Advanced Creep Testing of Refractories Providing New Insight into Material Behaviour

S. Jin, H. Harmuth, D. Gruber, S. Schachner, P. Meunier, R. Techer

Feasible high temperature testing approaches for ordinary ceramic refractory materials are of importance to understand material behaviour in service and further facilitate the material development. Aside from the wedge splitting test and modified shear test applied to investigate the tensile and shear failure respectively, an advanced high temperature compressive creep testing device was developed to characterize the creep behaviour of ordinary ceramic refractory materials. The application of rather low preload during heating up procedure and creep measurement on the cylindrical surface of specimens allows for determining the creep onset and deformation accurately. The study of an ultralow cement alumina castable demonstrated that the creep of the virgin castable below 1500 °C is overruled by sintering, which is sensitive to the preheating conditions of preload, dwell time and temperature. Caution shall be taken on the creep testing conditions for monolithics, and the consideration of service conditions is necessary. The identification of sintering contribution to the deformation during creep testing is also essential to gain advanced understanding on the thermomechanical behaviour of industrial vessel refractory linings.

1 Introduction

The rigorous requirements to build a sustainable society and support the advanced metallurgical technology dramatically challenge the development and application of refractories. Certain possible attracting features of refractories are low CO_2 emission, reduced material consumption, contribution to energy saving and metal quality improvement, etc. As reported by PRE (the European Refractories Producers Federation), refractories experiencing a firing procedure before being put into practice involve substantially higher equivalent tonne amount of CO_2 than those unfired [1]. In the last decades, the production and research activities of monolithic refractories increased evidently [2, 3].

An integrated engineering-targeted development activity of monolithic refractories shall involve the material design with various ingredients, the application of suitable testing approaches especially for elevated temperature purpose and the refractory lining concept optimisation of industrial vessels. The lack of comprehensive considerations might mislead the material development. For instance, the concept, "the stronger, the better", in the fine ceramic field deeply is misused in the development

Shengli Jin, Harald Harmuth, Dietmar Gruber, Stefan Schachner
Chair of Ceramics
Montanuniversitaet Leoben
Austria

Pierre Meunier, Romain Techer
Calderys
Sézanne
France

Corresponding author: *Shengli Jin*
E-mail: Shengli.Jin@unileoben.ac.at

Keywords: creep behaviour, monolithic refractories

Received: 17.01.2018
Accepted: 01.02.2018

The paper received the 3rd Prize of the Poster Award launched by refractories WORLDFORUM at ICR™ 2017 in Aachen/DE

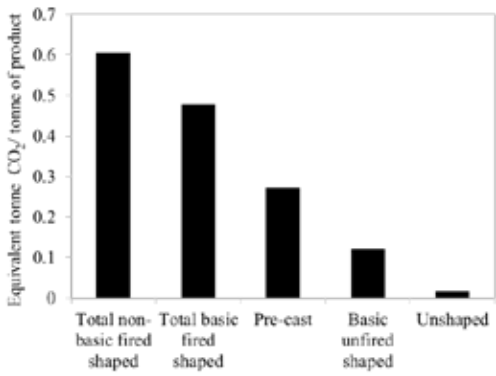

Fig. 1 **Carbon footprint of different refractory product groups [1]**

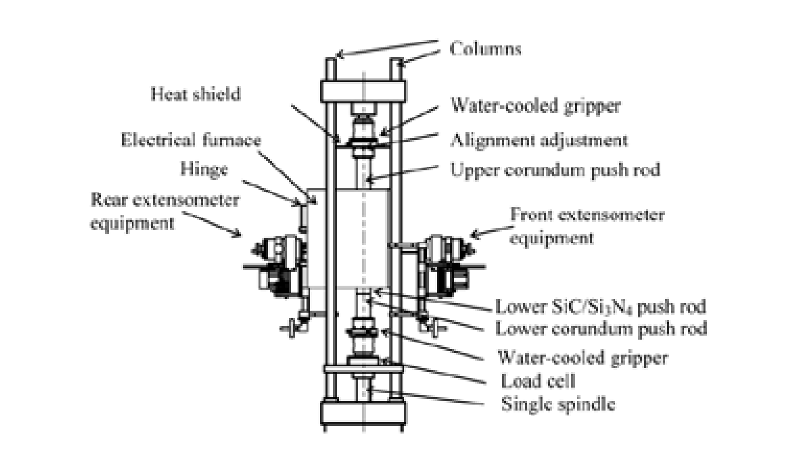

Fig. 2 Schematic diagram of the testing machine by courtesy of MESSPHYSIK/AT

of ordinary ceramic refractory materials that often demonstrate decreased brittle behaviour [4, 5]. The results from wedge splitting tests according to Tschegg proved that a better thermal shock resistance can be achieved by reducing the tensile strength whilst maintaining the specific fracture energy [6]. The fractographic results clearly indicated that a microstructural measure can be taken to reduce the brittleness of ordinary ceramic refractory materials, of which a greater amount of crack propagation along the grain/matrix boundaries shall predominate. Following this study, many researchers shift their attentions from the conventional testing approaches, such as three point bending test, quenching thermal shock test, into the wedge splitting test [7, 8] or that combined with fractographic observation for the advanced material development [9, 10].

Creep under load at high temperatures is another important issue for refractories, and an appropriate investigation of refractory creep will contribute to the proper material development and application. The standard testing approach for refractories – creep in compression (CIC, EN993-9) – only applies a rather low load limited to 0,2 MPa. Under such a low load, many materials may need extremely long testing time to reach the secondary creep stage. Furthermore, in order to receive information suitable for the later simulation of case studies, it is necessary to apply loads related to service conditions. Besides, with the CIC testing approach, the creep onset is not well defined, and thus the method is not suitable to accurately identify the material law that accounts for creep in the thermomechanical modelling of refractory linings. To overcome these disadvantages of the CIC test, an advanced compressive refractory creep testing device was developed [11]. The present paper applied this testing device for the creep study of an ultralow cement alumina castable and investigated the influences of preheating conditions on the creep determination under constant loads and temperatures.

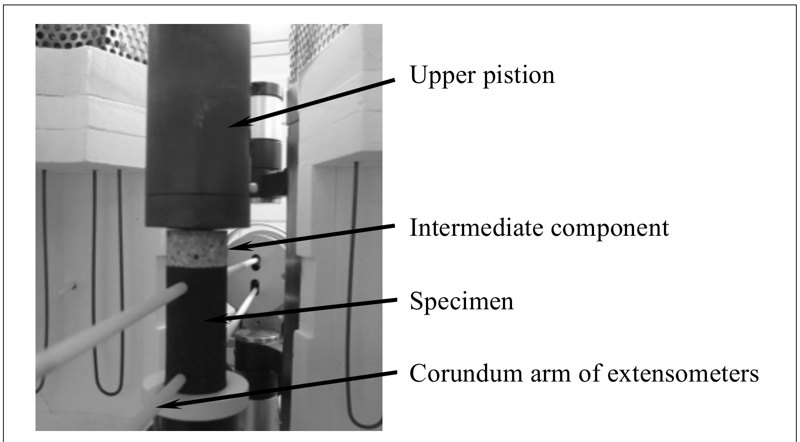

Fig. 3 Setup of the high temperature compressive creep test with furnace opened

2 Advanced compressive refractory creep testing device [11]

Shown in Fig. 2, the whole setup is based on a spindle-driven universal testing machine of sufficient stiffness, and an electrical furnace is inserted. The furnace is composed of two symmetrical parts, held together by means of hinges in the rear and can be closed tightly with two buckles in the front. Two platinum-rhodium thermocouples are used to measure the temperatures, one of which is located near the specimen to monitor its temperature and the other one is placed close to the heating elements to control the furnace temperature. Two pairs of extensometers with corundum arms are placed in the front and the rear of the furnace, respectively. The loading is realized by a single spindle connected to a load cell. Water-cooled grippers are placed at the cold ends of corundum push rods to protect the metal components. To guarantee coincidence of the axis of the lower and upper piston an alignment adjustment is provided. Compared to the standard CIC test, this device possesses several advanced features. Loads up to approx. 20 kN on a specimen can be applied; the height/diameter ratio of 2 allows the deformation measurement excluding the friction effect of the end faces; two pairs of extensometers with corundum arms are used to directly measure the deformation of specimens; creep onset is well defined, by recording the deformation as the loading procedure starts; A cap-like shape of the upper piston end is applied to avoid uneven loading. Both end faces of the cylindrical specimens are prepared by diamond grinding. A less brittle refractory material is placed between the upper piston and specimen, as labelled as intermediate component in Fig. 3.

3 Experimental

An ultralow cement alumina castable was investigated, and its chemical composition and physical properties are shown in Tab. 1. The castable was prepared by adding 3,5 mass-% water and cast into blocks of 100 mm × 100 mm × 100 mm. Compaction was performed by vibration. After hardening in a sealed cabinet at room temperature for 24 h, the cast blocks were cured at 110 °C for another 24 h. Cylindrical specimens (∅35 mm × 70 mm) were drilled from the cured blocks and placed in an oven at 110 °C for 24 h for drying.

In the case of creep testing, the virgin specimen was situated on the lower piston and a certain preload was applied. The alignment of loading and specimen axes was adjusted at room temperature before the heating procedure started. Different preloads (5 N and 100 N), dwell time (with and without dwell) and dwell temperatures were applied before creep measurement started. During the creep testing, a constant load of 3 MPa was applied at 900–1100 °C, and 0,2 MPa at 1200–1400 °C, and 1 MPa at 1500 °C, respectively.

Tab. 1 Typical chemical composition and properties of the alumina castable

Chemical composition [mass-%]	Al_2O_3	93,38
	SiO_2	5,63
	CaO	0,38
	Fe_2O_3	0,14
	Na_2O	0,33
	K_2O	0,14
Bulk density after drying at 110 °C [kg · m^{-3}]		3100
Thermal conductivity at 800 °C [W · m^{-1} · K^{-1}]		3,56
Cold crushing strength after firing at 800 °C [MPa]		130

4 Results

Fig. 4 a–b shows the total strain curves after creep testing at 900 °C and 1000 °C under the preheating conditions of 5 N preload and 3 h dwell/without dwell. The curves received from the tests without dwell were steeper at the beginning of creep testing resulting in higher strains compared to those with 3 h dwell time. The ultimate strain differences after 5 h creep testing between the curves with and without dwell were 6,5 · 10^{-4} for 900 °C, and were 5,1 · 10^{-3} for 1000 °C. At 1100 °C, the difference of ultimate strains with and without dwell under 5 N preload became rather evident and amounted to 2,86 · 10^{-2} (Fig. 4 c). Without dwell, when the preload was 100 N, the ultimate strain was 1,1 · 10^{-2} lower than that with 5 N preload. Moreover, without dwell, the creep strain increased pronouncedly at the beginning of creep testing and flattened after 1 h. In contrast, creep slowly developed when the preheating conditions

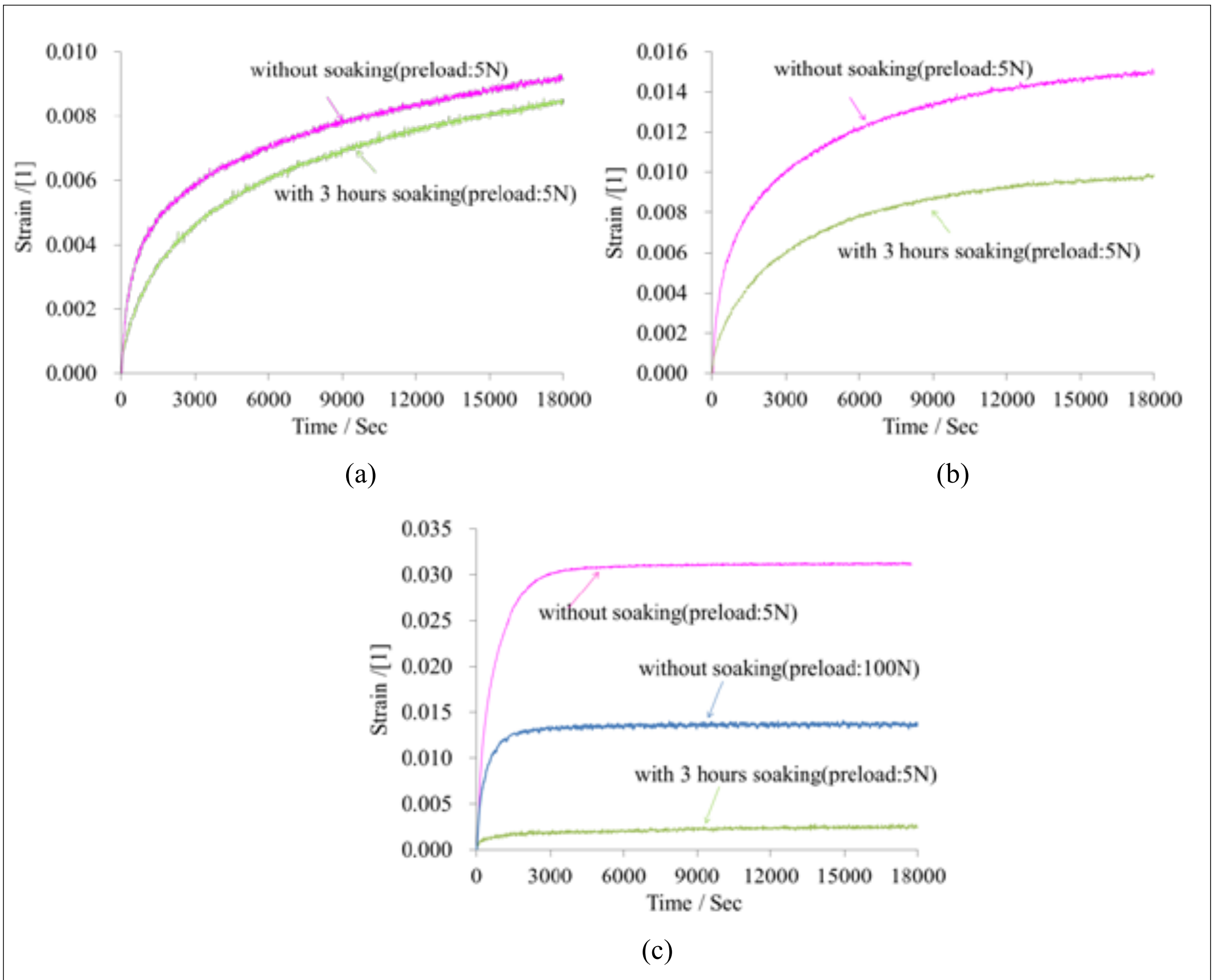

Fig. 4 a–c Creep curves under a constant load of 3 MPa at a) 900 °C, b) 1000 °C, and c) 1100 °C with different preheating conditions

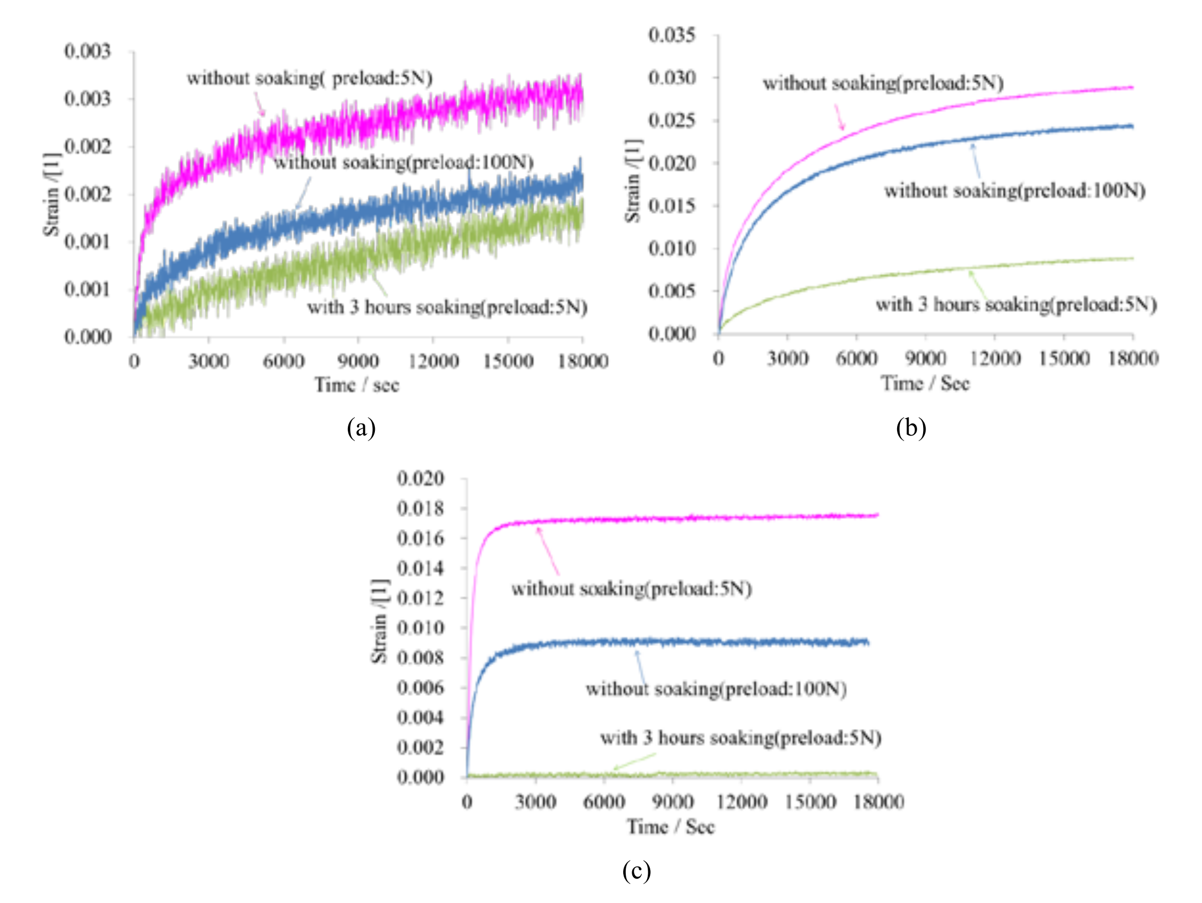

Fig. 5 a–c Creep curves at a constant load of 0,2 MPa at a) 1200 °C, b) 1300 °C, and c) 1400 °C with different preheating conditions

of 5 N and 3 h dwell were applied. It is clear that the impact extent of 3 h dwell was higher than the preload 100 N.

In 1200–1400 °C (Fig. 5 a–c), the creep tests were carried out at the constant load of 0,2 MPa. The figures also show that a higher preload or 3 h dwell contributed to a decrease of total strain. At 1200 °C (Fig. 5 a), the ultimate strains of curves with 100 N and 3 h dwell were 10^{-3} lower than that with 5 N and without dwell. At 1300 °C (Fig. 5 b), the 3 h dwell brought about a significant increase of creep resistance with an ultimate strain difference of $2 \cdot 10^{-2}$ to that with 5 N and without dwell. The preheating conditions of 100 N preload without dwell reduced the creep deformation by $4{,}6 \cdot 10^{-3}$. At 1400 °C (Fig. 5 c), creep curves looked substantially different to those at 1200 and 1300 °C. It is noticeable that after a sharp increase of creep strain in half an hour, the creep curves were nearly flat in the residual 4,5 h when the specimens did not experience a dwell procedure. In contrast, the specimen after 3 h dwell at 1400 °C showed insignificant creep. At 1500 °C (Fig. 6), the creep strain difference of curves with 5 N and 100 N preloads was negligible, and the 3 h dwell procedure accelerated the creep.

The above results exemplarily demonstrate the impact of preheating conditions on the measured creep deformation of a virgin castable. The equilibration processes in the virgin castable occurring at elevated temperatures account for this. The equilibration might bring about diffusion, sintering, shrinkage and formation of new phases. The phase equilibrium calculation applying FToxid oxide database of FactSage 7.0 [12] indicates that the liquid phase occurs around 1100 °C and the amount increases with respect to temperature, and above

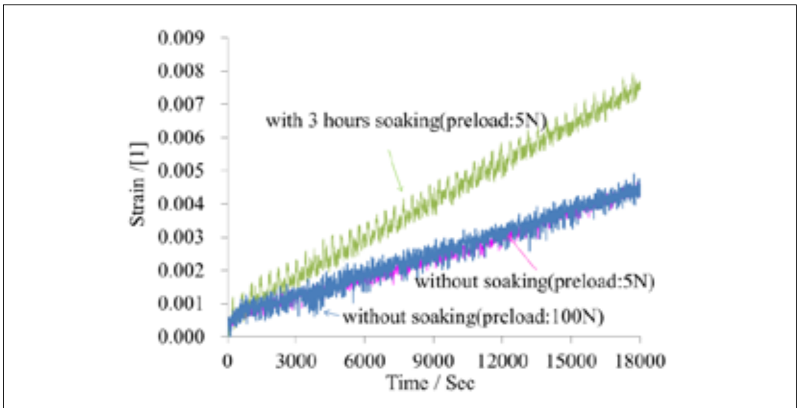

Fig. 6 Creep curves at a constant load of 1 MPa, and 1500 °C with different preheating conditions

1280 °C mullite is formed. The microstructure study showed that the virgin specimen contained aggregates of corundum and quartz. The XRD analysis of tested specimens after 3 h dwell showed that anorthite and cristobalite formed additionally after testing at 1300 °C, whilst only corundum and mullite were identified after testing at 1400 °C. The discrepancy between XRD analysis and FactSage calculation is caused by the equilibrium dynamics. It is assumed that below 1400 °C or at 1400 °C without sufficient dwell duration, the equilibration between silica carriers of quartz and microsilica added to the mixture composition and alumina was not yet achieved. The solid state sintering at low temperatures accompanied by the Ostwald ripening process [13] could also result in shrinkage of the virgin castable. Therefore, a significant deformation was always observed. A long dwell period or high preload promotes the equilibration processes and grain growth accompanied by the Ostwald ripening process, which enhance the creep resistance. The formed liquid phase of great amount at 1500 °C controls the creep of the virgin castable and the role of sintering is undermined.

5 Conclusion

An advanced high temperature compressive testing device was developed to characterize the creep behaviour of ordinary ceramic refractory materials. With this creep device, the contribution of sintering to the observed strain was identified for the monolithic materials exemplarily. In practice, the monolithic materials may experience complex cyclic thermal conditions, for instances, preheating of monolithic linings in a virgin state, thermal shock at the hot face, and various thermal histories of different zones in a monolithic lining. It is necessary to investigate the influences of service related thermal histories on the irreversible deformation of monoliths, and further develop feasible material constitutive models to describe the coexisting sintering and creep, for the sake of elaborate material and lining concept designs.

Acknowledgements

Financial support by the Austrian Federal Government (in particular from Bundesministerium für Verkehr, Innovation und Technologie and Bundesministerium für Wissenschaft, Forschung und Wirtschaft) represented by Österreichische Forschungsförderungsgesellschaft mbH and the Styrian and the Tyrolean Provincial Government, represented by Steirische Wirtschaftsförderungsgesellschaft mbH and Standortagentur Tirol, within the framework of the COMET Funding Programme is gratefully acknowledged (K2 MPPE). Federation for International Refractory Research and Education (FIRE) together with several of its members have been partners of this project.

References

[1] PRE-Product carbon footprint report. European Refractories Producers Federation, Brussels, September 2013, www.pre.eu

[2] Paula da Luz, A.; Braulio, M.A.L.; Pandolfelli, V.C.: Refractory castable engineering. Baden-Baden 2015, 3–8

[3] Semler, C.E.: The world's most important but least known products. Amer. Ceram. Soc. Bull. **93** (2014) [2] 34–39

[4] Semler, C.E.: Review of refractories markets and research – 2016. refractories WORLDFORUM **9** (2017) [2] 30–34

[5] Bradt, R.C.; Harmuth, H.: The fracture resistance of refractories. refractories WORLDFORUM **3** (2011) [4] 129–135

[6] Harmuth, H.; Bradt. R.C.: Investigation of refractory brittleness by fracture mechanical and fractographic methods. Interceram, Refractories Manual (2010) 6–10

[7] Miyaji, D.Y.; et al.: Fracture energy and thermal shock damage resistance of refractory castables containing eutectic aggregates. Ceramics Int. **40** (2014) [9] 15227–15239

[8] Böhm, A.; et al.: Thermal shock and thermomechanical behaviour of carbon-reduced and carbon-free refractories. J. of Ceram. Science and Technol. **7** (2016) [2] 155–164

[9] Zhu, T.; et al.: Fracture behaviour of low carbon MgO–C refractories using the wedge splitting test. J. of the Europ. Ceram. Soc. **37** (2017) [4] 1789–1797

[10] Preux, N.; et al.: Development of a fractography method to investigate the crack path, induced during wedge splitting test of alumina-spinel castables. Proc. of 60th Int. Coll. on Refractories, 2017, Aachen, Germany, 108–113

[11] Jin, S.; Harmuth, H.; Gruber, D.: Compressive creep testing of refractories at elevated loads – Device, material law and evaluation techniques. J. of the Europ. Ceram. Soc. **34** (2014) [5] 4037–4042

[12] FactSage 7.0. http://www.factsage.com

[13] De Jonghe, L.C.; Rahaman, M.N.: Handbook of Ceramics, S. Somiya, Ed., Chapter 4.1, Sintering of ceramics. Amsterdam 2003