situ, time-resolved 3D imaging to understand the changes in the porous microstructure and oil uptake dynamics. Key findings suggest that the pore network integrity significantly impacts oil absorption rates and distribution, and that a two-stage deep-frying process at different temperatures can reduce oil content while achieving the desired crust.

2. Limitations:

a. Spatial and Temporal Resolution: The study utilizes high-speed synchrotron radiation tomography, but the resolution may still be insufficient to capture the smallest microstructural changes, particularly during the rapid phase transitions in the early stages of deep-frying.

b. Sample Size and Representativeness: The research is based on WFD samples, which may not fully represent the diversity of deep-fried food products. The generalizability of findings to other types of food matrices is limited.

c. Experimental Conditions: The controlled laboratory conditions, including the use of specific temperatures and sample holders, may not perfectly replicate real-world deep-frying scenarios. The influence of variations in oil type, frying duration, and food composition on the results was not extensively studied.

d. Machine Learning Segmentation: The segmentation process, while advanced, introduces potential inaccuracies due to classification uncertainty, particularly in regions of phase transition and boundary determination.

3. Significance of Limitations:

a. Spatial and Temporal Resolution: Importance: High - Accurate resolution is critical for understanding the detailed microstructural changes and their impact on oil uptake. Insufficient resolution can lead to incomplete or misleading interpretations of the dynamics involved. Explanation: Detailed imaging is essential to capture the nuances of pore formation, expansion, and oil ingress. Any loss in resolution could obscure critical transitional phases, affecting the study's conclusions about oil absorption mechanisms.

b. Sample Size and Representativeness: Importance: Medium - While WFD provides a useful model, deep-fried foods vary widely in composition and structure. Explanation: Broader applicability requires testing on various food types. Limited sample diversity restricts the ability to generalize findings and apply them to different culinary practices or industrial applications.

c. Experimental Conditions: Importance: Medium - Laboratory conditions provide control but may not fully mimic commercial frying environments. Explanation: Real-world variability in frying conditions, such as oil degradation, varying moisture content, and different frying techniques, could influence the microstructural changes and oil uptake dynamics, potentially altering the study's practical relevance.

d. Machine Learning Segmentation: Importance: Medium - Segmentation accuracy is vital for quantitative analysis. Explanation: Errors in segmentation can lead to incorrect quantification of oil and gas phases, affecting the reliability of the data on oil uptake and microstructural changes. This limitation impacts the precision of the study's findings and their subsequent applications.

Conclusion: The study significantly advances the understanding of oil uptake dynamics in deep-fried foods through innovative 4D imaging techniques. However, limitations in resolution, sample diversity, experimental conditions, and segmentation accuracy must be addressed in future research to enhance the applicability and precision of these findings across a broader range of food products and real-world conditions.

A: We sincerely appreciate your valuable suggestions and feedback on the relevance of this work. These have been instrumental in enhancing the manuscript. The limitations mentioned by the reviewer have been addressed below:

- Spatial and Temporal Resolution: This study presents a first of its kind use of time-resolved synchrotron tomography to capture immediate structural changes and oil uptake during the challenging conditions of the deep-frying process. Previous studies have been limited either by their spatial, temporal resolution, or both, and did not account for any changes occurring in the time between the sample removal and the imaging. We acknowledge that the spatial resolution may not capture the smallest microstructural changes during deep-frying. However, it effectively resolves key features relevant to frying dynamics within the scope of the study. The strength of current study lies in eliminating the need of sample manipulation during and after frying, crucially capturing events not previously observed (e.g., the effect of pore connectivity on oil uptake).
- Sample Size and Representativeness: WFD samples were chosen as a model system, the sample provides a sufficient applicability in terms of formation of distinct crust and core regions during deep-frying similar to many other products (e.g., donuts, noodles, breaded products). Additional food matrices can be investigated in future studies using this technique.
- Experimental Conditions: Laboratory conditions, including specific temperatures and controlled setups, were designed to ensure repeatability. While these conditions may not fully replicate industrial frying, they allow for fundamental insights. Variations in oil type and food composition are noted as areas for future exploration.
- Machine Learning Segmentation: We acknowledge potential uncertainties in segmentation, particularly in complex regions. This was addressed in the manuscript in the methods section. These were observed mostly in the solid phase undergoing extensive deformation and not between oil and gas phases. Given the amounts of data generated and nature of the dataset, ML assisted segmentation has proven indispensable in segmenting the different phases (during post-frying cooling). The uncertainty, while undesirable, is unavoidable. Validation steps, including visual cross-check between segmented datasets and corresponding grey-scale images during training, were performed to minimize inaccuracies. Limitations of using ML-segmentation were addressed in the methods section and **Table S2** in Supplementary information.

We believe that this work highlights the importance of high speed 4D imaging technique in the context of food processing. This has a potential to study and enhance the understanding previously unobservable dynamic changes in other processes, thus open new possibilities for product improvement.

Based on these comments, additions have been made to the discussion section in the manuscript.

Lines 395-406: see reply to comment 5 from Reviewer #2 – [page 27](#)

All other comments have been addressed below.

Details:

1、 Abstract needs to be rewritten again. Lack data results

A: The abstract has been updated based on the reviewer's suggestion to reflect some quantitative results. However, we are somewhat restricted by the character limits based on the journal guidelines.

Oil serves as both the high-temperature heating medium during deep-frying unit operations and a contributor to the organoleptic properties of deep-fried foods. Its absorption is linked to structural deformations during deep-frying that create pathways for oil to enter the food microstructure. This study proposes a 4D imaging system (three spatial dimensions and time) using fast synchrotron radiation tomography for in-situ visualization during deep-frying and post-frying cooling to understand the mechanisms behind oil absorption. Using wheat flour dough as a model food system, we investigate the impact of frying oil temperature on structural deformation and pore formation in the crust and core, pore structure integrity, and oil uptake and distribution. The results show that higher temperatures lead to the formation of a distinguished crust with surface openings, facilitating greater oil absorption in small crust pores through capillary action. Comparing 3D microstructures attained at different frying oil temperatures, final oil content was measured as 14.4 % at 180 °C, 12.2 % at 150 °C, and 1.3 % at 120 °C, with temperature-dependent structural changes in pore connectivity and network integrity significantly impacting the rate of oil uptake and its distribution.

2、 Keywords failed to reflect the main aspects of the MS

A: We have updated the keywords based on this comment. The updated list is as follows:

X-ray micro-CT, 4D imaging, food microstructure, machine learning assisted segmentation, deep-frying, porous media, oil uptake, heat and mass transfer.

3、 Which kind of deep frying products? Why choose wheat dough not French fries?

A: Wheat flour dough was selected as a model food system to showcase the versatility of the 4D imaging technique, the samples exhibit distinct features of crust and core, that are analogous to various other deep-fried products. Products such as doughnuts, noodles, and breaded snacks are based on wheat-flour, and these undergo similar deep-frying processes with distinct microstructural behaviours. Unlike potato-based products, which have a more homogenous cellular structure, wheat flour dough enables exploring a wider range of structure deformations and interactions due to its complexity and variability across different formulations.

French fries are beyond the research focus of the research groups that were involved in this study. However, the investigation of potato-based products (e.g., French fries) is indeed an interesting use case for this technique. At this point, however, this would stretch beyond the scope of this work and is

therefore left for future studies. To clarify the use of selected samples in the presented work, we have made additions in the **lines 39-43** to highlight the versatility of wheat flour dough.

Lines 39-43: Deep-fried products based on wheat flour (ca. 70-75 % starch and 10-12 % proteins) are popular across several product categories (e.g., donuts, noodles, breaded products) and culture-specific product variations within each category (e.g., the donut-style beignet in France, churros in Spain, youtiao in China) have made deep-fried foods based on wheat flour ubiquitous worldwide.

4、 What are the frying oils? Why choose sunflower frying oil not palm oil or others?

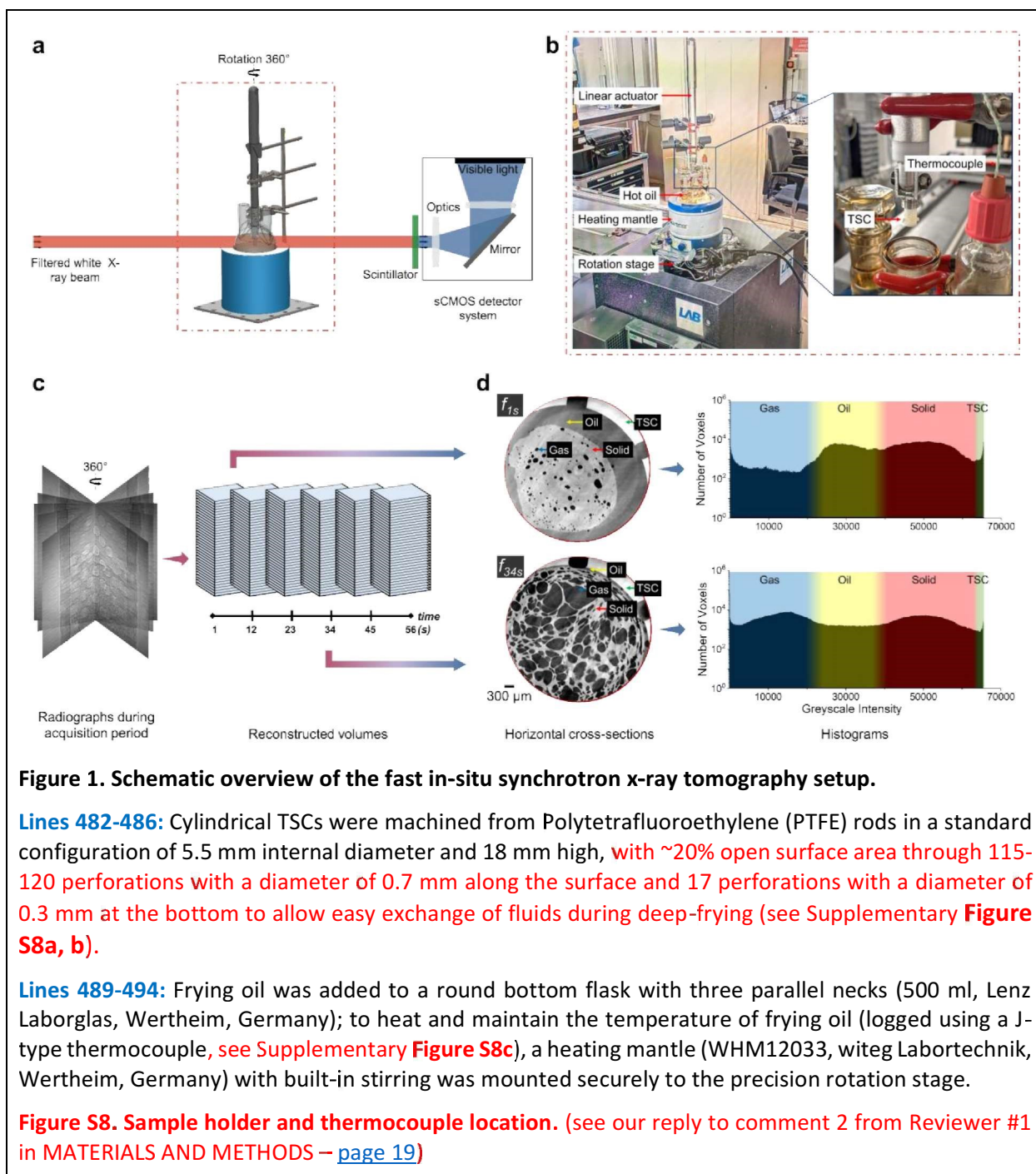
A: The focus of this work was to extend the fundamental understanding of microstructural changes during deep-frying and their effect on oil uptake. Sunflower oil was chosen as a frying medium due to its high applicability in industrial deep-frying applications and relevance to the study objectives. The presented results are broadly applicable to other deep-frying applications with different frying oils. There is a possibility that other frying oils (e.g., palm oil) with different physical properties (e.g., viscosity) may influence the final oil content, however, there is also growing interest in alternative oils to palm oil due to the significant environmental effects from its production. Future work could examine how different frying oils influence microstructural changes and oil absorption, as mentioned in the updated discussion:

Lines 480-482: Sunflower frying oil was obtained from Agristo (Nazareth, Belgium), and was chosen as a frying medium due to its popularity in industrial deep-frying applications.

Lines 466-472: To build further on this work, a multifaceted approach is advised to gain a better understanding of the relationship between deep-frying temperature and sensory and nutritional properties of the product. Furthermore, future investigations to gain insights into how other factors (e.g., food formulations, oil types, physicochemical properties of carbohydrate or protein constituents etc.) contribute to pore network connectivity are recommended as high-resolution, time-resolved imaging methods become more accessible.

5、 The sample holder is not clear in Figure 1, please provide a clearer picture and mark the location where the sample is placed. In addition, the position of the thermocouple should also be marked.

A: We thank the reviewer for pointing this out. The image of the sample holder presented in **Figure 1** has been enlarged for better clarity. In addition, we have provided additional description about the sample holder in the materials and **Figure S8** in Supplementary information. The FOV marked in **Figure S8b** highlights the scanned region during acquisition and thus the location of the sample. **Figure S8c** shows the location of the thermocouple measuring the temperature of the oil, a reference for the image has been added to the manuscript in **line 491**.



6、 How are gas, oil and solid precisely defined in Figure 1D? Is there a basis for this definition?

A: This has been addressed in our reply to comment 1 by Reviewer #1 in MATERIALS AND METHODS – page 17.

7、 What do the different colors represent in Figure S2.

A: We have updated the figure caption for better explanation:

Figure S2. 3D volume render of labelled pore space inside WFD samples deep-fried at 180 °C after 56 s of frying. A small region of interest was segmented, and the pore space was separated from the solid structure (grey) to investigate the connectivity between the crust and the core regions. The same label colour of adjacent objects depicts a connected pore network. Pores present in the outer crust regions were distinctly disconnected to the core regions of the sample.

8、 In lines 252-253, no pore filling was observed in the core area? You can see pore in the central regions in Fig. S3.

A: Original sentence has been combined with previous sentence for more clarity.

Lines 288-290: These findings also confirmed that oil uptake was mainly limited to the outer crust region of the sample (see Supplementary Fig. S3).

9、 In lines 271-272, where is the crust microstructure of WFD deep-fried at 180°C ?

A: We thank the reviewer for pointing out this mistake. We have updated these lines 317-319 (previously lines 271-272) with the correct references to figures in the updated manuscript as below:

Lines 317-319: A distinct, thick crust was observed in Figure 8b with small, flattened pores analogous to the crust microstructure of WFD deep-fried at 180 °C (Fig. 2b).

10、 In lines 276-279, the paper stated that the sample shrink to less than half of their initial gas porosity during the cooling period, which does not suitable for all fried samples, such as the samples fried in 180°C. What is structure thickness? In Table 1, the oil content and porosity of fried WFD are obtained from image analysis, which is somewhat subjective and can be added objective test indicators, such as the use of Soxhlet extraction to measure oil content and mercury meter to measure the porosity.

A: **Lines 333-337** (previously lines 276-279) correspond to the subsection: “Microstructure and oil uptake influenced by oil temperature”, discussing the WFD samples deep-fried at 120 °C and 150 °C shrinking during the cooling period. The same was observed visually in the 3D volume renderings of the sample microstructures during cooling (Figure 6 in the updated manuscript) and Table 1 (column 5, Gas porosity). This behaviour of structure collapse was not observed in the samples deep-fried at 180 °C and was not mentioned as such.

Structure thickness refers to the local thickness of the gas cell walls (expressed in μm). This is calculated using a sphere-fitting algorithm as described in the methods section. We have further expanded upon this metric by evaluating the differences in structure thickness of the crust and core regions of the deep-fried WFD samples in the updated manuscript (see Supplementary Figure S6 and Table S1).

With regards to the evaluation of oil content and porosity of fried WFD using image analysis, the comment has been partially addressed (for porosity measurements using mercury intrusion porosimetry) in our reply to comment 6 from Reviewer #2 – [page 27](#).

The determination of oil content using μ CT has been found to yield comparable results to those obtained using Soxhlet extractions in a number of previous studies (Contardo et al., 2020; Contardo & Bouchon, 2018; Riley et al., 2023). This method has an added benefit in the form of spatial context of oil presence and pore connectivity that highlight the heterogeneity of the deep-fried samples, that cannot be achieved using these analytical methods (Ho & Hutmacher, 2006; Zhang et al., 2024). For these reasons, we have not performed additional Soxhlet extractions.

We have made the following additions to the manuscript.

Lines 256-259: The morphological properties of the crust (considered as a 0.5 mm thickness subsurface layer) and core (remaining internal regions) were evaluated by using the local structure thickness of the solid gas cell walls (presented in Supplementary Fig. S6 and Table S1) and pore size distributions (Fig. S7a).

Lines 413-424: See our reply to comment 6 by Reviewer #2 – [page 28](#).

Lines 643-652: To investigate the microstructural differences between the crust and core regions of the deep-fried samples, an outer layer with a constant thickness of 0.5 mm was separated at the surface using the chamfer distance map in Avizo. The crust and core regions were masked from the segmented dataset and pore size distribution was evaluated at 8 s and 63 s during the post-frying cooling following a watershed separation. The local structure thickness distribution (expressed in μm) of the starch-gluten matrix was evaluated for the sample and the separated crust and core regions using CTAn (Version 1.18.8.0, Bruker SkyScan, Kontich, Belgium). The sphere-fitting algorithm in the 3D analysis module provided the local thickness of the structure as represented by the diameter of the largest enclosed sphere⁷⁸.

11、 In lines 287-290 and 338-340, there is a contradiction in the explanations of these two parts. The former states that structural deformation hinders oil absorption, while the latter states that deformation of pore space contributes to oil uptake. The structure should contain the pore.

A: We thank the reviewer for pointing out these confusing statements. We wanted to highlight that oil uptake was influenced by structure collapse of samples deep-fried at 120 °C and 150 °C during cooling due to their respective differences in crust formation during deep-frying.

We have rephrased the **lines 359-371** (previously lines 287-293) and **lines 433-439** (previously lines 338-340) to convey the intended meaning:

Lines 359-371: Similar to the samples deep-fried at 180 °C, most of the oil absorption in samples deep-fried at lower temperatures occurred during cooling. Changing capillary pressures in crust pores and vapour condensation from temperatures decreasing below 100 °C inside the sample during cooling are both expected to have contributed to oil uptake. The samples deep-fried at 120 °C experienced significantly lower ($p < 0.05$) oil uptake across all samples due to the limited interconnectivity of pores at the surface due to lack of a distinct crust formation during deep-frying (Fig. 7c). Extensive structure collapse led to shrinking of pores inside the sample and collectively hindered oil ingress during cooling (Table 1 and Fig. S7). In contrast, samples deep-fried at 150 °C displayed oil penetration in pores further below the crust during cooling (Fig. 7b). As the structure collapsed, pore connectivity between the crust and core was disrupted. The presence of a rigid crust led to the formation of surface-connected pores

that extended deeper into the sample, allowing greater amounts of oil to enter below the crust (**Fig. 6b**).

Lines 433-439: The formation of a distinct crust in samples deep-fried at 180 °C and 150 °C was more conducive for oil uptake due to **surface openings in** a deformed microstructure. Differences in structural transformation (of starch and protein constituents) due to differing rates of moisture loss **from heat conduction within the samples** contributed to the distinctly different **oil uptake rates** and oil distribution inside the microstructure. **For samples deep-fried at 120 °C, limited deformation during deep-frying resulted in a significantly lower final oil content due to reduced connectivity of the collapsed pore-space during cooling.**

References:

- Contardo, I., & Bouchon, P. (2018). Enhancing Micro-CT methods to quantify oil content and porosity in starch-gluten matrices. *Journal of Food Engineering*, 237(June), 154–161. <https://doi.org/10.1016/j.jfoodeng.2018.05.038>
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- Zhang, J., Li, J., & Fan, L. (2024). Comparative analysis of oil absorption and microstructure of fried potato chips treated with different pretreatment via X-ray micro-computed tomography and mercury intrusion method. *Journal of Food Composition and Analysis*, 129(February), 106129. <https://doi.org/10.1016/j.jfca.2024.106129>

Response to reviewer's comments (NCOMMS-24-35988A)

Title: High-speed 4D computed tomography for uncovering the dynamics of oil uptake in deep-fried food products

Authors: U. Verma, I. M. Riley, B. Lukić, L. Broche, P. Verboven*, J. A. Delcour, B. M. Nicolai

We sincerely thank the reviewers for taking their time to evaluate the revised manuscript. We have addressed all the comments and revised the manuscript accordingly. Below, we provide our responses to the referees' comments in blue, with major changes in the main text and supplementary information highlighted in red. The significant additional work included recalculating the moisture content with new interpretation regarding profiles and mechanisms, additional analysis of the temperature profiles in light of the different mechanisms, and a more elaborate discussion of the limitations.

REVIEWER COMMENTS

Reviewer #1 (Remarks to the Author):

Dear Authors,

I sincerely congratulate you on the excellent revision you have provided. The additional experiments, descriptions, and discussions have significantly improved the manuscript. However, I have a few additional minor (but important) aspects that need to be addressed:

A: Thank you very much for your kind words and constructive feedback, we are grateful for the positive evaluation of our revised manuscript. We have responded to the individual comments and suggestions below.

1) To carry out appropriate comparisons, the moisture content must be calculated on an oil-free and water-free basis, that is, $\text{mH}_2\text{O}/\text{m defatted-dried-solids}$, that is, the mass of solids exclude oil and moisture, to ensure a constant basis. Accordingly, the associated figure and discussion must be modified.

AND

2) When presenting results on moisture loss, describe the possible mechanisms of water loss (would it be diffusion? Convection? Capillary flow? etc.), and discuss them, according to the literature. Would there be any differences in the mechanisms and driving forces involved when working below or above the boiling point of water?

A: We thank the reviewer for this remark and suggestion. As both these comments concern the same sections of text in the manuscript, we have replied to these comments together.

With respect to the comment # 1, in this revised version, we have re-calculated the moisture content on the suggested dry defatted matter basis to ensure appropriate comparisons. The associated figure and relevant sections in the manuscript have been updated.

With respect to the comment # 2, the sections where moisture content is presented has been modified to reflect possible mechanisms of water loss. Based on heat and mass transfer fundamentals and literature

evidence, there are certain differences in moisture loss mechanisms when the temperatures are above the boiling point of water; these continually evolve during the deep-frying process.

Additions have been made to the following lines in the updated manuscript:

Lines 705-711: The moisture content of the WFD and deep-fried samples at 180 °C, 150 °C or 120 °C for 5, 10, 15, 20, 30, 40, 50 and 60 s was evaluated using the AACC method 44-15.02⁷⁹. Samples were deep-fried using the same experimental setup and cooled for 60 s before moisture content determination. The measurements were repeated for a total of six replicates for each of the time points. **The moisture content was calculated on a dry matter basis [$m_{\text{H}_2\text{O}} (m_{\text{dough DM}})^{-1}$], where mass (m) is expressed in grams and DM indicates dry matter of the solids (excluding oil absorbed as a consequence of deep-frying).**

Lines 184-195: The structure expanded during the early stages of deep-frying (i.e., between 1 s and 13 s) and was transformed into a highly porous solid matrix (**Fig. 3a**). Parts of the starch-gluten network were observed to rupture under continued expansion, thus forming large pores as smaller adjacent gas pores coalesced. **At this stage, the moisture content was approximately 0.544 g/g DM ($MC_{180^\circ\text{C},15\text{s}}$, **Fig. 4a**) indicating that the sample had lost a small portion of water. Moisture was primarily removed through vaporization at the surface, where it was convected away by the oil²³. Internally, as vapour pressure increased due to evaporation at pore-matrix interfaces throughout the material, moisture gradually diffused from regions of higher saturation towards the surface^{11,55}. This thus substantiates that persistent volumetric expansion induced by water evaporation and consequent enlargement of steam-filled pores inside the WFD solid structure increasingly exerted more forces on the internal walls resulting in thinning of the solid matrix.**

Lines 204-223: Temperatures steadily increased above 100 °C in the crust, while stabilizing around the boiling point of water in the core (**Fig. 4b**). **Prolonged moisture loss ($MC_{180^\circ\text{C},30\text{s}}$ approximately 0.356 g/g DM, **Fig. 4a**) prompted the formation of a distinct region representing a crust-like structure after 34 s of deep-frying (**Fig. 2c**). Moisture migration from the core to the crust continued through a combination of pressure-driven convection and capillary diffusion for both vapour and liquid water^{11,57}, while moisture was continually removed from the crust through diffusion and convection of vapor. Crust formation was facilitated by the slower moisture migration from the core compared to the rapid vaporization at the surface²³. The crust was largely composed of numerous small, flattened pores with thick walls, while the core region consisted of larger irregular pores with thin walls. Expansion inside the highly porous structure successively slowed down between 45 s and 56 s of deep-frying, which was attributed (in part) to the substantial loss of moisture (final $MC_{180^\circ\text{C},60\text{s}}$ approximately 0.167 g/g DM, **Fig. 4a**) and the increased structural rigidity of the biopolymers in the WFD system³³. The majority of internal heat flow was directed towards converting liquid water to vapour by boiling observed by the continued core temperature plateau at 100°C. Studies on the microstructure of potato-based products have attributed the formation of small pores in the crust to sample shrinkage induced by moisture loss after prolonged deep-frying²⁵. However, dynamic imaging of the deep-fried WFD system here shows that these small pores are formed due to the expansion of the core regions and the subsequent compression of pores below the surface.**

Line 327-347: Comparing the microstructures of deep-fried (56 s) WFD samples at 180 °C and 120 °C, those deep-fried at 120 °C presented a more uniform pore morphology without a distinct crust (see Supplementary **Fig. S4a**). As the internal temperatures of samples deep-fried at 120 °C did not reach the boiling point of water, the structure did not show signs of extensive deformation including thin wall rupture (**Fig. S4a**). **The higher residual moisture content after deep-frying (final $MC_{120^\circ\text{C},60\text{s}}$ approximately 0.599 g/g DM, **Fig. 4a**) suggests that most internal regions still existed in a rubbery state, preventing any apparent rupture of the thin-walled structures in the core. Moisture loss occurred at**

the surface through low equilibrium vapor pressures and low convective heat transfer coefficients⁶². The smaller temperature difference between the surface and the core regions indicates negligible water vapour gradients, suggesting that internal moisture migration was slow and driven by diffusion²³. On the other hand, the microstructure in WFD samples deep-fried for 56 s at 150 °C revealed a structure with larger, irregular pores formed due to the rupturing thin walls of the solid matrix and more advanced gas cell coalescence (see Supplementary **Fig. S4b**). A distinct, thick crust was observed in **Figure 8b** with small, flattened pores analogous to the crust microstructure of WFD deep-fried at 180 °C (**Fig. 2b**). Heat flow was faster than at 120 °C but slower than at 180 °C. The slower increase in surface temperatures during deep-frying at 150 °C reduced the overall moisture loss from the crust compared to deep-frying at 180 °C. This resulted in higher residual liquid water content throughout the process (final $MC_{150^{\circ}\text{C},60\text{s}}$ approximately 0.405 g/g DM **Fig. 4a**), as evidenced by the temperature plateau around the boiling point of water in both the crust and core (**Fig. 4c**).

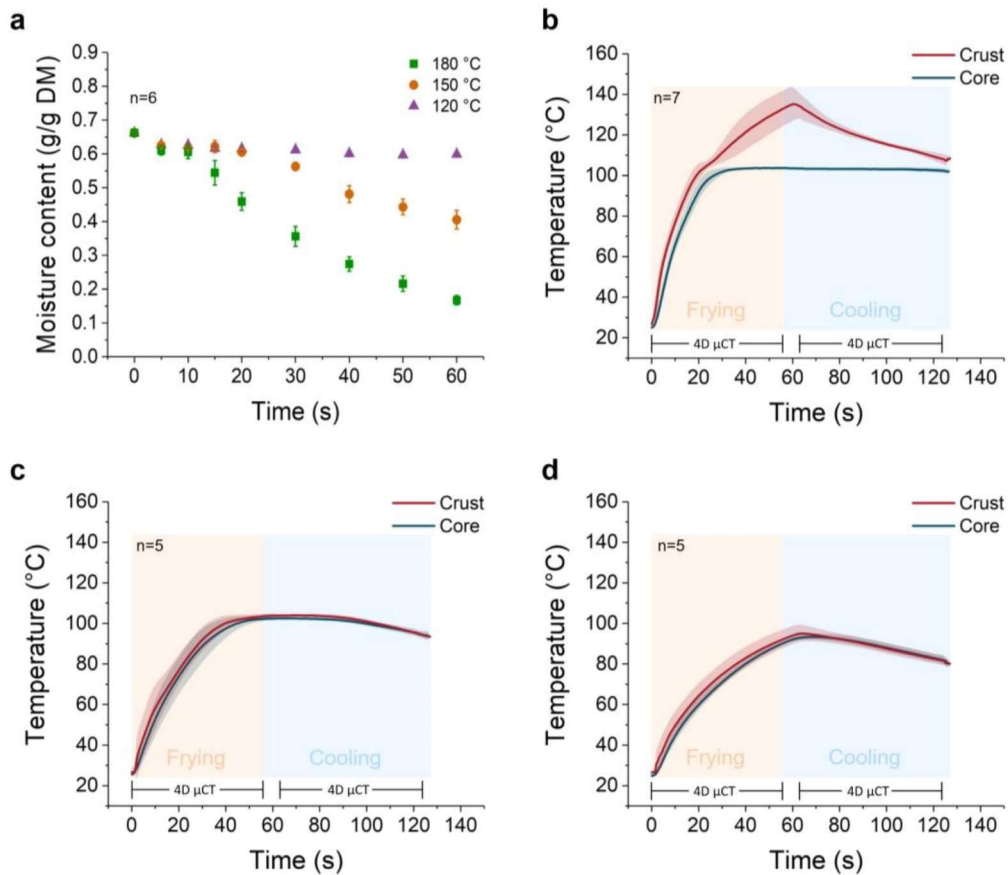


Figure 4. Moisture content and temperature measurements of WFD samples. (a) Moisture content of non-fried (0 s) and deep-fried (5 – 60 s) WFD samples with initial hydration levels of 40 % deep-fried at different temperatures (180 °C, 150 °C and 120 °C). Means and standard deviations (error bars) of six replicates are displayed; $MC, \%$ is calculated on an oil free dry matter basis $[m_{\text{H}_2\text{O}} (m_{\text{dough DM}})^{-1}]$, where mass (m) is expressed in grams and DM indicates dry matter of the solids; Local temperature measurements inside the WFD samples during deep-frying and post-frying cooling at different temperatures (b) 180 °C, (c) 150 °C, and (d) 120 °C. Each plot presents mean temperature profiles in the crust and core regions inside the samples (shadows plot the standard deviation across five to seven

replicates). The temperature measurements shown here corresponded to the deep-frying phase (orange shaded background) and post-frying cooling phase (blue shaded background), aligned with periods of 4D μ CT observations.

Lines 342-356: No significant oil ingress was observed during deep-frying in samples deep-fried at low temperatures (see Supplementary Fig. S4). Oil temperature significantly affected the moisture loss kinetics in WFD samples due to differences in the rate of heat conduction (Fig. 4). Gradual internal structure expansion was observed at temperatures approaching the boiling point of water, as a result of slow vapour generation. Similar microstructural features were seen among samples deep-fried at 180 °C after 12 s (Fig. 3b), 150 °C after 23 s (Fig. S4b) and 120 °C after 34 s (Fig. S4a) with moisture content measuring at approximately 0.606 g/g DM in all samples (Fig. 4a). At temperatures around 100 °C, more violent structure deformations such as formation of large pores and thin wall rupture were induced by continuous evaporation and increasing vapour pressure inside the structure. This was apparent from the reconstructed images for samples deep-fried at 180 °C after 23 s (Fig. 3b) and at 150 °C after 45 s (Fig. S4b), which corresponded to similar moisture content values of 0.459 g/g DM ($MC_{180^{\circ}\text{C},20\text{s}}$) for 180 °C and 0.443 g/g DM ($MC_{150^{\circ}\text{C},50\text{s}}$) for 150 °C (Fig. 4a). The slower rate of moisture loss in samples deep-fried at 150 °C, compared to those at 180 °C, can be attributed to the lower convective heat transfer coefficients and lower equilibrium vapour pressures⁶².

Line 357-365: After removal from the hot oil (i.e., 8 s of cooling), the WFD samples deep-fried at 120 °C and 150 °C preserved their porous microstructures (Fig. 6), thus resembling the structures observed at the final time point of deep-frying (Fig. 8). However, these samples progressively collapsed during the cooling period, shrinking to less than half of their initial gas porosity (Table 1) as the internal core collapsed (see Supplementary Fig. S5 and Table S1). This phenomenon was likely related to the sustained rubbery state of biopolymers due to higher residual moisture (final $MC_{120^{\circ}\text{C},60\text{s}}$ approximately 0.599 g/g DM, $MC_{150^{\circ}\text{C},60\text{s}}$ approximately 0.405 g/g DM Fig. 4a) resulting from steam accumulation inside the core regions of the structures during deep-frying. Core regions lacking sufficient rigidity were unable to maintain the expanded shape as steam condensation induces negative vapour pressures due to reducing temperatures during cooling^{2,11,63}.

3) You specify that “In the case of WFD samples deep-fried at 120 °C and 150 °C, the difference between the temperature profiles in the crust and core were less pronounced.” Why might this occur?

A: Thank you for this comment. An explanation addressing the observed temperature profiles in the crust and core regions of the samples deep-fried at different temperatures has now been integrated in the text. The revised text now discusses how the slower heat transfer at this temperature results in a more uniform temperature distribution, contrasting with the samples deep-fried at 180 °C. We hope this addition clarifies the likely mechanism behind the temperature profiles.

Additions have been made to the following lines in the updated manuscript, in addition to the above changes:

Lines 316-326: Temperatures within the samples did not exceed 95 °C (Fig. 4d) and 100 °C (Fig. 4c) when deep-fried at 120 °C and 150 °C, respectively. The more uniform temperature distribution observed in these samples, compared to those deep-fried at 180 °C, resulted from coupled heat and mass transfer mechanisms in the deforming structure. Slower heating occurred due to lower convective heat transfer coefficients at the surface, leading to lower vapour pressures, reduced surface water loss, and incomplete liquid water evaporation^{11,62}. Consequently, the temperatures did not rise above the boiling point during deep-frying, allowing for a more uniform temperature distribution within the

sample. In contrast, deep-frying at 180°C resulted in significant temperature gradients between the crust and the core, driven by higher heat flux, increased vapour pressures, and rapid vaporization at the surface²³.

Reviewer #3 (Remarks to the Author):

The objectives and the rationale of the study have been clearly stated.

A: We sincerely thank the reviewer for their constructive comments which have greatly improved our manuscript after revision.

The limitations of current work should be elaborated upon in the text.

A: We appreciate the reviewer's suggestion to elaborate on the limitations of the current work. While we had answered the concerns in our previous response, the reviewer was likely not able to trace it back fully in the text itself. Therefore, the discussion has now been more clearly restructured to highlight the limitations as below with a separate dedicated paragraph:

Lines 429-463: Despite the clear advantages offered by the presented technique, certain limitations remain. First, the water phase was not resolved in the images and was lumped together with the solid matrix due to complex interactions between the sample constituents, which made it challenging to validate established moisture loss mechanisms^{11,55}. However, ongoing improvements in state-of-the-art instrumentation at neutron sources are generating interest in the combined use of fast neutron and X-ray imaging⁶⁴. The higher neutron sensitivity of hydrogen atoms holds great potential for visualizing moisture transport in deforming media, such as during deep-frying⁶⁵.

Second, motion artifacts hindered phase segmentation of images captured during deep-frying. Balancing the spatial and temporal resolution, alongside data processing demands remains a key challenge. However, innovations in rotation free X-ray multi-projection imaging are showing promise in capturing ultra-fast phenomenon⁶⁶.

Third, oil uptake that may be occurring in the small nano-pores remains undetectable at current voxel resolutions. This could be improved by using a more tailored setup and sample environment to enable higher sensitivity, contrast and spatial resolution, particularly with pink beam imaging⁶⁷. While reliance on synchrotron light sources currently limits accessibility, rapid advances in lab-based systems and iterative reconstruction algorithms hold significant promise for broader application of this technique for similar applications in food processing^{46,47}.

Finally, the WFD model system represents a similar class of products (*e.g.*, donuts, noodles). Applying this technique to more traditional potato-based products (*e.g.*, French fries, potato chips) and breaded foods under varying experimental conditions will provide valuable insights transferable to other mass-consumed products. Food microstructure analysis using various imaging techniques has already contributed to significant advancements across different product types^{68,69}. As in-situ imaging methods continue to evolve, they are likely to enable further breakthroughs. While the current limitations pose challenges, they also present opportunities for future improvements and innovations.

The large amounts of time series data generated during imaging make machine learning assisted segmentation indispensable for the processing and segmentation of the different phases for quantification of important parameters during post-frying cooling. Uncertainty in prediction maps during classification, especially at phase transition boundaries, is unavoidable. Validation steps, including visual cross-checks between segmented datasets and corresponding grey-scale images, with a human-in-the-loop during training, were applied to minimize errors. Advances in deep learning algorithms for segmentation of large imaging datasets are expected to further improve segmentation accuracy⁷⁰.