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Original Article

# Setting kinetics and mechanical properties of flax fibre reinforced glass ionomer restorative materials

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## Abstract

Regardless of the excellent properties of glass ionomer cements, their poor mechanical properties limit their applications to non-load bearing areas. This study aimed to investigate the effect of incorporated short, chopped and randomly distributed flax fibers (0, 0.5, 1, 2.5, 5 and 25 wt%) on setting reaction kinetics, and mechanical and morphological properties of glass ionomer cements. Addition of flax fibers did not significantly affect the setting reaction extent. According to their content, flax fibers increased the compressive (from 148 to 250 MPa) and flexure strength (from 20 to 42 MPa). They also changed the brittle behavior of glass ionomer cements to a plastic one. They significantly reduced the compressive (from 3 to 1.3 GPa) and flexure modulus (from 19 to 14 GPa). Accordingly, flax fiber-modified glass ionomer cements could be potentially used in high-stress bearing areas.

**Keywords:** setting kinetics, flax fibre, glass ionomer cements

## Introduction

Due to their adhesive properties and anticariogenic action, conventional glass ionomer cements (GICs) have been widely used with best clinical outcomes as liners, bases, and filling materials in low stress bearing areas as cervical cavities. GICs also have favorable biologic properties and a similar coefficient of thermal expansion to dental tissues. GICs, however, have some drawbacks including high solubility<sup>[1]</sup>, relatively poor mechanical properties<sup>[2]</sup>, poor fracture toughness and long setting time<sup>[3]</sup>.

A series of modifications to the GIC powder or liquid component have been attempted to improve their

properties. Resin modified GICs (RMGICs) are an example of liquid modification that involve the addition of polymerizable hydrophilic monomers, such as 2-hydroxyethyl methacrylate (HEMA), to the polyacrylic acid liquid of conventional GICs. This modification improved the mechanical properties and resistance of GICs. The presence of uncured monomer, however, in RMGICs increased the concern about its biocompatibility with pulpal tissues<sup>[4–6]</sup>. Metal-reinforced GICs, as an example of powder modification, involve the addition of silver-amalgam alloy powder to conventional GICs powder; it increased the mechanical properties, reduced solubility, and induced radio-opacity to GICs. Metal-reinforced GICs, therefore,

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employed to restore cavities in high stress-bearing areas, e.g., Class II. The addition of metals to GICs powder, however, reduced fluoride release and bond strength to tooth structure<sup>[7]</sup>.

Experimental GICs, based on niobium modified calcium fluoro-alumino-silicate glass powders, were prepared by sol-gel method in an attempt mainly to improve the mechanical properties and degradation resistance<sup>[8–10]</sup>. These formulations have been initially shown to be biocompatible when injected subcutaneously, but further research is still required to verify their biocompatibility in the presence of dentin-pulp complex<sup>[11]</sup>. One of the latest attempt involves the incorporation of titanium dioxide nanoparticles into GICs' powder to improve the mechanical properties, as well as to induce antibacterial activity<sup>[12]</sup>.

The worldwide use of natural products for pharmaceutical purposes has increased over the last few decades. Flax fibers, as an example of biologic material (lingo-cellulosic), have been considered as multipurpose use fibers. They are commonly used for both industrial, e.g., automotive and construction industry<sup>[13]</sup>, and non-textile use, mainly in composites<sup>[14]</sup>. These fibers are renewable, nonabrasive and strong; their natural origin eliminates any concern with health and safety during handling and application<sup>[13]</sup>. Accordingly, their use has recently been extended to biomedical applications<sup>[15,16]</sup>. Due to their antibacterial action, genetically modified flax fibers were used as wound dressings for treatment of infection<sup>[15]</sup>. The development of biologically active dressings has also been attempted by immobilization of the required drug into flax fibers; this resulted in better clinical outcomes and reduction in both treatment time and post-traumatic suppuration<sup>[17–18]</sup>. For dental use, flax fibers were effective in improving compressive strength while reducing solubility and micro-leakage of zinc oxide eugenol cement<sup>[19]</sup>.

Through this study, short chopped flax fibers were randomly incorporated at different weights into GICs restorative materials' matrix to control GICs' mechanical properties. The setting kinetics of GICs, however, represents one of the main issues that would be considered when designing new materials. The aim of this study was, therefore, to study the effect of flax fiber incorporation on setting reaction/kinetics, mechanical and morphological properties of GICs using Perkin Elmer ART-FTIR, Instron testing machine and SEM, respectively. The two null hypotheses were: (1) there was no significant difference in the setting reaction/kinetics of GIC and flax fiber modified GIC; (2) there was no

significant difference in the flexure and compressive properties of GIC and flax fiber modified GIC.

## Materials and methods

### Reagent

Natural short flax fibers and glass ionomer filling materials (Ketac™ Fil Plus, 3M ESPE, Germany, shade 3.5A) were used for this study. Fibers were included at 0, 0.5, 1, 2.5, 5 and 25 wt %. These were coded as GIC, 0.5FFRGIC, 1FFRGIC, 2.5FFRGIC, 5FFRGIC and 25FFRGIC. The powder/liquid (P/L) ratio was fixed at 3:1. The required amount of powder and liquid was weighted using an electronic balance (Shimadzu Corporation, Tokyo, Japan). For proper distribution of flax fibers, they were properly mixed with the powder before the addition of liquid. The cement was mixed according to the manufacturer's instruction.

### Fourier transform Infra-Red and setting reaction monitoring

The setting reaction kinetics of each formulation has been studied using ATR-FTIR spectrometry (Perkin Elmer Series 2000, UK) at 37°C. The mixed cement was placed in a rubber ring of 8 mm diameter and 1 mm depth positioned centrally on the Golden Gate Single Reflection Diamond ATR. With this technique, the bottom few micron of each sample contacting the diamond are analyzed. The top surface of each sample, however, was initially sealed with an acetate sheet to avoid possible dehydration of the samples. FTIR spectra between 500 and 4,000 cm<sup>-1</sup> were then obtained using Timebase software with a resolution of 4 cm<sup>-1</sup> every 12 seconds for 30 minutes. The reaction extent ( $\zeta$ ) versus time was calculated from the absorbance change ( $\Delta A$ ) and the maximum absorbance change ( $\Delta A_{\max}$ ) at 1565 cm<sup>-1</sup> using the following equation<sup>[20]</sup>:

$$\zeta = \frac{\Delta A}{\Delta A_{\max}} = \frac{A_t - A_0}{A_f - A_0}$$

Where  $A_0, A_t$  and  $A_f$  are the absorbance initially, at time  $t$  and finally.

Spectra of the starting materials (Ketac™ Fil Plus powder and liquid, as well as the flax fibers) were also obtained.

### Compression test

Disc shaped specimens ( $n = 6$ ) of 10 mm diameter and 3 mm thickness were prepared from each composition using a transparent rubber mold that can be easily

removed from the material after its setting. The rubber mold was set on the top of a glass slab and an acetate sheet. The material was inserted into the mold with a slight excess of material used to fill the mold. Another acetate sheet was then placed on the top of the filled mold; the material surface was pressed flat with another glass slab. The glass slab was held firmly in place for a few minutes to avoid air bubbles from forming and to obtain a flat and smooth surface. After ~15 minutes, the celluloid strips and glass slabs, as well as excess material were removed. The surface of each disc, however, was left untouched. The samples were then kept in a sealed plastic bag at room temperature for 1 week before testing.

The dimensions of each specimen were measured with a micrometer prior to testing using the Universal testing machine (Instron 5969, USA) at a cross head speed of 0.25 mm/minutes and 50 kN load cell. The compressive strength, modulus and failure strain were recorded using Bluehill 3 software.

### Three-point bending test

Rectangular specimens ( $n = 6$ ) of  $25 \times 2 \times 2$  mm<sup>3</sup> were used to measure the flexure strength and modulus of each composite using the three-point bending test. Samples were prepared as described above for the compression test.

The dimensions of each specimen was measured with a micrometer prior to testing using a Universal testing machine (Instron 5944, USA) at a cross head speed of 0.25 mm/minutes and 2 kN load cell. The flexure strength ( $\sigma_f$ ) in MPa and modulus ( $E_f$ ) in GPa were calculated using Bluehill 3 software.

### Scanning electron microscopy

For scanning electron microscopy, samples were mounted on aluminum stubs, sputter coated with gold-palladium alloy, and viewed using a Stereoscan 90B scanning electron microscope at 15kV (Cambridge instruments Ltd., UK).

### Statistical analysis

Numerical data was presented by mean  $\pm$  standard deviation. Due to heterogeneity of variance, the data was analyzed using the non-parametric Kruskal-Wallis test. Since Kruskal-Wallis testing does not identify where the stochastic dominance occurs, a pairwise comparison between groups was then carried out to test the significant differences between them. The statistical analysis was carried out using IBM SPSS for Windows (SPSS 20, SPSS Inc., Chicago, IL, USA). The significance level was set at 0.05.

## Results

### Fourier transform infrared and setting reaction monitoring

#### Starting components

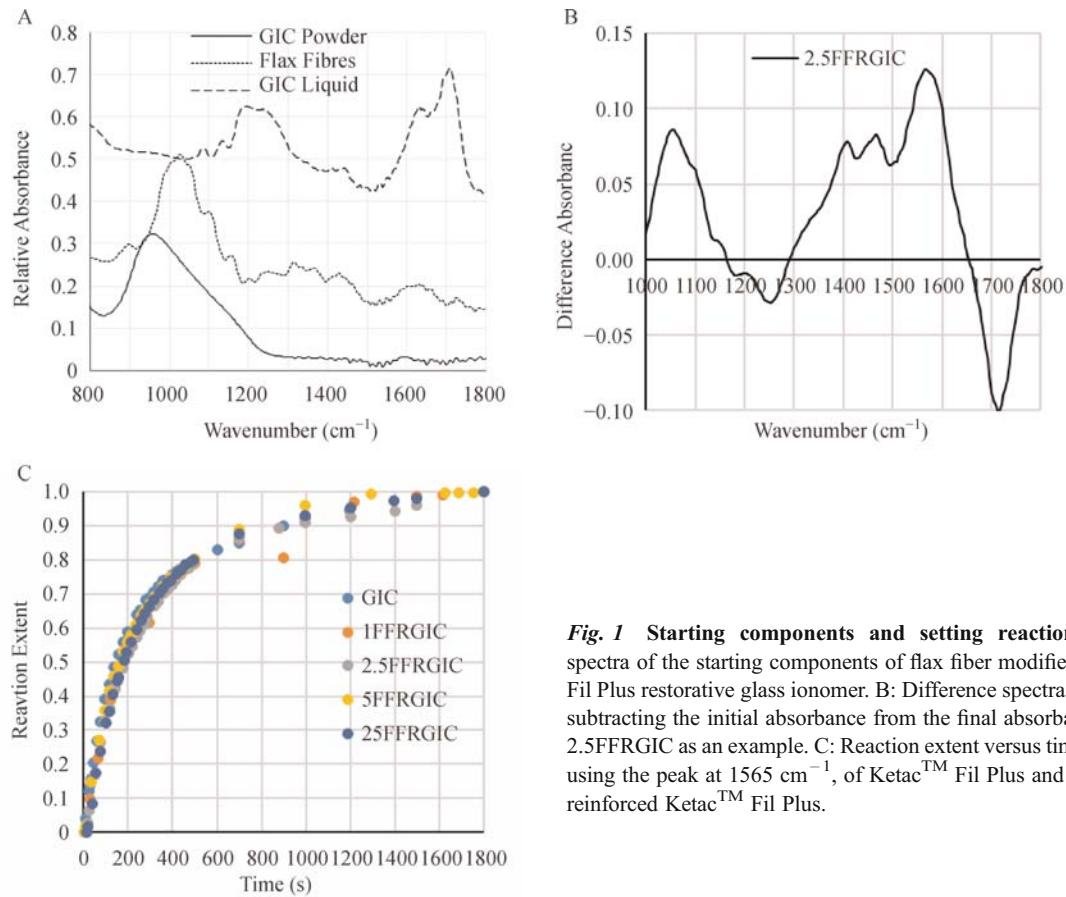
**Fig. 1A** shows the FTIR spectra of the starting components of flax fiber modified formulations. The powder of Ketac<sup>TM</sup> Fil Plus is characterized by the presence of peaks at 540 and 970 cm<sup>-1</sup> that could be assigned to Si-O stretch<sup>[21]</sup>. The liquid has sharp peaks at 1,249, 1,635, 1,707 and 3,380 cm<sup>-1</sup>. The shoulder at 1,635 cm<sup>-1</sup> O-H stretch of water<sup>[21]</sup> while the peaks at 1,249 cm<sup>-1</sup> at 1,707 cm<sup>-1</sup> are due to C-O and C=O stretch of polyacrylic acid, respectively<sup>[21]</sup>. Flax fibers have strong peaks at 540 and 1,027 cm<sup>-1</sup>. The peak at 1,027 could be assigned to out-of-plane bending  $\gamma$ (O-H.....O) vibration of the intermolecular hydrogen bonds<sup>[22]</sup>. A very broad peak in the region 2,500-3,600 cm<sup>-1</sup> was assigned to the -OH vibration of several hydroxyl groups in the cellulose matrix<sup>[23]</sup>.

#### Setting reaction

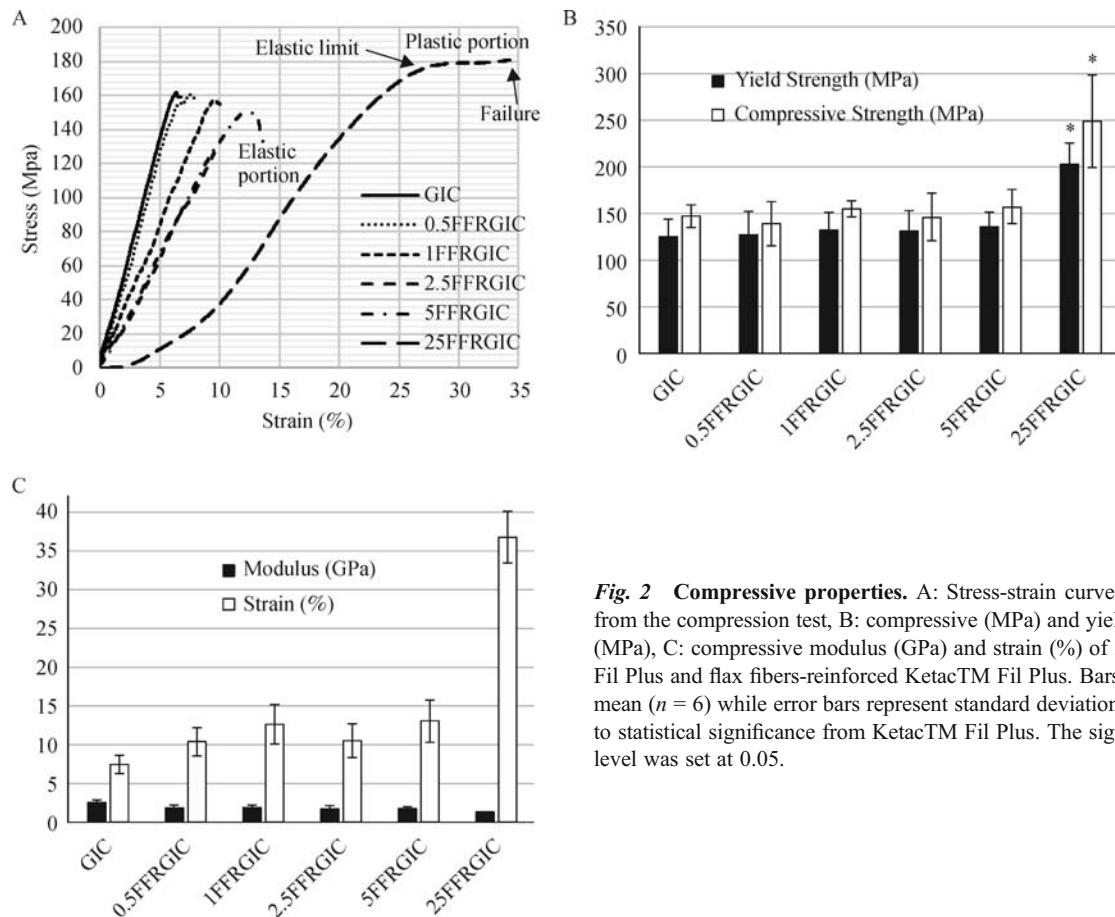
An example of change in the FTIR spectrum as the GIC reaction progresses is provided in **Fig. 1B**. The different spectra obtained have peaks and troughs in the same positions irrespective of flax addition or time of reaction and are characteristic of a glass ionomer setting reaction. As is the case with all cements, substantial changes in the spectra were observed during the first 30 seconds. The troughs seen at 1,249 and 1,707 cm<sup>-1</sup> can be assigned to C-O and C=O stretch bands of polyacrylic acid which are shifted when it is neutralized<sup>[21,24]</sup>. By comparing the intensity of 1,707 cm<sup>-1</sup> as the time proceeds, nearly 50% and 90% of polyacrylic acid has been consumed in the acid-base reaction by the first 2 and 10 minutes, respectively. The loss of intense polyacid peaks coincided with the gradual appearance of broad symmetric (1,400 and 1,470 cm<sup>-1</sup>) and asymmetric (1,565 cm<sup>-1</sup>) COO polyacrylate salt peaks<sup>[21]</sup>. The addition of flax fibers had no significant effect on either the different spectra or on the calculated reaction extent (**Fig. 2C**).

#### Compressive strength

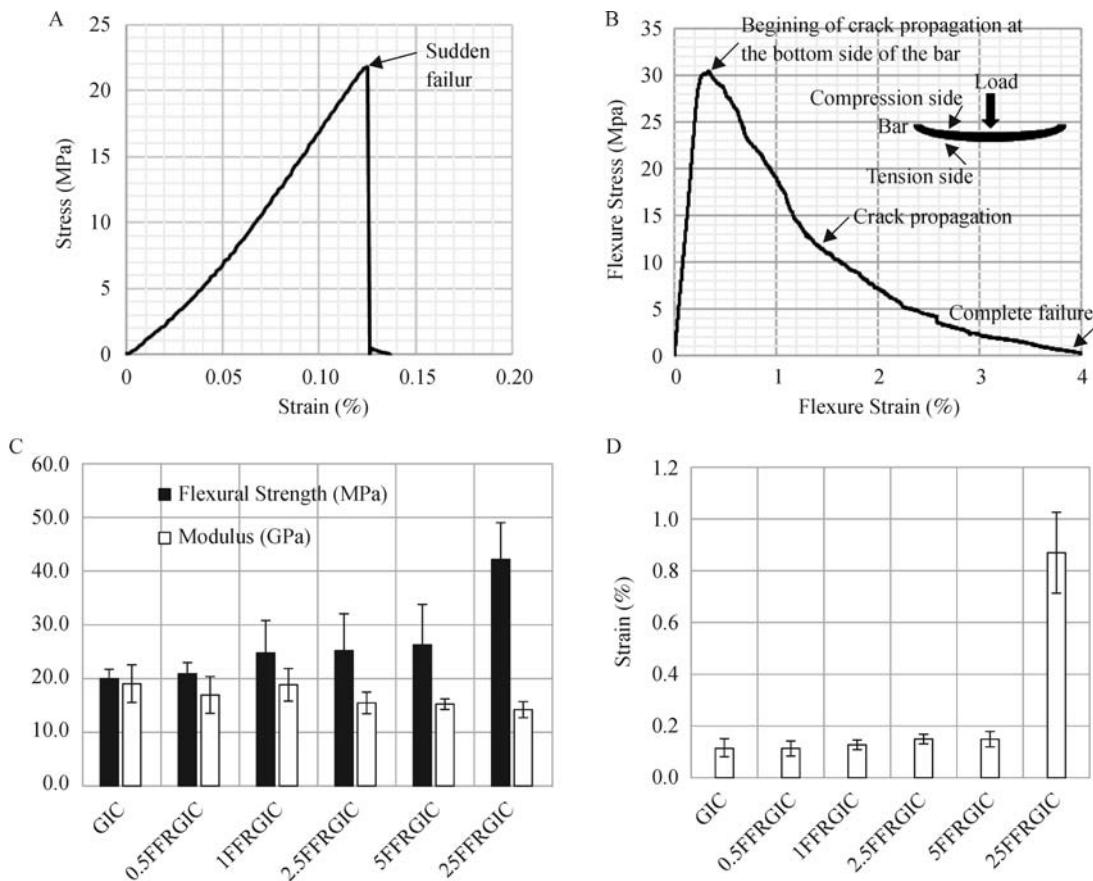
**Fig. 2A** shows the stress-strain curves of Ketac<sup>TM</sup> Fil Plus and flax fiber reinforced formulations. The stress-strain curve of Ketac<sup>TM</sup> Fil Plus is characterized by a linear elastic region terminated by peak stress called



**Fig. 1 Starting components and setting reaction.** A: FTIR spectra of the starting components of flax fiber modified Ketac<sup>TM</sup> Fil Plus restorative glass ionomer. B: Difference spectra, obtained by subtracting the initial absorbance from the final absorbance, for 2.5FFRGIC as an example. C: Reaction extent versus time, measured using the peak at  $1565 \text{ cm}^{-1}$ , of Ketac<sup>TM</sup> Fil Plus and flax fibers reinforced Ketac<sup>TM</sup> Fil Plus.



**Fig. 2 Compressive properties.** A: Stress-strain curves obtained from the compression test, B: compressive (MPa) and yield strength (MPa), C: compressive modulus (GPa) and strain (%) of Ketac<sup>TM</sup> Fil Plus and flax fibers-reinforced Ketac<sup>TM</sup> Fil Plus. Bars represent mean ( $n = 6$ ) while error bars represent standard deviations. \* refers to statistical significance from Ketac<sup>TM</sup> Fil Plus. The significance level was set at 0.05.



**Fig. 3** Flexural properties. Stress-strain curves of (A) KetacTM Fil Plus and (B) 5wt% flax fibers-reinforced GIC as obtained from the three-point bending test as an example to show the ductile behavior of fibers-reinforced formulations with 5 and 25wt. % fibers content. The inset represent the compression and tension side of the bar under three-point bending; the failure normally starts at the tension (bottom) side (C) Flexure strength (MPa) and modulus (GPa). (D) Flexure strain (%) of KetacTM Fil Plus and flax fibers-reinforced KetacTM Fil Plus. Bars represent mean ( $n = 6$ ) while error bars represent standard deviations. \* refers to statistical significance from KetacTM Fil Plus. The significance level was set at 0.05.

'elastic limit' where the failure occurred. In such cases, the compressive strength is equal to the elastic limit of the material. This type of behavior is described as brittle. Generally, KetacTM Fil Plus is stiff, strong and brittle (i.e., no yielding before failure). The addition of flax fibers changed the stress-strain behavior of the material, where the materials showed yielding before failure. This region of yielding is called plastic region; the extent of yielding increased in proportion to the fibers' content. The failure mode changed from being brittle to plastic as the content of fibers increased. This type of failure becomes clearly evident with 5FFRGIC and 25FFRGIC. Furthermore, a toe-in region, at the beginning of load application, became evident in the stress-strain behavior of 25FFRGIC. Generally, the flax-modified formulations are resilient, strong and ductile.

The addition of flax fibers produced an increase in both compressive and yield strength. This increase, however, was not statistically significant ( $P > 0.05$ ) for all formulations except 25FFRGIC ( $P = 0.002$  and  $0.001$  for compressive and yield strength respectively).

when compared with KetacTM Fil Plus (Fig. 2B). The compressive strength increased from  $148 \pm 12$  for Ketac Fil Plus to  $250 \pm 50$  MPa for 25FFRGIC. All fiber reinforced formulations, however, showed statistically significant difference from 25FFRGIC. As shown by the pairwise comparison, there was a statistically significant difference between 0.5FFRGIC and 25FFRGIC ( $P = 0.000$ ), between 1FFRGIC and 25FFRGIC ( $P = 0.018$ ), between 2.5FFRGIC and 25FFRGIC ( $P = 0.001$ ), and between 5FFRGIC and 25FFRGIC ( $P = 0.043$ ). Regarding the yield strength, all fiber reinforced formulation showed significantly different statistics from 25FFRGIC. As shown by the pairwise comparison, there was a statistically significant difference between 0.5FFRGIC and 25FFRGIC ( $P = 0.002$ ), between 1FFRGIC and 25FFRGIC ( $P = 0.006$ ), between 2.5FFRGIC and 25FFRGIC ( $P = 0.003$ ), and between 5FFRGIC and 25FFRGIC ( $P = 0.012$ ).

On the other hand, a significant linear reduction in Young's modulus was observed when incorporating flax fibers into KetacTM Fil Plus (a reduction from  $2.6 \pm 0.4$

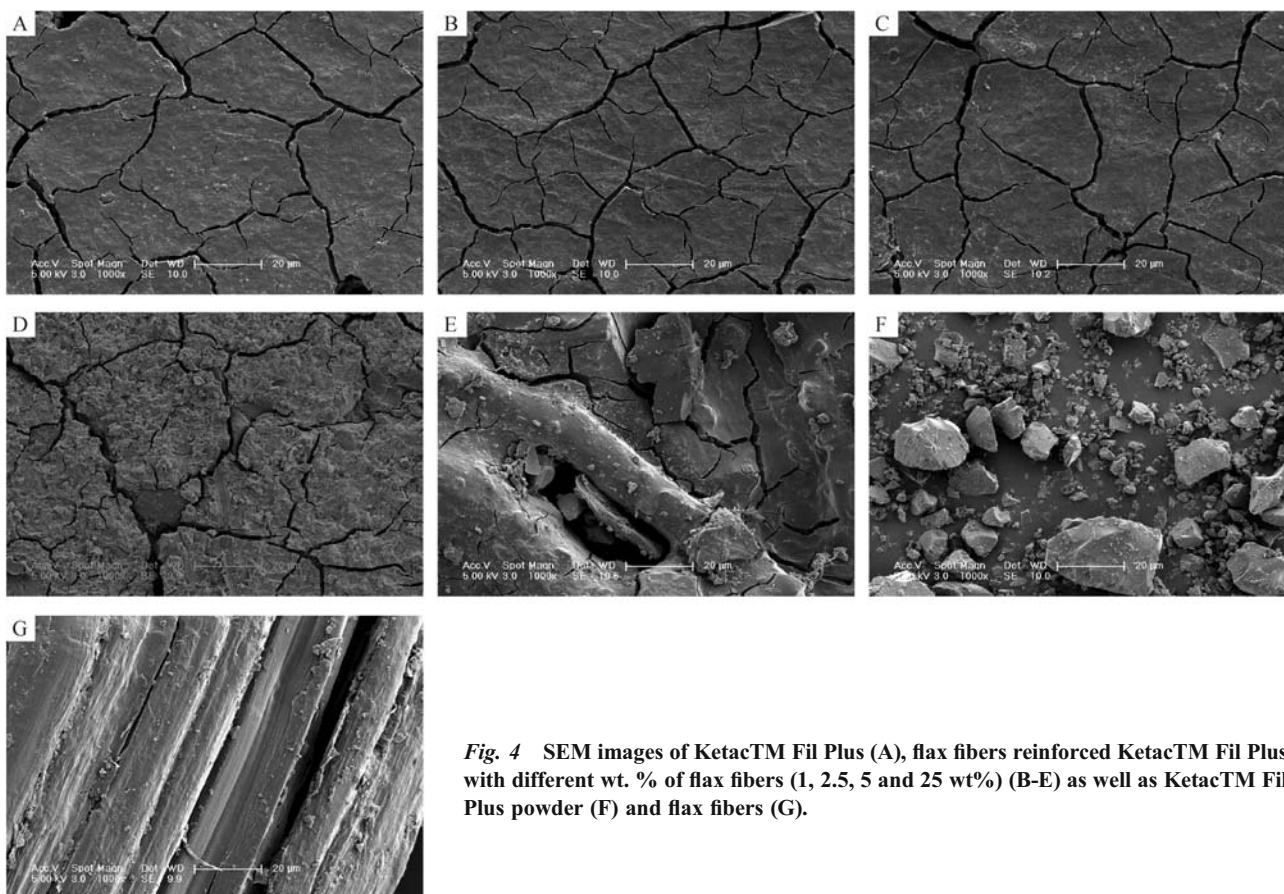
GPa for Ketac Fil Plus to  $1.5 \pm 0.3$  GPa 25FFRGIC) (**Fig. 2C**). All fiber reinforced formulation showed significantly lower modulus than GIC. The *P* values were 0.007, 0.042, 0.19, 0.013 and 0.000 for 0.5FFRGIC, 1FFRGIC, 2.5FFRGIC, 5FFRGIC and 25FFRGIC when compared with GIC. There was also a statistically significant difference between 1FFRGIC and 25FFRGIC (*P* = 0.036).

The reduction in modulus was associated with a significant linear increase in strain (%) recorded at the material failure (**Fig. 2C**). A pairwise comparison test showed that, statistically, there is a significant difference GIC and 1FFRGIC (*P* = 0.007), between GIC and 5FFRGIC (*P* = 0.003) and between GIC and 25FFRGIC (*P* = 0.000). There was also a significant difference between some flax fiber reinforced formulations (0.5FFRGIC, 1FFRGIC and 2.5FFRGIC) and 25FFRGIC; the "P" values are 0.001, 0.042 and 0.003 respectively.

#### Flexural strength

**Fig. 3** shows the brittle failure of Ketac<sup>TM</sup> Fil Plus under the three-point bending test. Similar behavior was relatively seen for 0.5FFRGIC, 1FFRGIC and 2.5FFRGIC. **Fig. 3B**, however, shows the characteristic

plastic behavior of formulations with high flax fiber content, 5FFRGIC and 25FFRGIC. As expected, the failure under the three-point bending condition normally started at the tension side. This failure was catastrophic once the elastic limit of the material had been approached; this was observed for Ketac<sup>TM</sup> Fil Plus. Addition of flax fibers, however, increased the strain % before failure; this behavior was clearly seen for formulations with 5 and 25 wt%. This type of behavior was expected for fiber-reinforced formulations. While the flexural strength and strain linearly increased, the modulus decreased with increased fiber content (**Fig. 3C and D**). Kruskal-Wallis testing showed that the addition of flax fibers produced a statistically significant change in flexural strength (*P* = 0.001), strain (*P* = 0.005) and modulus (*P* = 0.004) when compared with GIC. So there is strong evidence to reject the null hypothesis. Only 5FFRGIC and 25FFRGIC showed a significantly higher strength than GIC; the *P* values are 0.037 and 0.000 respectively. The remaining fiber reinforced formulations (0.5FFRGIC, 1FFRGIC and 2.5FFRGIC) showed higher strength than GIC, but this increase was not statistically significant (*P* > 0.05). Furthermore, there was a statistically significant difference in flexural strength of 0.5FFRGIC, 1FFRGIC, 2.5FFRGIC and 5FFRGIC when compared with GIC;



**Fig. 4** SEM images of Ketac<sup>TM</sup> Fil Plus (A), flax fibers reinforced Ketac<sup>TM</sup> Fil Plus with different wt. % of flax fibers (1, 2.5, 5 and 25 wt%) (B-E) as well as Ketac<sup>TM</sup> Fil Plus powder (F) and flax fibers (G).

the *P* values were 0.001, 0.012, 0.012 and 0.034, respectively.

Regarding the flexural modulus, only 2.5 and 25 showed significantly lower modulus than GIC; the *P* values are 0.001 and 0.045, respectively. Also both 0.5FFRGIC and 1FFRGIC showed significantly higher modulus than 25FFRGIC; the *P* values are 0.025 and 0.001 respectively. Both 2.5FFRGIC and 5FFRGIC showed significantly lower modulus than 1FFRGIC; the *P* values are 0.028 and 0.032, respectively.

Regarding the strain, only 25FFRGIC showed significantly higher strain than GIC; the *P* value was 0.002. Furthermore, 0.5FFRGIC, 1FFRGIC, 2.5FFRGIC and 5FFRGIC showed significantly lower strain than 25FFRGIC; the *P* values were 0.002, 0.000, 0.002 and 0.002, respectively.

Comparing the compressive strength with that of flexure strength (that mostly measure the tensile strength); all tested materials had flexure strength around 6-8 times lower than the compressive strength. This indicates the brittle nature of these materials. The ratio of compressive: flexure strength decreased with increasing the filler contents indicating the move from brittle to plastic nature (or indicating a reduction in brittle behavior). The compressive moduli were 6-11 times lower than the flexure moduli. The ratio increased with increasing the fiber content.

#### Scanning electron microscopy

**Fig. 4** shows SEM images of the set Ketac<sup>TM</sup> Fil Plus (A), fiber modified formulations (B-E) as well as the powder (F) and flax fibers (G). The surface morphology of GIC and flax fiber modified GIC exhibited a high degree of integrity and smooth surface. Accordingly, addition of flax fibers produced no difference in the morphology of the set materials. The flax fibers become only visible on the surface when presented in high content (25 weight %) and these fibers were completely coated with the GIC matrix. The cracks seen on samples could be due to dehydration during sample preparation for SEM<sup>[25]</sup>. The powder particles had an angular shape; they varied in sizes from 2 to 30 micron in size. Short flax fibers ( $675 \pm 255 \mu\text{m}$  length and  $\sim 10 \mu\text{m}$  diameters).

## Discussion

Despite the excellent adhesive properties of glass ionomer cement, its weak mechanical properties limits its application in stress-bearing areas. The present study aimed to modify a commercially available conventional

glass ionomer cement with naturally occurring flax fibers to produce new formulations with enhanced mechanical properties while maintaining the same setting reaction kinetics of the conventional cement. The effect of flax fiber incorporation on setting, compressive and flexure properties of Ketac<sup>TM</sup> Fil Plus glass ionomer restorative material has been investigated throughout this study. The morphological character of the produced formulations were also obtained.

After mixing, the acid-base reaction between the weak polyacrylic acid and the finely powdered calcium fluoroaluminosilicate glass commences. This reaction was observed from the absorbance changes seen in FTIR spectra as a gradual reduction in the intensity of polyacrylic acid peak at  $1,707 \text{ cm}^{-1}$ . The intensity of COO salt peaks increased with time. Young observed the presence of symmetric asymmetric COO stretch bands of polyacrylate salts with mono or divalent counterions at  $1,420$  and  $1,540 \text{ cm}^{-1}$  but those of trivalent counterions closer to  $1,460$  and  $1,600 \text{ cm}^{-1}$ <sup>[24]</sup>. Nicholson *et al.*, however, claimed that the position of these peak could vary with the physical environment in which the acid-base reaction occurs<sup>[26]</sup>. The addition of flax fibers had no significant effect on the setting reaction/kinetics of Ketac<sup>TM</sup> Fil Plus. Accordingly, there was not enough evidence to reject the first null hypothesis.

As seen from the stress-strain behavior, the Ketac Fil Plus failed under compression at the elastic limit i.e., the peak stress at the end of the linear elastic region. This could occur due to crack formation within GIC matrix. Once the crack started, an immediate catastrophic failure occurred as expected for brittle materials as GIC. With fiber modified formulations, 5FFRGIC and 25FFRGIC in particular. However, the failure happened at higher stress and at the end of the plastic portion of the stress-strain curve. This plastic region may represent a crack propagation stage which was absent from GIC i.e., application of stresses higher than the elastic limit of 5FFRGIC and 25FFRGIC resulted in a progressive crack propagation before failure. In case of fiber modified formulations, the crack initiation would be expected at the fibers-GIC matrix interface, the weakest area in these formulations. The progression of crack through the plain GIC matrix is expected to be fast; the presence of short, discontinuous flexible flax fibers, however, could absorb some of the stresses or energy required for cracks propagation. The flax fibers could then redistribute these stresses or energy to the nearby GIC matrix, thus suppressing the strain localization. This could explain the presence of ductile, plastic behavior of fiber modified formulation i.e., absence of a catastrophic failure characteristic to the plain GIC

matrix. The large area under the stress-strain curve, seen with fiber modified formulations, indicated a significant increase of the work-of-fracture compared to the brittle characteristics of Ketac<sup>TM</sup> Fil Plus. The crack propagation in GIC matrix resulted in a crack pattern that divided the material into a number of portions that can break off<sup>[27]</sup>.

The compressive strength indicates the resistance of a material to the masticatory forces. The mean values obtained for the compressive ( $148 \pm 12$  MPa) and flexure ( $20 \pm 2$  MPa) strength of Ketac<sup>TM</sup> Fil Plus in the present study are similar to those reported by Molina *et al.*<sup>[28]</sup>. The ratio of compressive to flexure strength is 7.5; this is also in agreement with those reported by Molina *et al.*<sup>[28]</sup>. Generally, Ketac<sup>TM</sup> Fil Plus is stiff, strong and brittle (i.e., no yielding before failure) while the fiber modified Ketac<sup>TM</sup> Fil formulations are resilient, strong and ductile. The reduction seen in compressive modulus with addition of flax fibers was also observed in our previous work using amplitude modulated force microscopy mode (AMFM, 3D-Bio, Research Asylum), which enable continues mapping of surface mechanical properties and recording the force-deflection curve under indentation. The results obtained in the current study, fall in the same range obtained for modulus using AMFM. As also indicated from AMFM, the set GIC represents a single phase material while the fiber modified formulations are bi-phasic materials. Lack of chemical bonding between the flax fibers and the GIC matrix could account for the non-significant increase in the compressive strength particularly at low filler content. Furthermore, absence of chemical bonding between the flax fibers and GIC matrix will produce a weak interface. Surface modification e.g., silane treatment, of the flax fibers will therefore be considered in our future work.

Similar findings were also seen from the three-point bending test. The flexure strength increased but the modulus decreased with addition of flax fibers. The resilience increased linearly with increasing the flax fiber content. This may account for the reduction seen in modulus with increasing the fiber content. The compressive and flexure strength of flax-fiber modified formulations are higher than those obtained by Lohbauer *et al.*, who used reactive glass fibers for reinforcing GIC. According to the results of this study, the second null hypothesis was rejected since there are statistically significant statistical differences in the compressive and flexure properties of Ketac<sup>TM</sup> Fil Plus and flax fiber modified Ketac<sup>TM</sup> Fil Plus.

In conclusion, short flax fibers with  $675 \pm 255$   $\mu\text{m}$  length and  $\sim 10$   $\mu\text{m}$  diameter were used for the reinforcement of glass ionomer to be employed as a

dental restorative material with improved mechanical properties. The addition of flax fibers up to 25 weight % had no significant effect on the acid-base reaction of glass ionomer cement. The acid neutralization was observed by the loss of intense C=O polyacid peak, identified mainly at  $1707\text{ cm}^{-1}$  and concomitant appearance of broad symmetric and asymmetric COO polyacrylate salt peaks. The intensity of COO polyacrylate salt peaks increased with time. Incorporation of flax fibers showed a significant increase in the compressive and yield strength only at high weight %, 25FFRGIC. Low fiber content significantly improved compressive strength obtained by silane treatment of flax fibers. Incorporation of flax fibers  $\geq 1$  wt%, however, significantly increased flexure strength, elasticity and resilience and changed the material's behavior from brittle to ductile. Addition of fibers produced no morphological changes in the set materials. The improvement obtained in the mechanical properties with incorporation of flax fibers could extend the application of GIC to stress-bearing areas.

## References

- [1] Hamouda IM. Effects of various beverages on hardness, roughness, and solubility of esthetic restorative materials[J]. *J Esthet Restor Dent*, 2011, 23(5): 315–322.
- [2] Elsaka SE, Hamouda IM, Swain MV. Titanium dioxide nanoparticles addition to a conventional glass-ionomer restorative: influence on physical and antibacterial properties[J]. *J Dent*, 2011, 39(9): 589–598.
- [3] Crisp S, Pringuer MA, Wardleworth D, et al. Reactions in glass ionomer cements: II. An infrared spectroscopic study[J]. *J Dent Res*, 1974, 53(6): 1414–1419.
- [4] Mendonça AAM, Oliveira CF, Hebling J, et al. Influence of thicknesses of smear layer on the transdental cytotoxicity and bond strength of a resin-modified glass-ionomer cement[J]. *Braz Dent J*, 2012, 23(4): 379–386.
- [5] Selimović-Dragaš M, Huseinbegović A, Kobašlija S, et al. A comparison of the in vitro cytotoxicity of conventional and resin modified glass ionomer cements[J]. *Bosn J Basic Med Sci*, 2012, 12(4): 273–278.
- [6] Geurtzen W, Spahl W, Leyhausen G. Residual monomer/additive release and variability in cytotoxicity of light-curing glass-ionomer cements and compomers[J]. *J Dent Res*, 1998, 77(12): 2012–2019.
- [7] Hattab FN, Amin WM. Fluoride release from glass ionomer restorative materials and the effects of surface coating[J]. *Biomaterials*, 2001, 22(12): 1449–1458.
- [8] Bertolini MJ, Palma-Dibb RG, Zaghet MA, et al. Evaluation of glass ionomer cements properties obtained from niobium silicate glasses prepared by chemical process[J]. *J Non-Cryst Solids*, 2005, 351(6-7): 466–471.

- [9] Bertolini MJ, Zaghet MA, Gimenes R, et al. Determination of the properties of an experimental glass polyalkenoate cement prepared from niobium silicate powder containing fluoride[J]. *Dent Mater*, 2008, 24(1): 124–128.
- [10] Bertolini MJ, Zaghet MA, Gimenes R, et al. Preparation of new glass systems by the polymeric precursor method for dental applications[J]. *J Non-Cryst Solids*, 2004, 344(3): 170–175.
- [11] Boaventura JM, Bertolini MJ, Padovani GC, et al. Tissue response to experimental dental cements prepared from a modified power glass composition[J]. *Dent Mater J*, 2012, 31 (4): 583–592.
- [12] El-Negoly SAEFA, El-Fallal AA, El-Sherbiny IM. A new modification for improving shear bond strength and other mechanical properties of conventional glass-ionomer restorative materials[J]. *J Adhes Dent*, 2014, 16(1): 41–47.
- [13] Stamboulis A, Baillie CA, Garkhail SK, et al. Environmental durability of flax fibres and their composites based on polypropylene matrix[J]. *Appl Compos Mater*, 2000, 7(5/6): 273–294.
- [14] Kromer KH. Physical properties of flax fibre for non-textile-use [J]. *Res Agr Eng*, 2009, 55(2): 52–61.
- [15] Czemplik M, Kulma A, Bazela K, et al. The biomedical potential of genetically modified flax seeds overexpressing the glucosyltransferase gene[J]. *BMC Complement Altern Med*, 2012, 12(1): 251.
- [16] Hu C, Yuan YV, Kitts DD. Antioxidant activities of the flaxseed lignan secoisolariciresinol diglucoside, its aglycone secoisolariciresinol and the mammalian lignans enterodiol and enterolactone in vitro[J]. *Food Chem Toxicol*, 2007, 45(11): 2219–2227.
- [17] Adamyan AA. Current approaches to the development and clinical use of effective dressings, sutures, and polymer implants[J]. *Proceedings of the International Conference 2006*([in Russian], Institut Khirugii im. A. V. Vishevskogo, RAMN, Moscow):19–20.
- [18] Moryganov AP, Galashina VN, Dymnikova NS, et al. Modification of flax fibres: from research to realization[J]. *Fibre Chem*, 2008, 40(3): 234–240.
- [19] Shehata MM, Mona HAD, El-Hariri DM. Compressive strength, solubility and micro-leakage of flax fibers reinforced zinc oxide eugenol dental cement material[J]. *Flax and other bast plants Symp, September 30 and October 1, INF Poznan Poland 1997*.
- [20] Abou Neel EA, Salih V, Revell PA, et al. Brushite and Self-Healing Flexible Polymer-Modified Brushite Bone Adhesives for Fibular Osteotomy Repair[J]. *Adv Eng Mater*, 2014, 16(2): 218–230.
- [21] Young AM, Rafeeka SA, Howlett JA. FTIR investigation of monomer polymerisation and polyacid neutralisation kinetics and mechanisms in various aesthetic dental restorative materials [J]. *Biomaterials*, 2004, 25(5): 823–833.
- [22] Wróbel-Kwiatkowska M, Czemplik M, Kulma A, et al. New biocomposites based on bioplastic flax fibers and biodegradable polymers[J]. *Biotechnol Prog*, 2012, 28(5): 1336–1346.
- [23] Wróbel-Kwiatkowska M, Zuk M, Szopa J, et al. Poly-3-hydroxy butyric acid interaction with the transgenic flax fibers: FT-IR and Raman spectra of the composite extracted from a GM flax[J]. *Spectrochim Acta A Mol Biomol Spectrosc*, 2009, 73(2): 286–294.
- [24] Young AM. FTIR investigation of polymerisation and polyacid neutralisation kinetics in resin-modified glass-ionomer dental cements[J]. *Biomaterials*, 2002, 23(15): 3289–3295.
- [25] Sidhu SK, Pilecki P, Sherriff M, et al. Crack closure on rehydration of glass-ionomer materials[J]. *Eur J Oral Sci*, 2004, 112(5): 465–469.
- [26] Nicholson JW, Brookman PJ, Lacy OM, et al. Fourier transform infrared spectroscopic study of the role of tartaric acid in glass-ionomer dental cements[J]. *J Dent Res*, 1988, 67 (12): 1451–1454.
- [27] Lima PRL, Toledo Filho RD, Melo Filho JA. Compressive stress-strain behaviour of cement mortar-composites reinforced with short sisal fibre[J]. *Mater Res*, 2014, 17(1): 38–46.
- [28] Molina GF, Cabral RJ, Mazzola I, et al. Mechanical performance of encapsulated restorative glass-ionomer cements for use with Atraumatic Restorative Treatment (ART)[J]. *J Appl Oral Sci*, 2013, 21(3): 243–249.