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► **To cite this version:**

Aurélien Mazzoni, Gérard Bernhart, Thierry Cutard. Ceramic fibre reinforced refractory castables for very high temperature superplastic forming. *Materials Science and Engineering Technology / Materialwissenschaft und Werkstofftechnik*, 2012, 43 (9, SI), pp.758-762. 10.1002/mawe.201200032 . hal-01687324

**HAL Id: hal-01687324**

**<https://hal.science/hal-01687324>**

Submitted on 6 Nov 2018

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# Ceramic fibre reinforced refractory castables for very high temperature superplastic forming

## Keramikfaser verstärkte hochschmelzende Gießmassen für superplastisches Umformen bei sehr hohen Temperaturen

A. Mazzoni<sup>1</sup>, G. Bernhart<sup>1</sup>, T. Cutard<sup>1</sup>

In the frame of the development of very high temperature die material solutions, two grades of alumina based ceramic fibres with a diameter close to 12  $\mu\text{m}$  have been investigated as reinforcement solutions for an andalusite refractory castable. High temperature mechanical tests were performed up to 1200 °C in four-point bending conditions: rupture tests at high temperature and anisothermal creep tests. After testing, microstructural and rupture surface were observed in order to get information on deformation mechanisms and rupture processes. Results show that significant mechanical improvement were obtained up to 1100 °C when compared to non reinforced refractory castables.

**Keywords:** Refractory castable / superplastic forming / ceramic dies / ceramic fibre reinforcement / microstructure

**Schlüsselwörter:** Hochschmelzende Gießmassen / superplastisches Umformen / Keramikgesenk / Keramikfaser Verstärkung / Mikrostruktur

### 1 Introduction

Since several years, Fibre Reinforced Refractory Castables (FRRC) have been developed as an emerging material solution to manufacture tools in order to propose a low cost solution for Ti alloy sheet Superplastic Forming [1, 2]. Non-reinforced refractory castables keep good mechanical properties until 1000 °C and are characterized by a good thermal shock resistance because of their low thermal expansion coefficient and low Young's modulus. Unfortunately, their mechanical behaviour remains quasi-brittle. It was demonstrated that the reinforcement with stainless steel fibres gives some ductility to the resulting FRRC and avoids a catastrophic failure after crack initiation.

Unfortunately when addressing higher temperatures, up to 1100 °C as those that may be required for TiAl alloy, Superpalloy or duplex steel sheet superplastic forming, classical metallic reinforcement solutions are no more possible. In order to overcome this limit, alumina based ceramic fibre reinforcement solutions were investigated. This change does not only introduce a composition difference but also a major morphological evolution; as a matter of fact ceramic fibres have a diameter close to 12  $\mu\text{m}$  compared with the previous 500  $\mu\text{m}$  for the metallic ones and aggregates sizes that are close to 3 mm.

After the description of fibre and castable matrix compositions, the processing route is shortly described and the high temperature mechanical properties under four-point bending conditions are reported. The non-reinforced castable is compared with two alumina fibre reinforcing solutions over the range from 700 °C to 1200 °C. Interrupted and post rupture microstructural investigations have been performed in order to get an understanding of the internal micro-mechanical damage and fibre-matrix load transfer mechanisms.

### 2 Materials and experiments

#### 2.1 Materials

A commercial grade of an andalusite based refractory concrete (And-NF: Andalusite No Fibres) is considered in this work [3–5]. It is made of aggregates with a 5 mm maximum size, of silica fume, of alpha-alumina and of a low content of a calcium aluminate cement. The main properties of the two alumina-rich fibres investigated as reinforcing solutions (Nextel 720<sup>®</sup> and Nextel 610<sup>®</sup>) are listed in *Table 1*. The as delivered long mineral fibre tows were first cut to produce 12 mm short fibres and then mixed in a planetary mixer with the castable raw material and with 5 wt% water addition to produce the test samples.

After casting in a wood mould, mould and samples are then immediately wrapped in plastic. They are kept and dried at room temperature during 24 h and then extracted from the moulds before a 110 °C – 24 h drying step. Results on two grades of fibre reinforced andalusite concretes are reported in this paper and compared with the non reinforced one (And-NF):

<sup>1</sup> Université de Toulouse; INSA, UPS, Mines Albi, ISAE; ICA (Institut Clément Ader), Albi cedex, France

**Table 1.** Designation and properties of reinforcing fibres

Parameter	Nextel 720® (N7)	Nextel 610® (N6)
Diameter [µm]	12	12
Length [mm]	12	12
Main constituents [wt%]	85% Al <sub>2</sub> O <sub>3</sub> 15% SiO <sub>2</sub>	> 99% Al <sub>2</sub> O <sub>3</sub>
Young's modulus [GPa]	260	380
Tensile strength [MPa]	2100	3100
Maximum use temperature [°C]	1800	2000
Expansion coefficient [K <sup>-1</sup> ]	6.0 · 10 <sup>-5</sup>	8.0 · 10 <sup>-5</sup>

- grade And-N7 : And-NF reinforced with 2 vol.% of Nextel 720® short fibres
- grade And-N6 : And-NF reinforced with 2 vol.% Nextel 610® short fibres

Before performing microstructural or mechanical tests, a firing cycle is applied with a heating and cooling ramp of 60 °C/min including an isothermal dwell of 5 hours at maximum firing temperature. Before high temperature mechanical testing samples are heated with the same heating rate of 60 °C/min and subjected to a half hour dwell.

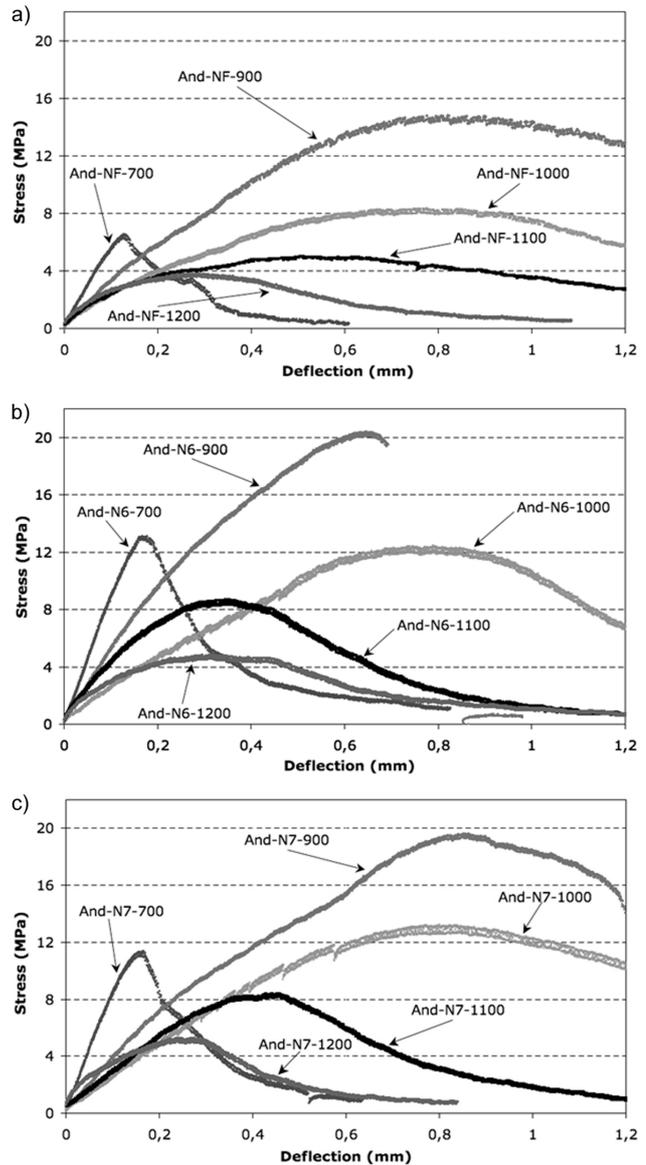
## 2.2 Experiment procedures

The four-point bending tests were carried out on a MTS810 servo-hydraulic universal testing machine and more details can be found in previous papers [6–8]. The samples, with a size of 150 × 25 × 25 mm<sup>3</sup>, were loaded at a constant crosshead speed of 0.2 mm/min. Applied load and beam deflection were recorded during the tests. Monotonic tests are performed at high temperature between 700 °C and 1200 °C using a resistive heating furnace and alumina testing fixtures. Fracture surfaces were observed at room temperature on a FEI-XL30-ESEM-FEG environmental scanning electron microscope.

## 3 High temperature mechanical behaviour

An increase of the non-linear domain of the behaviour curves is observed combined with an important increase of the strain capacity. *Figure 1* shows that reinforcing the refractory concrete with alumina fibres modifies its behaviour over the whole temperature range. Moreover, this figure allows to notice that the mechanical characteristics of the refractory concrete is hardly modify with the testing temperature.

Figures 1a, 1b and 1c show bending curves for the unreinforced And-NF refractory concrete and for Nextel 610® reinforced (And-N6) and Nextel 720® reinforced (And-N7) grades. Tests have been performed at 700 °C, 900 °C, 1000 °C, 1100 °C and 1200 °C. At 700 °C, the behaviour is quite similar for the three materials. After a linear elastic part, when the stress level increases, the behaviour moves to a non-linear one due to damage processes by microcracking. At higher stress levels, damage localizes and one or several macrocracks appear. After the stress peak, the behaviour curve exhibits an extended softening part. When comparing to stainless steel reinforced castables, fibre



**Fig. 1.** Four-point bending behaviour curves of concrete fired and tested at high temperature between 700 °C et 1200 °C: a) unreinforced (And-NF), b) Nextel 610® (And-N6) and c) Nextel 720® (And-N7).

bridging is not so efficient and the post peak behaviour shows a rapid stress decrease, that is nevertheless not brittle [1, 2]. At temperatures higher than 900 °C, general behaviour moves to a typical elasto-visco-plastic behaviour.

Reinforcing the And-NF concrete with Nextel fibres leads essentially to large modifications of the pre-peak behaviour. They mainly deal with an increase of the width of the pre-peak linear domain and with an increase of the stress peak and deflection peak levels. Nextel fibres can also be considered as delaying diffuse damage phenomena and to limit the visco-plastic behaviour of the concrete. Because of the high alumina content of Nextel fibres, the temperature range in which such phenomena can be

**Table 2.** Peak stress relative variation at high temperature (four-point bending test)

Testing temperature	700 °C	900 °C	1000 °C	1100 °C	1200 °C
And-N7	86%	79%	96%	52%	31%
And-N6	117%	65%	73%	51%	41%

Unreinforced concrete is the reference

**Table 3.** Apparent elastic modulus relative variation at high temperatures (four-point bending test)

Testing temperature	700 °C	900 °C	1000 °C	1100 °C	1200 °C
And-N7	44%	16%	110%	12%	49%
And-N6	36%	22%	85%	49%	49%

Unreinforced concrete is the reference

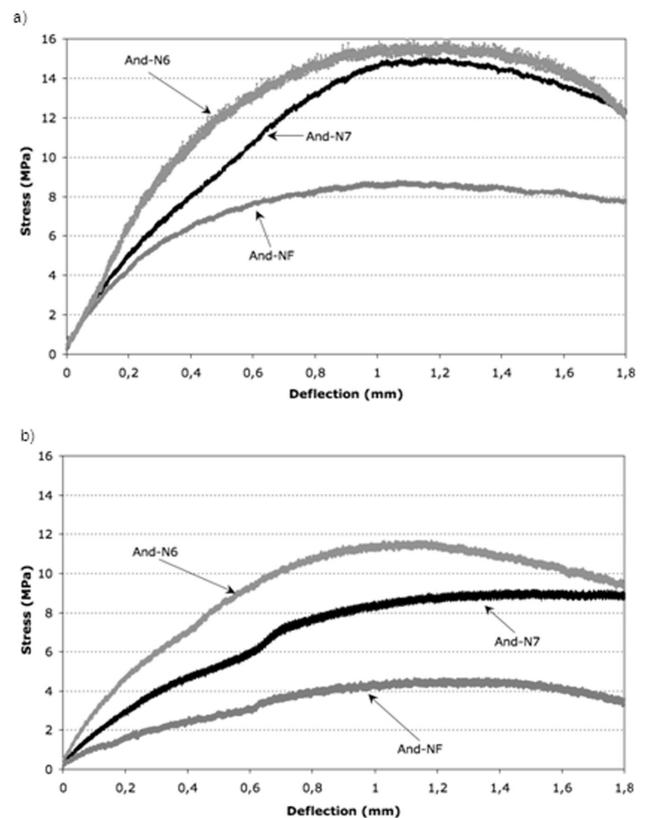
obtained is largely increased. No benefit effect is nevertheless observed in the post-peak domain

Table 2 and 3 gather respectively the peak stress and apparent elastic modulus relative variations of the reinforced And-NF castable and allow the quantification of the reinforcing effects. It can be seen that the addition of alumina-rich fibres increases the mechanical characteristics over the whole temperature range. The gain on the peak stress values varies between 9% and 96% for the And-N7 grade and between 41% and 117% for the And-N6 grade. For the apparent elastic modulus the gain varies between 12% and 110% for And-N7 grade and between 22% and 85% for And-N6 grade.

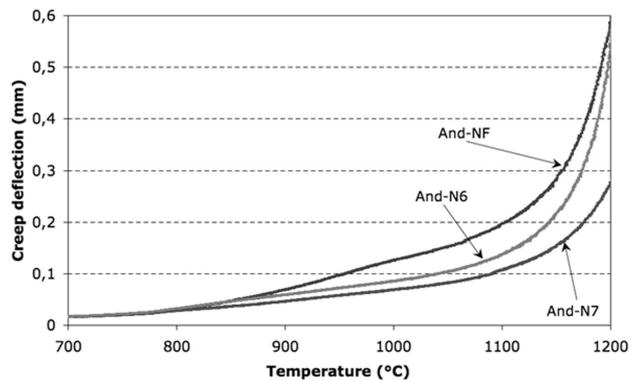
These first results of high temperature behaviour indicate that alumina-rich fibres have the capacity to delay the diffuse damage phenomena; fibres also modify the mechanisms that drive the visco-plastic concrete behaviour.

In order to study the visco-plasticity effect of the concrete, bending test were performed at 900 °C in modifying the crosshead speed from 0.2 mm/min to 0.02 mm/min, Figure 2. As it can be seen, dividing the displacement rate by a factor of 10 reduces the peak stress and the apparent modulus of the concrete without permitting a higher deformation capacity. For a given deflection value, a decrease of the strain rate causes a decrease of the stress values that has been related to the appearance of a liquid phase in the cementitious matrix [3]. The lower displacement rate allows a higher stress relaxation of the concrete. In Figure 2, the benefits of the alumina-rich fibre reinforcement can be observed, the higher the Al<sub>2</sub>O<sub>3</sub> content the higher the load capacity: for the And-N6 grade tested at 0.2 mm/min, the stress gain is 80% for grade whereas for a rate of 0.02 mm/min, the gain is as high as 150%.

Creep behaviour was also investigated under non-isothermal creep conditions: four-point bending samples were submitted to a constant 2 MPa stress level whereas temperature was raised from 20 °C to 1200 °C, Figure 3. Under such testing conditions, creep starts to occur between 750 °C and 800 °C whatever the grade. Results confirm that reinforcing the And-NF concrete with Nextel fibres increases its high temperature mechanical behaviour. Indeed, in all the temperature range, the measured



**Fig. 2.** Crosshead speed effect on four-point bending behaviour at 900 °C of the three grades: (a) 0.2 mm/min and (b) 0.02 mm/min.

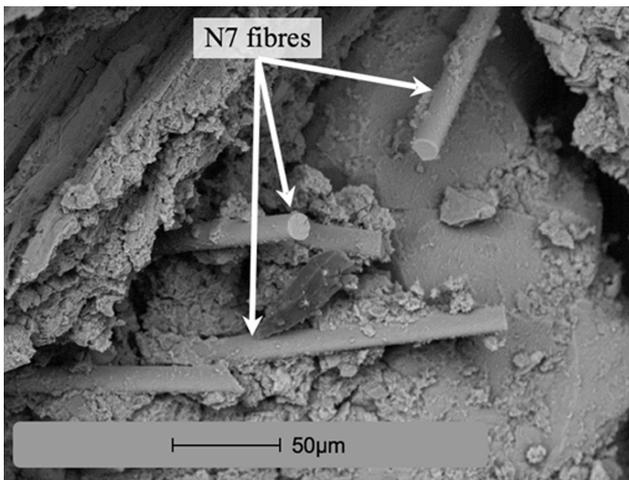


**Fig. 3.** And-NF, And-N6 and And-N7 grades anisothermal creep behaviour under 2 MPa (four-point bending) until 1200 °C.

deflection levels are really lower for the reinforced grade up to 1100 °C, the And-N7 grade showing the better performances. After 1100 °C, a rapid creep deflection was observed for the And-N6 grade whereas And-N7 grade retains his creep resistance.

#### 4 High temperature microstructural evolution

In order to identify the phenomena involved in the high temperature range, SEM observations were performed on samples after testing.



**Fig. 4.** Fracture surface of an And-N7 sample after a bending test at 900 °C (SEM micrograph).

For the 900 °C tested samples, *Figure 4* shows a fracture surface with fibres that have been pulled out of the concrete. This indicates that debonding and sliding mechanisms act at the fibre/concrete interfaces. Such mechanisms must be considered as having a major role on the high temperature behaviour of the reinforced grades.

Previous studies have shown the microstructural evolution of the matrix concrete at high temperature (starting at 900 °C) and in particular the apparition of liquid phases in the matrix [4, 5]. In order to observe the interaction between liquid phase and fibres, high magnification observations were completed on samples ruptured in the range 700 °C–1200 °C. *Figure 5* shows clearly the importance of the matrix microstructural evolution from a still cementitious matrix at 700 °C towards a matrix with high liquid phases at higher temperatures. These images show that Nextel fibres reinforcement process involves also liquid phases at fibre/concrete interface.

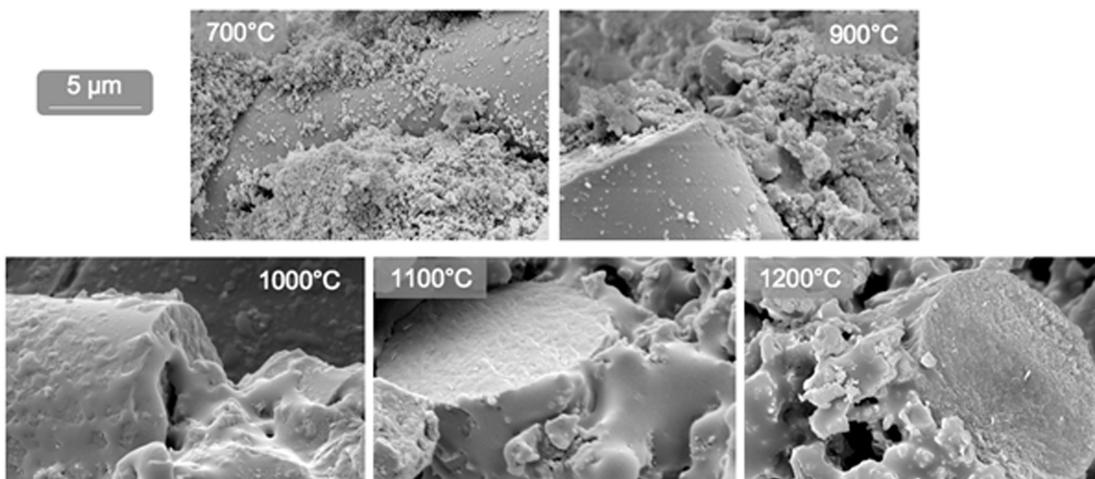
During high temperature testing, the fibres modify the natural network of microcracks of the concrete. This can be seen on *Fig-*

*ure 6* showing microstructural observations on four-point bending test samples submitted to interrupted tests at 900 °C (test stop took place before maximum stress peak). For both grades (And-N7 (a) and And-N6 (b)) numerous microcrack deviations, crack branching and bridging and fibre/matrix debonding configurations are visible (white arrows). As a consequence the presence of fibres slows crack propagation and opening. Debonding phenomena take extensively place both at matrix/aggregate interfaces and at matrix/fibre interfaces. The combination of all these phenomena is beneficial and partly explains the overall non-linear behaviour that is a mix between elastic-damageable and visco contributions. However, at this scale, the specific effect related to the presence of liquid phase is difficult to observe especially after return to room temperature for observations.

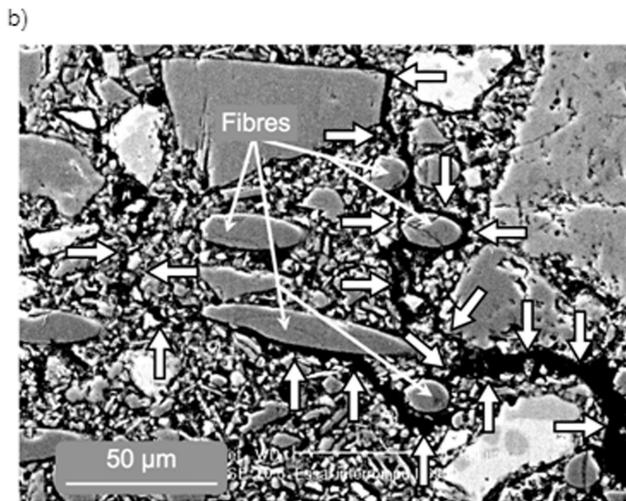
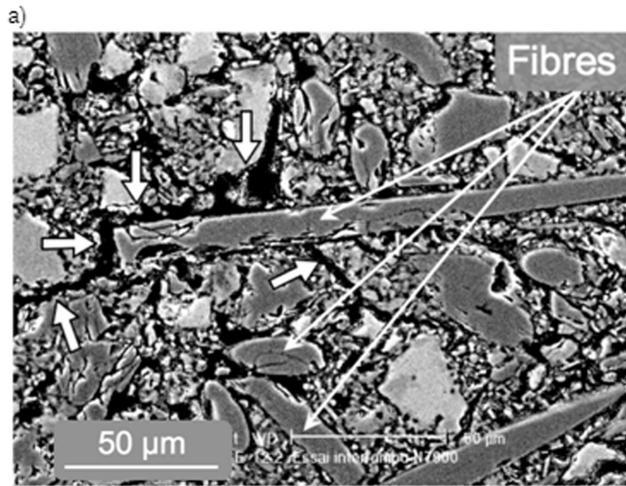
*Figure 7* highlights the role of fibres in the visco-plastic behaviour of the concrete at 1200 °C and before a high level of damage. At this temperature, liquid phase amount of matrix is important and some vitreous phases could be seen. The observations partly help to explain the macroscopic viscoplastic deformation as a sliding mechanism between solid particles immersed in a large amount of liquid matrix phase. Such mechanisms are consistent with the increase of the strain capacity of the concrete and the stress accommodation phenomena in this high temperature testing domain. As a consequence the reinforced concrete becomes sensitive to strain rate. Moreover, many vitreous bridges are observed at fibres/matrix interfaces or between fibres. From this it can be considered that fibres can slow down or stop the relative displacement between the solid constituents of the matrix and modifying the rheology of the matrix at high temperature.

## 5 Conclusion

Ceramic fibre reinforced refractory castable have been studied in order to propose material solutions for die manufacturing for very high temperature Superplastic Forming. Two grades of Alumina based ceramic fibres with a diameter close to 12 μm have been investigated as reinforcement solutions for an andalusite castable refractory and for temperatures higher than 900 °C.

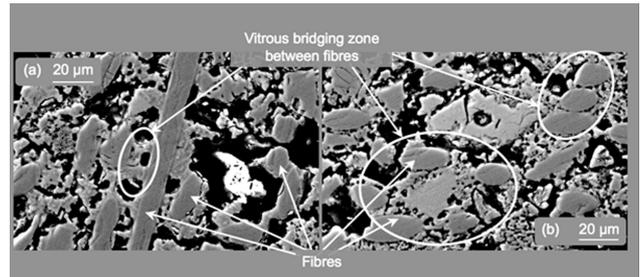


**Fig. 5.** Fracture surfaces of Nextel 720® fibres and surrounding matrix after four-point bending tests in the temperature range 700 °C to 1200 °C.



**Fig. 6.** Observation of diffuse damage developed during a 900 °C four-point bending test (interrupted before the peak stress) in an andalusite based castable reinforced with 2 vol.% of: (a) Nextel720® fibres (b) Nextel610® fibres. White arrows indicate microcracks deviation phenomena to fibres.

High temperature mechanical tests were performed up to 1200 °C in four-point bending conditions followed by after testing microstructural and rupture surface observations. Results show that significant mechanical and creep improvement were obtained up to 1100 °C when compared to non reinforced refrac-



**Fig. 7.** Observation of And-N7 grade at room temperature after an interrupted four-point bending test at 1200 °C.

tory castables. These material solutions are now ready for industrial trial. At high temperatures, these materials exhibit a macroscopical visco-plastic behaviour, that has been related to microcracking branching and fibre/matrix debonding phenomena as well as numerous liquid phase appearance.

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