

# Surface analysis of retrieved bilateral UHMWPE tibial inserts under varus malalignment condition

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## ABSTRACT

Failure analysis on a retrieved ultra-high molecular weight (UHMWPE) knee tibial inserts of bilateral total knee replacement (TKR) was performed due to aseptic loosening detected after 16 years (left) and 12 years (right) in vivo services. Despite long implantation time, the effect of varus malalignment present on a 71 years old female patient (body mass index, 35.1) with a non-active lifestyle will be considered as a factor towards the TKR failure. We, therefore, determined whether implant malalignment was associated with increased surface damages in both retrieved tibial inserts. Surface damage morphology was assessed using a 3D laser microscope and Scanning Electron Microscope (SEM). ATR-Fourier Transform Infra-Red (ATR-FTIR), Differential Scanning Calorimetry (DSC) and Gel-Permeation Chromatography (GPC) were used to measure changes of chemical and physical properties of retrieved inserts. Results show left-16 years insert possesses more severe wear degradation (crater and cracks) compare to wear on right-12 years insert (delamination, multidirectional scratches, and ripple). The surface roughness on the medial compartment seems to be higher than the lateral side for both inserts which can be affected by uneven load distribution contribute by the varus deformity. Higher crystallinity of left-16 years insert (66.99%) compare to right-12 years insert (56.52%) were an indicator of major mechanical changes happen on left insert which was contributed by oxidation with respect to implantation time of both inserts. Our findings revealed that in vivo oxidation is a main contributing factor to the failure of implants, but not varus malalignment. The material properties in the oxidized layer are significantly altered, including a very substantial reduction in molecular weight displayed by both inserts.

## 1. Introduction

The survival rate of total knee replacement (TKR) has been reported to increase along with the development of advance technologies and operative techniques of current TKR [1,2]. However, failure rate of 5–7% of primary TKR remain to present on 10 year follow up studies in spite of 95.3–97.7% survival rate [3,4]. The major frequent cause of these implant failure, besides prosthesis joint infection, are found due to aseptic loosening especially on tibial part [5,6]. Bahraminasab et al. [7] reported that one of the factors that leads to the occurrence of aseptic loosening on TKR component are caused by wear debris that were generated during the tribological activities of bearing component in TKR causing reaction to surrounding tissue which consequently lead to osteolysis and

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aseptic loosening of prosthesis component. This can be generally concluded that wear degradation occur during in vivo application contribute to this aseptic loosening failure.

Wear degradation of the implant can be influenced from the condition of implant and patient itself such as type of implant, mechanical and chemical properties of implant, weight of patient, age of patient and gender of patient. Previously, oxidation and delamination fatigue were found as one of the major reasons of wear degradation in TKR [8]. Collier et al. [9] reported that this type of oxidation degradation occur over implantation time. Besides, due to this reason, Laska et al. [10] has also summarized that the lifespan of UHMWPE joint is often limited up to 15 years. She reported in her study that oxidative degradation has leads to chain scission and recrystallization, and thus, causing brittle behaviour to present on the UHMWPE component. The changes on this material properties will then resulted on mechanical degradation and thus limiting the performance of material.

However, various factors including contact stress and loading distribution are said to contribute to the extension of the previous degradation [11,12]. Maquet et al. [11] demonstrated that the resultant force on the knee must pass through the centre of gravity of total load-bearing surface of the knee. In static normal knee, the vertical force from the ground passes through the centre of knee and distribute equal load between lateral and medial compartment with respect to no horizontal force exist. Therefore, any changes in component and knee alignment can influence the loading distribution on femoral tibial interfaces associate with shear forces and which affect the polyethylene wear.

Varus deformation of knee component is one of the knee malalignment which also known as inward knee angulation that causes the load passes medially to the centre of knee and resulting greater load on medial compartment. The load distribution effect is said to increase as addition horizontal component applied during dynamic knee condition. Previous study by Kaissi et al. [13] concluded that varus deformity caused subsequent impaired distribution of force over the load-bearing of medial compartment and consequently effect the degenerative lesion of medial compartment. Another study by Srivastava et al. [14] discover the effect of varus component malalignment on wear failure in total knee arthroplasty. They reported that varus malalignment lead to higher linear and volume wear penetration, especially on medial part, which worsening local destruction of bearing surface and eventually cause osteolysis to occur.

From the above brief review, we found no further analysis was performed on wear damage mode possesses on the surface of the insert especially in this varus knee malalignment condition toward its contribution on failure in regards with implantation time. The study on surface damage mode is necessary to further explain the occurrence mechanism of wear degradation during in vivo services of the implant which said to correlated with loosening failure. Therefore, the present study attempt to examine the contribution of varus deformity on the surface damage mode of UHMWPE inserts from bilateral knee prosthesis which was retrieved from non-active elderly housewife with overweight symptom.

## 2. Material and methods

### 2.1. Retrieved sample

Bilateral knee prosthesis were retrieved for revision from a 71 years old female, housewife patient with obesity symptom (body mass index, 35.1). Previously, the patient was undergone primary bilateral knee replacement surgery in 2002 and 2006 for left and right knee respectively as of bilateral knee osteoarthritis treatment. However, after years of in vivo services, the patient started to feel severe pain on the left knee and followed by right knee 6 month afterwards. She also presented with complaint of limited daily activities such as unable to walk, climb up and walk down stairs and aggravated by movement of the knees especially when getting up from sitting position. Series of radiograph performed indicated that varus deformity were detected on both knee (Figs. 1 and 2) and severe loosening with changes on femoral and tibial tray position occur in vivo services (Fig. 3a and b). Both implants experienced

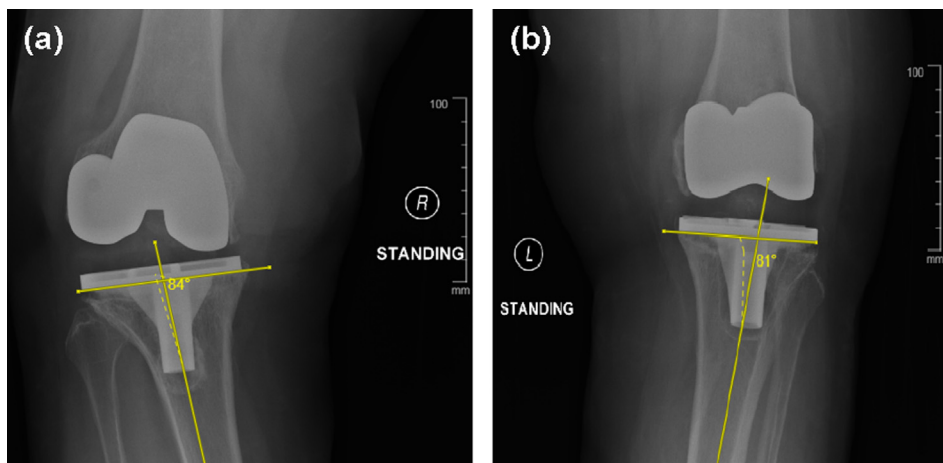
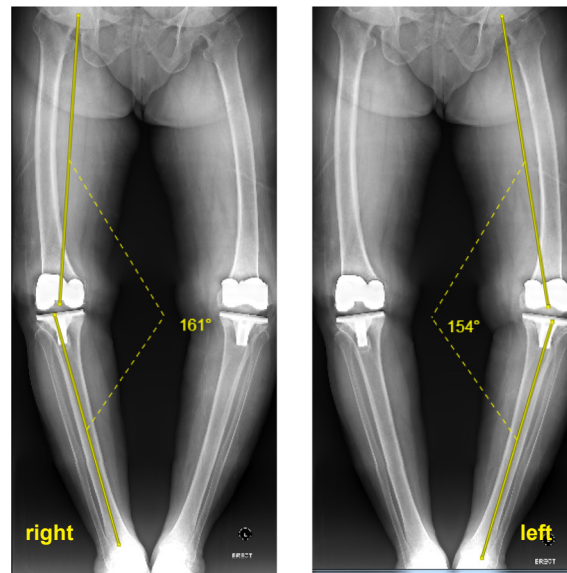


Fig. 1. Anatomic alignment for TKR in 2014 (4 years before revision) (a) right knee at 6° varus (b) left knee at 9° varus.



**Fig. 2.** Long-leg preoperative standing radiograph demonstrating a significant varus deformity with malalignment in 2018.

progression of varus deformities due to lysis. The failed knee replacement were then removed for revision and replaced with new implant from Johnson & Johnson (J&J) S-ROM hinge (Fig. 3c and d). Fig. 4a and b shows retrieval knee prosthesis for both left and right knee respectively consisting of three components which are femoral component, UHMWPE tibial insert and tibial tray.

#### 2.1.1. Right knee prosthesis

Primary right knee replacement was performed in 2006 by using fixed bearing knee prosthesis from Johnson & Johnson (J&J) Attune® Knee system. The sample was retrieved after 12 years in vivo services due to the complaining of pain at medial part of right knee. Radiograph performs shows that right knee possesses varus malalignment with inward angular of  $161^\circ$  (Fig. 2a). Retrieved knee prosthesis consists of femoral and tibial tray component made from cobalt-chromium (CoCr) alloys and tibial insert from UHMWPE material.

#### 2.1.2. Left knee prosthesis

Primary left knee replacement was performed in 2002 by using fixed bearing knee prosthesis from Zimmer Biomet Total Knee system. The sample was retrieved after 16 years in vivo services due to the complaining of pain at anterior aspect of left knee. Radiograph performs shows that left knee possesses varus malalignment with inward angular of  $154^\circ$  (Fig. 2b). Retrieved knee prosthesis from left knee consists of femoral and tibial tray component made from cobalt-chromium (CoCr) alloys and tibial insert from UHMWPE material.

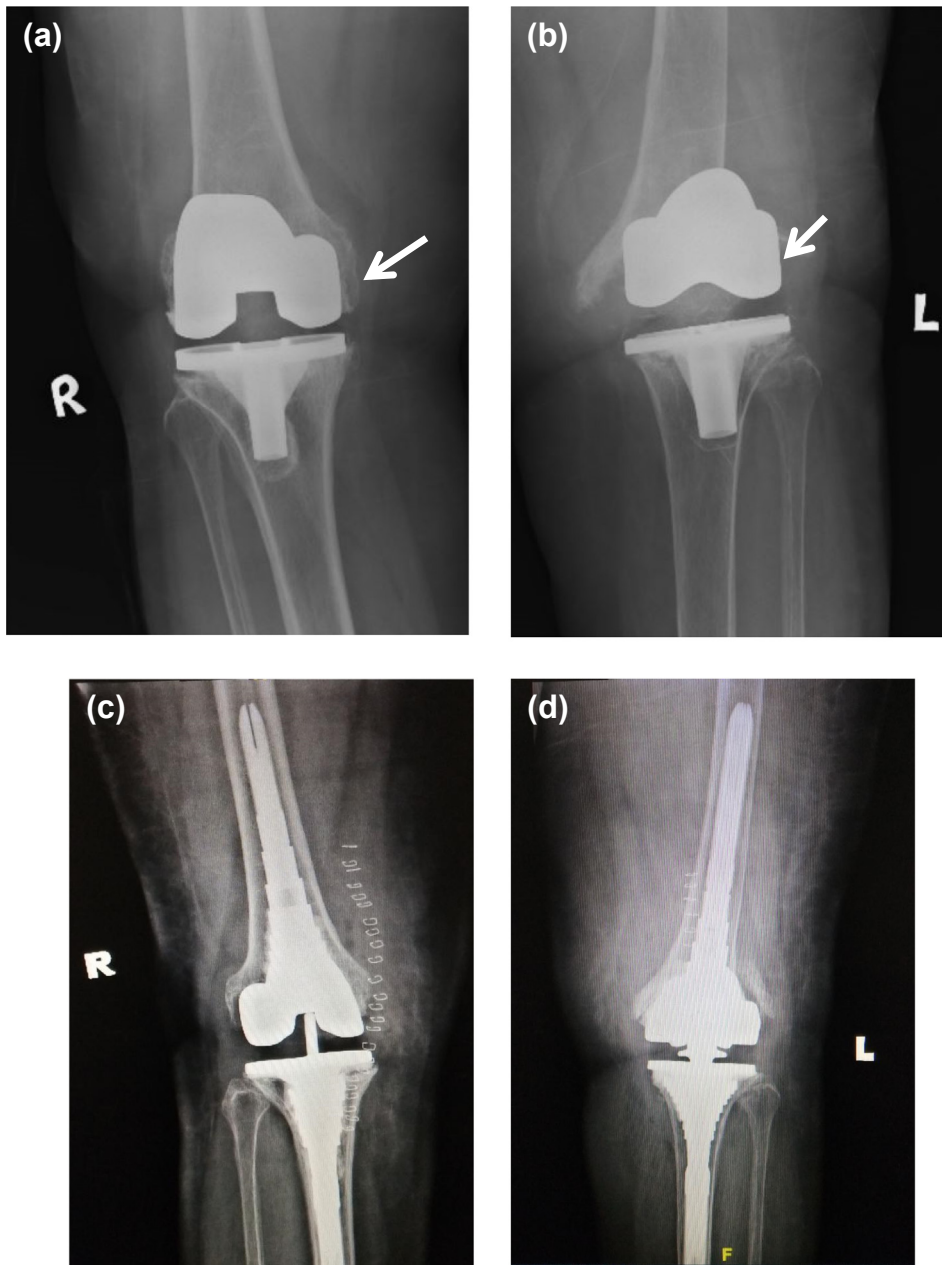
### 2.2. Surface evaluation (zoning)

The failure analysis of retrieved UHMWPE inserts was conducted by characterizing the morphological conditions of the sample surface. Due to physical contact of femoral metal to polymer tibial insert of retrieved bearing component in knee joint application for extended years, some changes in surface morphology were observed to occur. Changes in terms of surface roughness and surface degradation of the UHMWPE sample which mainly related to wear activities in vivo services will be evaluated in this study.

Surface assessment by scoring related to surface roughness will be assigned on each zone. The surface of the UHMWPE inserts were partitioned into three main zones (top, middle and bottom) on lateral-medial compartment to differentiate each location of the zone (Fig. 5a and b). This zone division resembles the pattern used by Currier et al. [15] which applies three zones in medial-lateral side and together with single zone in between the lateral and medial compartment.

Meanwhile, in association with surface roughness of the UHMWPE tibial insert, 3D laser microscopy (Olympus LEXT OLS5000) will be used further to measure the roughness of each zone by evaluating the depth of the scratch and the height of asperities on each surface. Area of  $643 \times 643 \mu\text{m}$  with  $430 \mu\text{m}$  in depth were detected by the microscopy. Repeated measurements were taken at different points for each zone and the average of the measurement was then compared between left-right UHMWPE inserts and with the other zone in medial-lateral side. Additional to that, high resolution 3D image can also be obtained regarding to respective surface roughness measurement.

Scanning electron microscope (JOEL JSM-6010 Plus/LV) will next be used to evaluate wear damage characterization of retrieved sample. Wear degradation is expected to produce higher surface roughness due to presence of wear damage mode such as abrasion, burnishing, cracking, creep, delamination, pitting and scratching that were produced by contact stress during wear activities.



**Fig. 3.** Initial anteroposterior radiographs of (a) right knee and (b) left knee (note that arrow pointing at hollow bone density due to osteolysis with femoral and tibial tray in changes position) and posteroanterior radiographs of (c) right knee (d) left knee.

Therefore, wear characterization of UHMWPE inserts were only performed on higher surface roughness of each medial–lateral compartment for both left and right knee.

### 2.3. Oxidation characterization

Oxidation of UHMWPE was reported to cause reduction in mechanical properties such as loss of toughness and decrease in wear resistance which consequently leads to degradation of the sample [16,17]. Attenuated Total Reflection-Fourier Transform Infrared Spectroscopy (ATR-FTIR)(IRTracer-100 Fourier Transform Infrared Spectrometer) was used to quantify oxidative degradation in UHMWPE sample by analysing the intensity of carbon bonding with oxygen molecules. The analysis was carried out in two distinct regions (oxidised surface area and bulk surface area) of both left and right knee tibial polymer with flexible thickness range as long as the retrieved sample has secure contact with the ATR crystal in order to obtain precise measurement. The scan spectra are detected in transmittance intervals from  $500\text{ cm}^{-1}$  to  $4000\text{ cm}^{-1}$ .



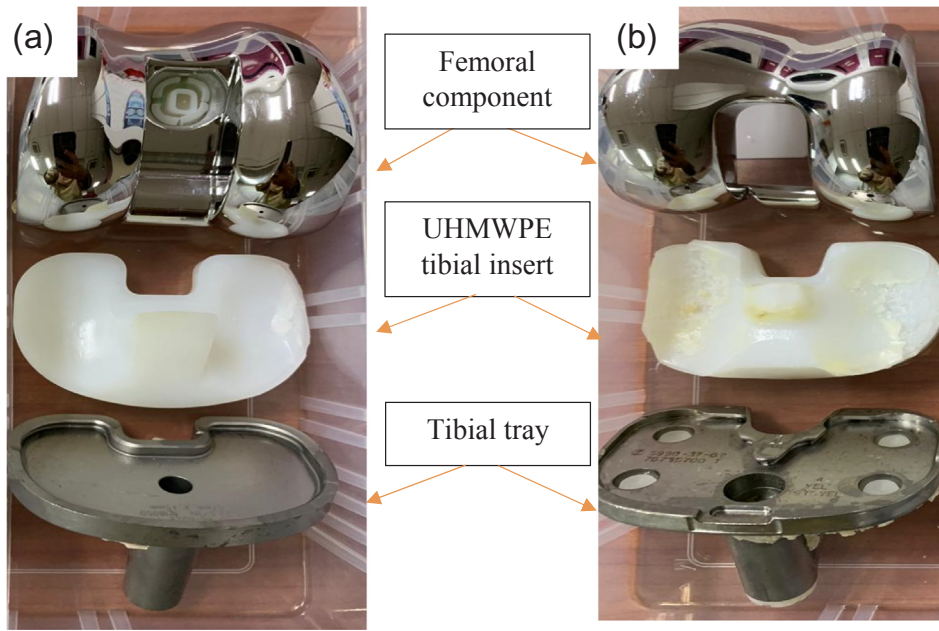


Fig. 4. Knee prosthesis component of (a) right-12 years; (b) left-16 years; knee after retrieval (cleaned).

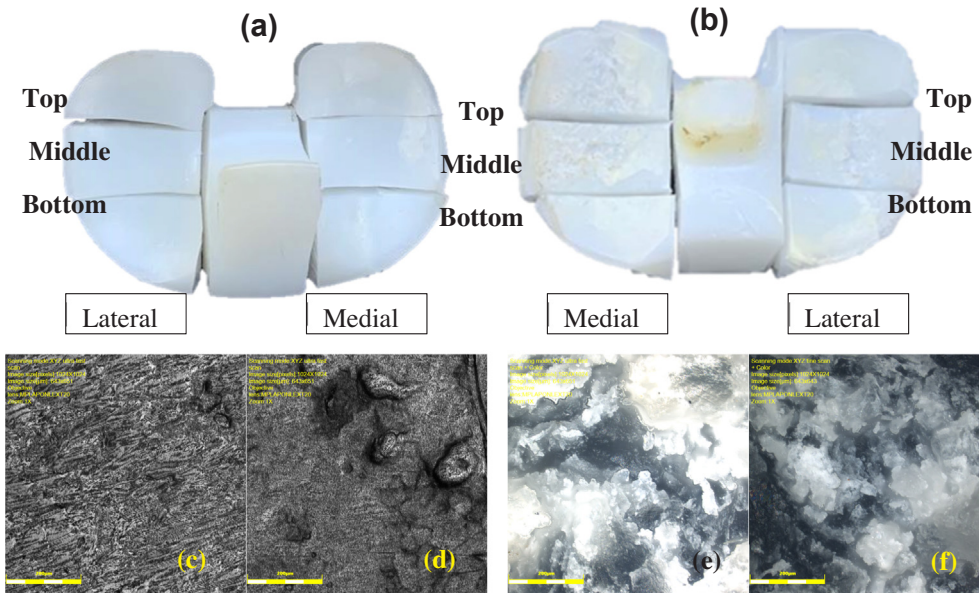


Fig. 5. General view UHMWPE tibial insert of (a) right-12 years; (b) left-16 years; and microscopic images of lateral (c and f) and (d and e) medial compartment for respective inserts.

#### 2.4. Crystallinity measurement

Measurement of degree of crystallinity of retrieved UHMWPE sample can provides fundamental which other physical properties of implant can also be predicted [18]. Differential Scanning Calorimeter (DSC) allows to determine degree of crystallinity by quantifying heat acquired to melt the polymer. The crystallinity percentage measurement can be obtain by comparing the total heat of melting  $\Delta H_m$  (the area under the endotherm) to the total heat of fusion  $\Delta H_f^\circ$  of that 100% crystalline polymer sample which in case of UHMWPE is 293 J/g [19]. The DSC experiment were performed using (Differential Scanning Calorimeter Q20) by purging nitrogen gas as at flow rate of 10 mL/min with heating and cooling rate of 10 °C/min on particulate sample with sample mass around 1–2 mg. The sample was heated from 30 °C to 250 °C and held isothermally for 5 min and then, cooled to 5 °C and held for another 5 min. Then, the sample undergoes heating process again at the same heat rate to obtain the total heat of melting for UHMWPE sample.

## 2.5. Molecular weight measurement

Gel Permeation Chromatography (GPC) (Gentech Scientific Waters 2414 refractive index (RI) detector) analysis was conducted on a small amount of UHMWPE (less than 5 mg) by dissolving the UHMWPE sample in Tetrahydrofuran at 180 °C in order to obtain the molecular weight of the UHMWPE tibial insert.

## 3. Results and discussion

There are several limitations of this study that require consideration. First, the study was lack of information on patient's medical history. The second limitation of this study was that information of implant prior the replacement surgery such as original dimensions and treatment were unavailable. Despite these limitations, this retrieval study help to better understand the relationship, if any, between the wear damage and uneven load distribution contribute by varus deformity, which encompass longer in vivo duration than had been available to earlier retrieval studies. A particular focus of this observational study is to define surface damage of tibial inserts, which failed due to aseptic loosening with varus malalignment of the mechanical axis, 6° and 9°, after 12 and 16 years implantations, respectively. In order to reveal the mechanism of surface damage, we performed detailed observation of the surface topographical and morphological properties of tibial insert by using 3D laser microscope and SEM. Due to long implantation (> 12 years after initial surgery), surface degradation due oxidation in vivo must be considered. Therefore, we also measured the chemical and physical properties of tibial inserts using FTIR, DSC and GPC.

Retrieved UHMWPE tibial inserts from bilateral knee prosthesis were accepted in a condition where there was significant different between surface appearance of right and left inserts. General visual inspection on the surface physical of both inserts indicate severity of surface damage are more prominent on left tibial insert compare to right (Fig. 5c–e). The distinct between these two surfaces condition could be resulted from difference years in vivo services of both inserts which left insert (16 years) possesses longer implantation time compare to right insert (12 years). Similarly, Kurtz et al. [17] proposed in his work that degradation of polyethylene occurs over a period prior to implantation (shelf ageing) and in vivo services of respective insert. The degradation of polyethylene are said to occur during in vivo services due to occurrence of extended cyclic loading and multiaxial sliding with reducing of mechanical properties of inserts during physical contact of femoral metal to polyethylene tibial insert of retrieved bearing component in knee joint application [20]. However, conformity and contact stress on the wear of tibial insert, with respective loading have become a concern since the retrieved inserts are from a patient with varus deformities background. Varus malalignment is said to alter the distribution of tibial loading at the femoral tibial interfaces which consequently could affected the polyethylene wear and thus contribute to implant failure [14].

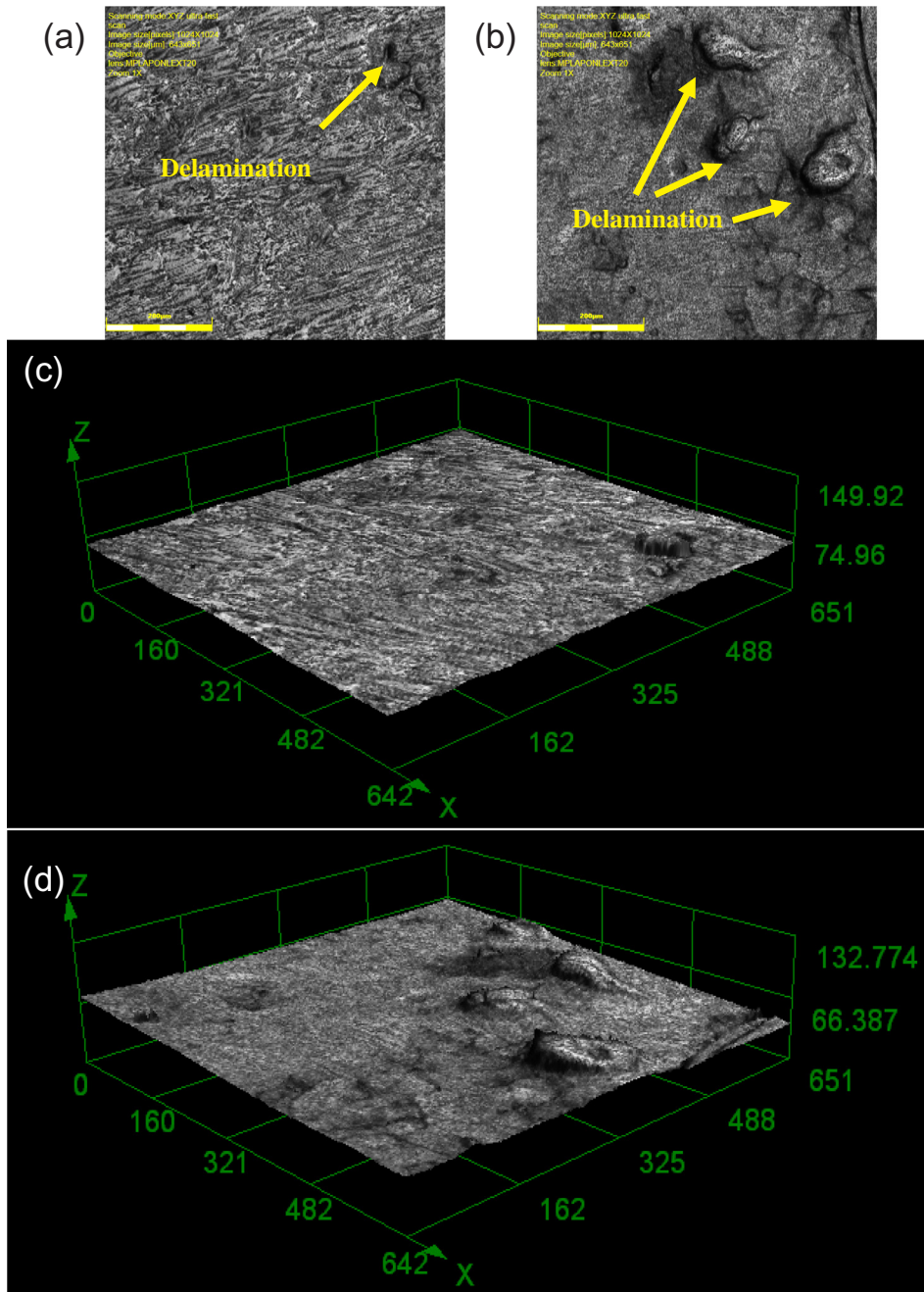
In order to understand this situation, further analysis of retrieved tibial insert was conducted by characterizing the morphological conditions of the insert surface. Surface condition of right and left tibial inserts were analysed according to modified Currier grading system [15]. The tibial inserts were assessed by scoring represented by surface roughness throughout three main zones (top, middle, and bottom as shown in Fig. 5a and b) of medial–lateral sides of tibial inserts. The surface roughness data evaluated along the medial–lateral compartment of both right and left tibial insert (Table 1) indicated that there was significant difference between right–left and medial–lateral part of the tibial inserts. The average roughness of medial part (1.665  $\mu\text{m}$ ) is higher than lateral part (1.286  $\mu\text{m}$ ) for right-12 years insert and the average roughness of medial part (32.649  $\mu\text{m}$ ) is higher than lateral part (24.838  $\mu\text{m}$ ) for 8 months insert. The average surface roughness of left-16 years insert was 94.8% higher than and right-12 years insert. Rough surface on left-16 years insert indicates surface might undergo more wear than right-12 years insert (smoother surfaces). Furthermore, in term of medial–lateral comparison, it can be clearly observed that medial side of tibial inserts of both knee possesses highest surface roughness which suggested that higher degradation occur on this medial part. We believe the greatest damage on the left-16 years insert may effect by the long implantation time. On the other hand, rough surface on the medial part was due to varus malalignment knee where the load passes to the centre of knee is correspondingly greater on the medial compartment and thus led to medial collapse [12].

However, this explanation is still limited and in order to explore the effect of varus malalignment the surface damage, we have done morphological analysis using 3D laser microscope. There are multidirectional scratching marks and delamination which more features can be observed on medial part of right-12 years insert is shown in Fig. 6. Nevertheless, for the left-16 years insert in Fig. 7, the sign of catastrophic wear become the major features dominating which resulted severe surface deformation. In this analysis (Figs. 6d and 7c), severe damages were more dominant on the medial part which has proved that wear degradation occur on these inserts are highly influenced by the present of varus deformity in regard with implantation time.

Wear damage characterization on articulating surface of both tibial inserts were then being further analysed using SEM in order to

**Table 1**  
Surface roughness measurement of UHMWPE tibial inserts.

| UHMWPE insert    | Compartment | Top                | Middle             | Bottom            | Average [ $\mu\text{m}$ ] |
|------------------|-------------|--------------------|--------------------|-------------------|---------------------------|
| Right (12 years) | Medial      | 2.106 $\pm$ 0.99   | 2.131 $\pm$ 1.05   | 0.759 $\pm$ 1.00  | 1.665 $\pm$ 0.78          |
|                  | Lateral     | 1.310 $\pm$ 0.15   | 0.943 $\pm$ 0.12   | 1.605 $\pm$ 0.22  | 1.286 $\pm$ 0.33          |
| Left (16 years)  | Medial      | 32.118 $\pm$ 17.45 | 60.630 $\pm$ 19.24 | 5.199 $\pm$ 5.06  | 32.649 $\pm$ 27.71        |
|                  | Lateral     | 4.617 $\pm$ 2.76   | 60.262 $\pm$ 39.17 | 9.634 $\pm$ 10.57 | 24.838 $\pm$ 30.78        |

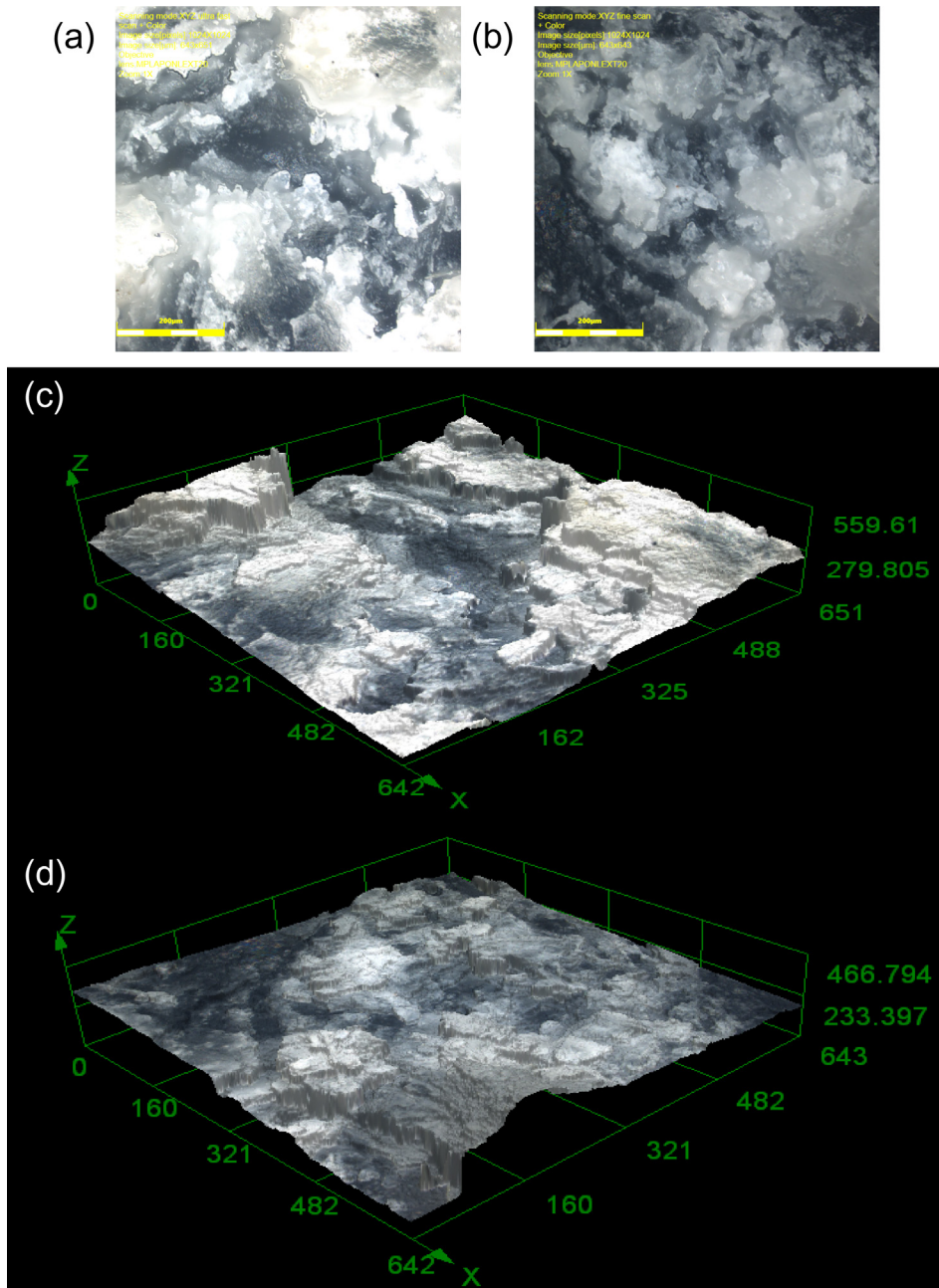


**Fig. 6.** 3D laser images taken for surface roughness measurement of right-12 years; UHMWPE tibial insert; (a and c) bottom zone of lateral compartment surface image and surface profile and (b and d) middle zone of medial compartment surface image and surface profile.

reveal the occurrence of the wear features under the influence of varus malalignment in a clearer way. This characterization was performed on high surface roughness zone on medial–lateral compartment for both right and left inserts as more wear features are expected to present on this zone. The microscopic images of wear features on medial and lateral side for both right-12 and left-16 years insert were shown in Figs. 8 and 9, respectively.

In the previous failure analysis, the retrieved polyethylene insert were assessed for seven damage modes; pitting, scratching, burnishing, third-body debris, abrasion, surface deformation and surface delamination in detecting surface damage severity [25]. The SEM evaluation revealed delamination and scratching were the most predominant damage present at all regions of both lateral and medial compartment for right-12 years shown in Fig. 8. There was also another wear features that can be seen on the medial compartment (Fig. 8e) is wave-like structure features known as ripple [22]. Many retrieval studies have characterized the degree of ‘damage’ apparent on the surfaces of retrieved polyethylene bearings for 10–12 years [8,23–24]. Medel et al. [24], reported that the



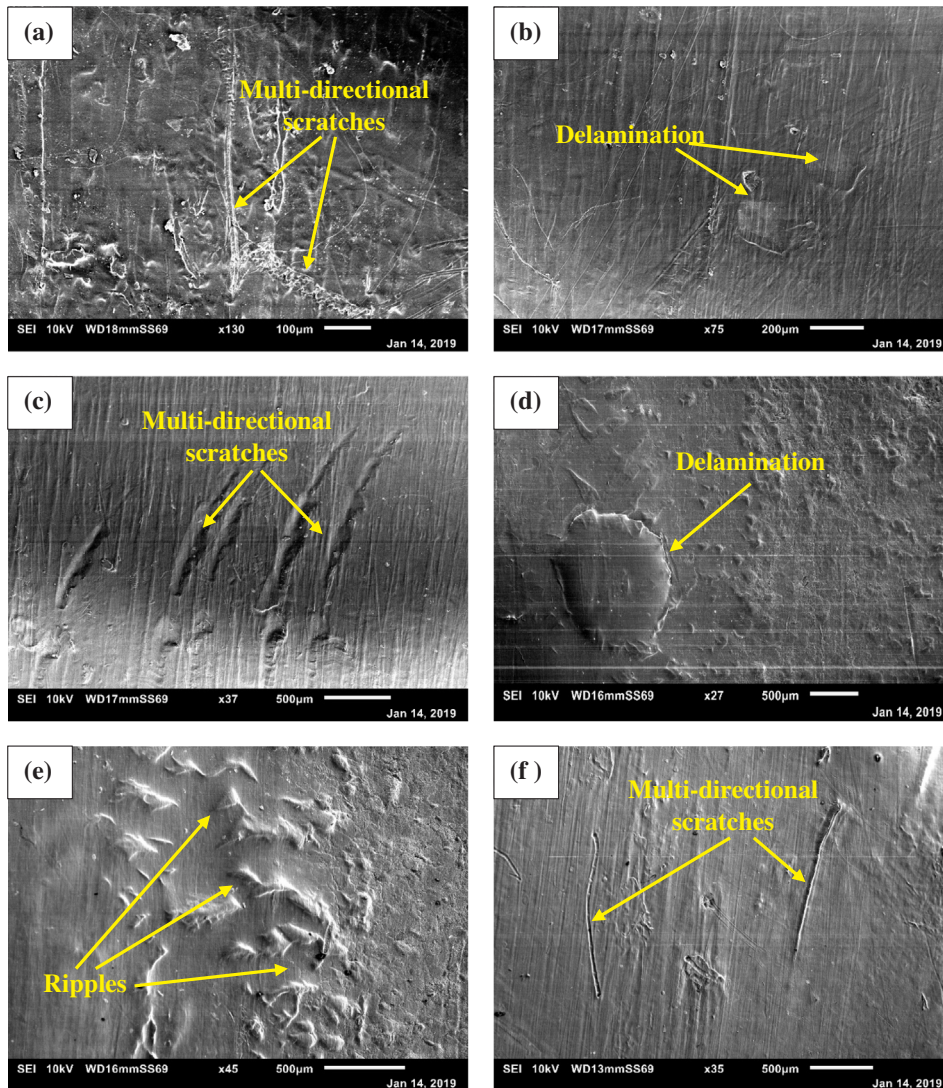


**Fig. 7.** 3D laser images taken for surface roughness measurement of left-16 years; UHMWPE tibial insert; (a and c) middle zone of medial compartment surface image and surface profile and (b and d) middle zone of lateral compartment surface image and surface profile.

delamination were significantly more prevalent and extensive at the articulating surface of polyethylene tibial inserts which implanted for 11–13 years.

Delamination known as the most severe form of polyethylene degradation, and the primary cause of failure of TKR. Delamination is a form of wear in which a large sheet of polyethylene separates from the deeper layers [25]. Delamination developed on the articulating surfaces is likely result from oxidation degradation during implantation or fatigue failure of the polythethylene as a result of repetitive cyclic loading during everyday activity [26]. In general, surface fatigue's wear appearance such as pitting and delamination is often reported for the polyethylene tibial insert [27]. In this study, it is difficult to predict when the TKR failed because of wear fatigue mechanism. This is because of the absence of pitting damage mode within the articulating surface for both compartments. Pitting has previously been associated only with third-body damage that were embedded in the articular surface and subsequently dislodged during repeated knee movement [28,29]. In addition, we could not find the appearance of debris in insert which support no accuracy of pitting. Thus, it is expected that in case of delamination formation is not related to fatigue-related failure. In general,



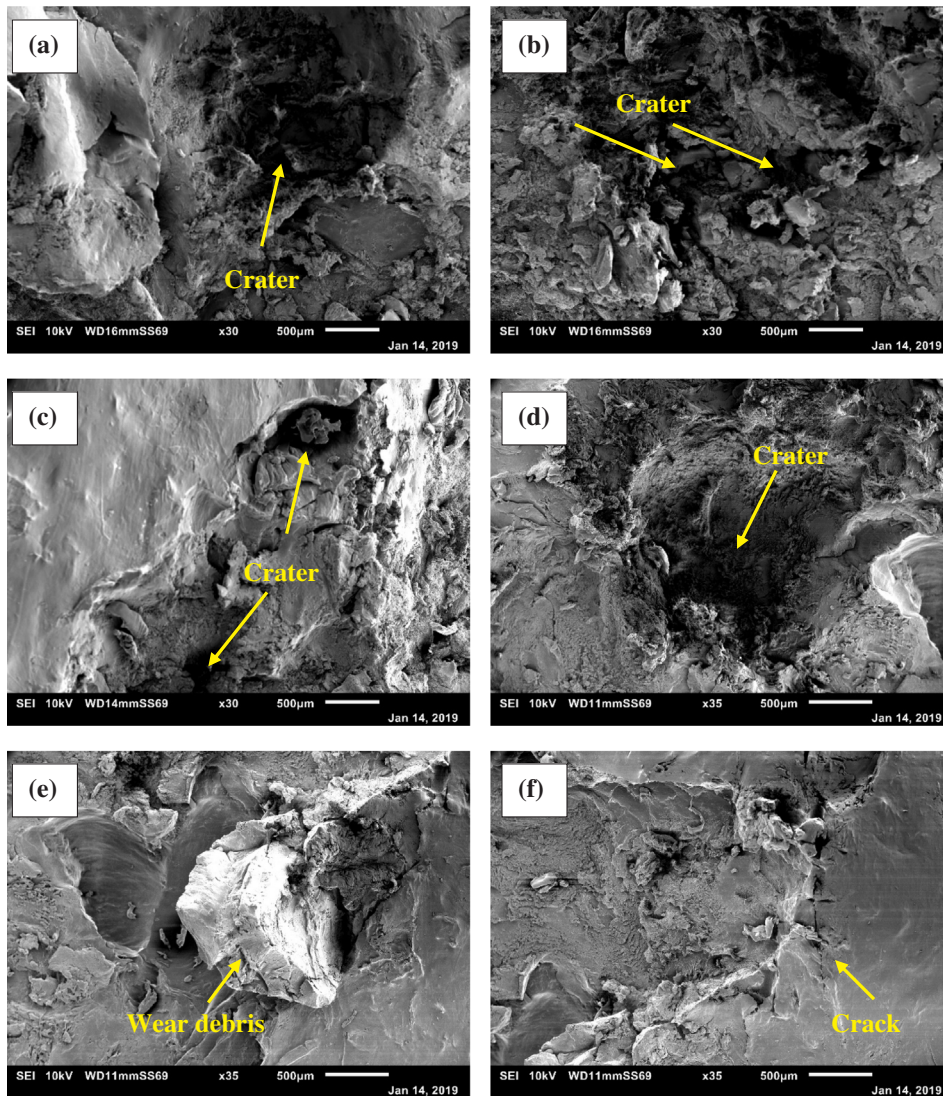


**Fig. 8.** SEM wear characteristics micrograph of right-12 years; UHMWPE insert on bottom zone of lateral compartment (a–c) and middle zone of medial compartment (d–f).

millions of cycles might be required to cause fatigue wear with the presence wear damage modes of pitting and delamination in TKR. In one simulator study performed by Bahce and Emir [30], it has been found that pitting starts appeared on the insert at  $1 \times 10^6$ , by using four axis knee joint prosthesis wear simulator. By considering the gait cycle in 12 years studied by Zahiri et al. [31], we believed these cycles can be considered a sufficient to produce pitting. However, since the patient had poor clinical function, which is inactive in daily activities indicated the 12-years insert has experienced lower gait cycle number which may also explain why pitting was not found on the articulating surface. The present findings believe that in vivo oxidation may be the primary contributor to delamination damage right 12-years insert. This finding is consistent with the previously reported finding that in vivo oxidation contributes to delamination not pitting [24].

Besides delamination, scratches tracks were the type of damage detected most frequently. The indented line was developed as a result particles separated from polyethylene which were loosened by movement between the femur and the insert component. In contrast, Bahce and Emir [30] reported that delamination can also caused scratch formation due to tangential growth of cracks on the polyethylene insert condyle surfaces. It is also interesting to consider the correlation between delamination and scratches in furthering the understanding of scratch formation. It is possible to hypothesise that delamination area and scratches formed due to separation of material in the shape of debris resulting in abrasive wear.

As previously mentioned, ripples were formed on the surface of middle zone, which the femoral component rolled on. This feature is usually associated with adhesive-abrasive wear of polyethylene in knee replacements. Ripples on polyethylene is known to be the precursor for the generation of wear particles and a higher wear mass loss of polyethylene will be induced. The present observation of ripples on the medial compartment indicated the asymmetry in surface damage which might due to the influence of malalignment



**Fig. 9.** SEM wear characteristics micrograph of left-16 years; UHMWPE insert on middle zone of medial compartment (a–c) and middle zone of lateral compartment (d–f).

which later contributed to higher surface roughness. However, we were unable to confirm the relationship of ripples damage mode and malalignment due to lack of complete understanding of the wear behavior of TKR. Interestingly, Cerquiglini et al. [32], found that only severe malposition can lead to asymmetry in surface damage of polyethylene tibial inserts. In this study, it appears that the medial surface of the component is covered with scratches, delamination and ripples led to increase of surface roughness - this is intended to promote asymmetry in surface damage of 12-years polyethylene tibial inserts.

Surface analysis of the left-16 years insert displayed severe surface deformation through the formation of crater which known as catastrophic wear. Crater can be defined as uneven surface formation due to uneven removal of surfaces bearing area by means of adhesion wear. Particulate debris is reported responsible in the formation of craters [33]. When the particle is completely disintegrated and remove from the surface, a crater is left behind. The formation of wear debris can be clearly seen on Fig. 9e. Hood et al. [21] described that wear debris can be recognized by difference in colour and/or texture within the polyethylene insert. However, further analysis of this wear debris cannot be made due to gold coating layer apply prior to sample preparation requirement for the SEM characterization may change the elemental properties of the debris. Particulate debris are not the only reason involved in crater formation, low material properties such as low molecular weight fraction at the articulating surface can fatigue and fracture when exposed to the cyclical stresses which in turn formation of craters and crack [33,34]. From the Fig. 9f, it can be obviously observed that severe surface cracking occurs on the insert. No difference of damage mode was noted across the medial and lateral compartment for 16 years insert.

The surface damage analysis in this study observed the delamination could be seen clearly on both lateral and medial compartment for the first insert (right-12 years). In the second insert (left-16 years) the crater was dominant on both lateral and medial

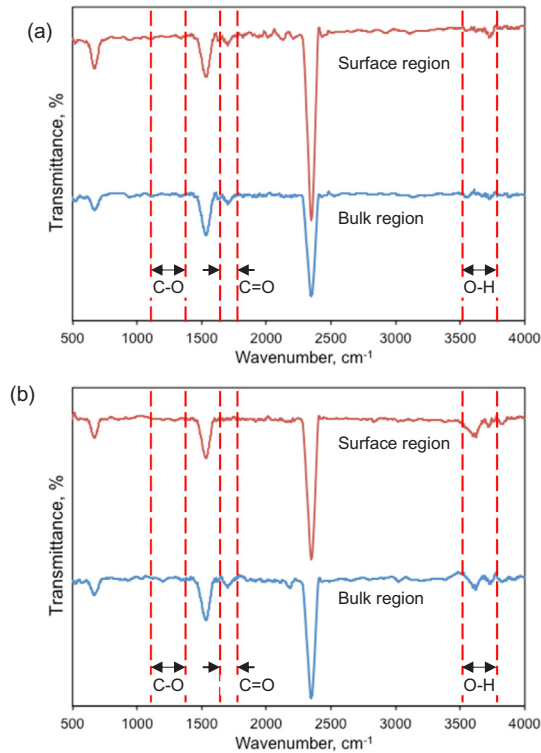


Fig. 10. ATR-FTIR spectra for (a) right-12 years; and (b) left-16 years UHMWPE tibial inserts.

compartment. Since both of the UHMWPE inserts were implanted in long years (left-16 years and right-12 years), delamination and crater formations found in these inserts are likely to correlate with the influenced of oxidative degeneration. Oxidation process may occur in term of these two main mechanism which firstly the residual free radical of UHMWPE insert would provide site for oxidation and react with dissolved oxygen in body fluids, while second mechanism suggested that free radical species present in synovial fluid triggered the oxidation to occur in vivo and consequently affect the UHMWPE insert itself [17]. This second mechanism highlight the free radical initiators that provided by the body system itself instead of the presence of free radicals within the UHMWPE for in vivo degradation to take place.

However, identification of mechanism involve for in vivo oxidation is beyond our research scope and for this current research, quantitative evidence of oxidation characterization will instead be determine using ATR-FTIR. This information would be beneficial in order to reveal significant oxidation degradation of UHMWPE insert occur in vivo application and thus support the hypothesis of the degradation of mechanical properties of the insert occurred along with the implantation.

Fig. 10 shows the FTIR spectra of UHMWPE tibial insert of surface and bulk region of both right and left UHMWPE tibial insert. There was no significant difference detected on both regions of each knee insert although few differences can be observed when comparing the spectra between right and left knee inserts. Several absorption peak indicate the product of oxidation were reflected on spectra of both region of right and left UHMWPE insert which is at  $1043\text{ cm}^{-1}$  (anhydride  $\text{CO}-\text{O}-\text{CO}$ ),  $1208\text{ cm}^{-1}$  (aromatic ester  $\text{C}-\text{O}$ ),  $1681\text{ cm}^{-1}$  (ketone  $\text{C}=\text{O}$ ),  $1703\text{ cm}^{-1}$  (amide  $\text{C}=\text{O}$ ),  $1350$  and  $1410\text{ cm}^{-1}$  (carboxylic acid  $\text{O}-\text{H}$ ). Additionally, there are some extra evident peak observed on left UHMWPE insert spectra which can be assigned to oxidation at  $3599$  and  $3629\text{ cm}^{-1}$  (alcohol  $\text{O}-\text{H}$ ) which mainly highlight the differences between these both inserts [16,35]. From this FTIR result, it can be confirmed that oxidation has occurred on both UHMWPE inserts which consequently lead to change of properties on both inserts and thus can be correlated with the occurrence of surface damage features mention on previous paragraph.

Consequently, further justification on oxidative degradation were carried out by measured the changes of crystallinity and molecular weight of UHMWPE insert of bilateral total knee replacement after 16 years (left knee) and 12 years (right knee) in vivo environment. The crystallinity of both right and left UHMWPE insert were obtained from DSC by intergrating the area under the melting endotherm and comparing with the heat of fusion of 100% crystalline UHMWPE. The degree of crystallinity of left UHMWPE insert display higher value of crystallinity which is 66.99% compare to right insert which is 56.52% as shown in Fig. 11. This may due to higher oxidation reactions of left insert with carbon bonding that leads to recrystallization of broken chains and thus devote to increasing in crystallinity [31]. In addition, it can be concluded that higher crystallinity would produce more wear debris as the macromolecular chains are shorter [22]. Despite of that, discussion of crystallinity changes in vivo application could not be provided due to insufficient original crystallinity data of both insert. Yet, the crystallinity percentage of both UHMWPE insert stills falls within the acceptable medical range which is from 39% to 75% [36].



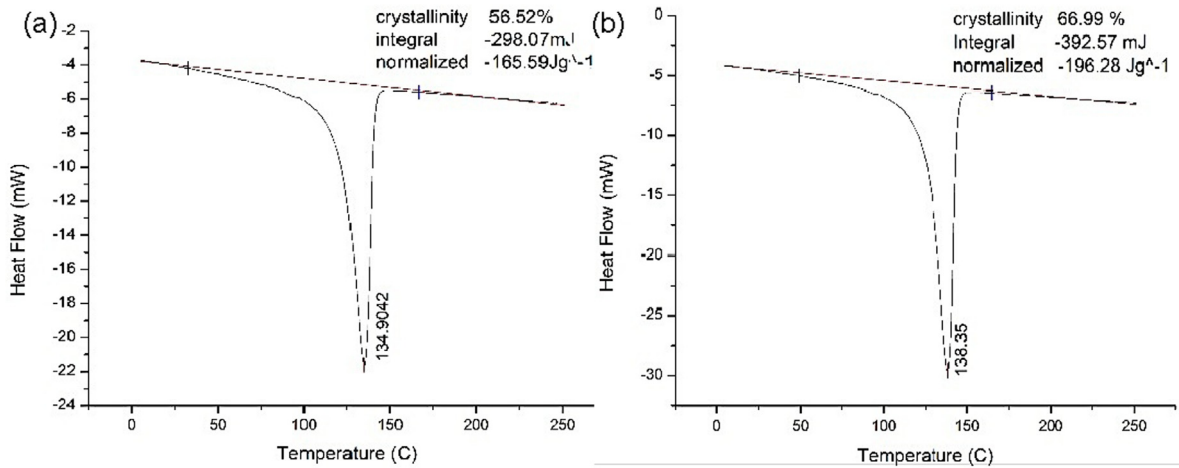


Fig. 11. Plot of heat flow versus temperature for (a) right-12 years; and (b) left-16 years UHMWPE tibial inserts.

On the other hand, the molecular weight of retrieved UHMWPE inserts for both right and left knee after in vivo implantation were measured by using GPC as presented in Table 2. High reduction of molecular weight from standard molecular weight of medical grade polyethylene which is  $1.5 \times 10^6$  g/mol [36] can be observed on both UHMWPE inserts. Molecular weight of 2103 g/mol of right insert and 1960 g/mol of left insert after in vivo application cause reduction approximately of 99.86% and 99.87% respectively on both knee inserts suggested that polyethylene properties has changes throughout years of implantation and thus consequently affected the mechanical and wear degradation of the inserts.

#### 4. Conclusions

Based on the morphological and oxidation related analysis, following conclusions can be drawn:

- Both left UHMWPE insert (16 years) and right UHMWPE insert (12 years) exhibit wear degradation which severity of damage are more prominent on left insert due to longer implantation time.
- (a) Severe surface deformation which also known as catastrophic wear of left inserts causes demolishing of other wear features. However, the presence of particulate debris still can be observed on the surface of insert. The present of particulate debris resulting crater formation and increase the surface roughness
- (b) Wear features present on right insert are mainly delamination and multiple scratching.
- (c) Extension of wear degradation (high surface roughness) due to uneven load distribution of varus malalignment (load pass medially) with respect to implantation time proposed that severe wear modes are prominent on medial compartment compare to lateral for both UHMWPE inserts.
- Significant correlation was found between surface degradation and oxidation degradation of inserts associate with higher crystallinity and higher molecular weight reduction of inserts which likely to occur in regards with implantation time.

#### Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Table 2

Molecular weight of UHMWPE tibial inserts from GPC analysis.

| UHMWPE insert    | Elution volume [ml] | Retention time [min] | Adjusted RT [min] | Mn   | Mw [g/mol] | MP   | Mz   | MZ + 1 | Mz/Mw |
|------------------|---------------------|----------------------|-------------------|------|------------|------|------|--------|-------|
| Right (12 years) | 24.832              | 24.832               | 24.832            | 1855 | 2103       | 2115 | 2345 | 2582   | 1.114 |
| Left (16 years)  | 24.912              | 24.912               | 24.912            | 1512 | 1960       | 2030 | 2260 | 2501   | 1.153 |



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