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Multi-physical analysis for refractory's design Case of slag impregnated refractory

E. Blond^a, N. Schmitt^b, J. Poirier^c

^aLMSP (UMR 8106 CNRS-ENSAM-Université d'Orléans), ERT « Design of High temperature ceramic structures », Polytech'Orléans, 8 rue Leonard de Vinci, F-45072 Orléans Cedex, France

^bLMT Cachan (UMR 8535 CNRS / ENS de Cachan / Université Paris 6), 61 avenue du Président Wilson 94 235 Cachan (France)

^cCRMHT (UPR 4212 CNRS), ERT « Design of High temperature ceramic structures »

Abstract

Today a better knowledge of degradations due to multiphysical phenomena is possible thanks crossanalyses permitting the development of new models. First, we discuss on the best way to account for couplings between mechanical, thermal, chemical and transport phenomena in refractory structures. Then, this approach is used to understand the causes of degradation in bauxite-based working lining used in steel ladles. It is shown that a new cause could explain the spalls observed in the impregnated zone of bauxite refractory.

Keywords: Multi-physics coupling, Refractory, Slag impregnation, Thermal spalling

Introduction

Thanks specific studies of corrosion¹, phase change² or thermomechanic³ problems occurring in fire industrial processes, made by specialists in different fields, important progresses of the life-time of refractories have been obtained. Although specialists acknowledge that pooling their resources could help to explore new ways to improve refractories, cross-studies are less common. In particular, thanks to the development of new experimental and numerical tools in different scientific fields^{4,5,6} a better knowledge of the causes of degradations due to the combination of multiphysical phenomena is now possible. We show how a multiphysical reasoning was developed for the study of degradation of slag impregnated bauxite-based refractory in steel ladle.

Cross-analyses associating both ceramic and mechanical approaches provide new tools for the modelling and understanding of the coupling effects^{7,8}. However, one difficulty is to estimate the weight of each mechanism responsible of these degradations. This presentation aims at demonstrating that general coupling approaches enable one to propose new ideas and to test their relevance.

The beginning is devoted to a discussion of a multiphysics coupling framework that allows, from a conceptual point of view, to consider all possible causes of degradations involving thermomechanic and / or chemistry with the aim of developing a multiphysical model. As it is not reasonable to account for all degradation mechanisms in only one model, the whole problem is decomposed into several simpler subproblems to bypass this difficulty. In the continuation, the proposed approach is applied on the case of the spalling of bauxite based refractories impregnated by slag in steel ladle. The chemical gradient produced by the slag impregnation is studied. The presence of a liquid in the porosity is underlined. Then, the fluctuation of the liquid pressure with thermal loading is studied. The non linear mechanical behaviour of bauxite at high temperature is discussed. Finally, the spalling risk due to the interstitial pressure of liquid oxides in refractory is quantified and the effect of permeability is discussed.

General coupling scheme

The common classification of the loads of refractory structures encourages the development of approaches which exhibit three different poles, namely a thermal pole, a chemical pole and a mechanical pole. This choice is not a well-adapted framework to deal with the phenomena describing the degradation process of refractory used in steel industries. The general coupling scheme proposed in figure 1 seems to be better adapted. The thermo-mechanical behaviour of the refractory structures, including complex mechanical behaviour of the material, classical momentum balance and heat transfer, interacts with transport phenomenon by capillary impregnation of liquid slag or chemical diffusion of species, and this at different scales. This interaction could be, for example, the temperature activation of diffusion or the interstitial liquid effect on the effective stress in a porous media.

The transport phenomenon is of major importance as it drives the chemical reactions. Indeed, it puts in presence (or not) chemical reagents producing an evolution of the state of the material (microstructure, physical and mechanical properties).

The last pole is not the least because "chemical reaction", such phase change or dissolution could directly interact with the stress field, due to chemically induced bulk variation, or modification of the mechanical behaviour. For example, the partial dissolution or melting of a bounding phase can induced a transition from elasticity to creep.



Figure 1: a general coupling scheme for the study of refractory structures

All classical degradation scenarios of damage observed in refractory lining used in steel industrial reactors can be described in this framework⁸. If one were able to model all the mechanisms of the coupling diagram, one would be able to account for all the possible degradations. However it is very difficult to model the whole degradation processes in their full complexity. Consequently, the decomposition of the whole problem into several simple sub-problems is necessary to bypass the difficulty.

From a general point of view, a refractory lining can be divided into two zones: an impregnated zone and an unimpregnated one. On the surface in contact with slag the impregnated zone is dissolved and the impregnation front moves inside the thickness. Consequently, the existence of the impregnated zone results from the competition between the dissolution rate and the impregnation rate. However, when there is an impregnation, it is observed that after a transient regime where the impregnated zone grows, the thickness of this layer remains constant. Consequently, the impregnation front moves with the same rate than the dissolved one. Then, in a first approximation, it is reasonable to consider only the steady state of impregnation, when the thickness of the impregnated zone remains constant. The habit to model the refractory by a classical continuous and homogenised medium cannot allow accounting well for the coupling between impregnation (transport phenomenon) and thermomechanics (Fig. 1). One option possible is to consider the slag-impregnated zone like a porous medium where the brick constitutes the skeleton, gases and molten slag the fluid phases. This approach has already been developed in civil engineering in the framework of the mechanic of porous media initially proposed by Biot⁹ and developed by Coussy⁵. In this work, we will adopt the framework proposed by Coussy⁵.

The coupling scheme proposed above is used herein to identify the main factors controlling spalling of bauxitebased working linings of steel ladles. First, the coupling between impregnation and phase change is studied by the classical crucible test and EDS analysis. It helps us to characterize the different zones (e.g., impregnation, dissolution) for the refinement of the mechanical analysis. Then, the mechanical effect of the liquid present in the porosity is quantified. Lastly, after a short presentation of the modelling of the asymmetric creep of such refractory, an acceptable scenario for spalling is proposed.

Chemical gradient linked to slag impregnation

Because of high temperatures in service (1200-1650°C), thermo-chemistry simulations based on data collected by EDS were carried out to estimate the likely state of mineralogical composition existing in service^{8,11}. Data were obtained at room temperature by observations using a scanning electron microscope (SEM) (Hitachi S-510) equipped with a dispersive X-Ray spectroscopy analyzer (Princeton Gamma-Tech XS14-H005).

The analysis was first performed on a crucible corroded by slag during 5 hours at 1600°C. It appears that phosphorus present in the bounding phase of the brick can be used as a natural tracer of impregnation. Results give guideline for the analysis of a worn sample taken from a steel ladle lining^{8,11}. The post-mortem analysis focuses on the content of phosphorus pentoxide (P₂O₅) initially present in the bounding phase of the refractory and lime (CaO) initially present in the slag. They are indicators of the impregnation state. The chemical content distribution is thus particularly interesting. Figure 2 shows the content of these oxides with the depth.



Figure 2: Impregnation gradient of a worn brick

Along the first millimeters, the slag dissolves the refractory phases. New phases are precipitated after saturation of the liquid, changing the chemical composition of the liquid. The latter is enriched with components of the dissolved bounding phase (phosphorus) and becomes impoverished in lime (precipitate).^{10,11} This liquid in equilibrium with the refractory components impregnates the lining behind the dissolution zone.

The obtained results allow for the identification of four distinct areas at 1600°C: the last slag deposit area; a mushy / dissolved zone where the slag dissolve the refractory; an impregnated zone where the liquid in equilibrium with the refractory totally fill the porosity; an unsaturated impregnated zone and an unimpregnated zone (Fig. 2).

As cracks which are at the origin of spalls are localized in the impregnated zone saturated by liquid, it asks about the role played by the liquid in the failure mechanism.

Mechanical effect of interstitial liquid

The influence of the liquid fully saturating the impregnated area is examined. The fluid flow is governed by Darcy's law, i.e., V = k grad P, where V is the filtration vector, k the hydraulic conductivity and P the fluid pressure. The following additional properties are used in the numerical model. Because the liquid content increases with temperature and the kinematic viscosity decreases with temperature both the porosity ϕ and the hydraulic conductivity k of the refractory depend strongly on the temperature. Yet, due to a lack of data, they are assumed equal to the value obtained at room temperature, namely, $\phi = 0.12$ and $k = 10^{-12}$ m.s⁻¹. The mass density of the liquid phase is equal to $\rho = 2,690$ kg.m⁻³. The thermal expansion of the liquid is assumed identical to that of the refractory (Table I).

According to these assumptions, the setting of the problem's equations¹² yields to the definition of the coefficient of relative bulk variation δ_v :

$$\delta_{\nu} = \alpha_o \left(b \left(\frac{1 + \nu_o}{1 - \nu_o} - 3 \right) + 3 \right) - 3 \left((1 - \phi) \alpha_s - \phi \alpha_{fl} \right) \quad (1)$$

where *b* is Biot's coefficient depending on the skeleton and fluid compressibility, v_o Poisson's ratio, α_s and α_{fl} the coefficients of thermal expansion of the skeleton and fluid, respectively and α_o the drained coefficient of thermal expansion. For the application $\delta_v \tilde{\approx} -1 \times 10^{-5} \text{ K}^{-1}$.

The analogy between heat conduction and mass flow diffusion, leads to define the hydraulic diffusivity¹²:

$$D_{H} = k \left(\frac{1}{M} + \frac{(1+\nu)b^{2}}{3(1-\nu)K_{o}} \right)^{-1}$$
(2)

where *M* is Biot's modulus depending on porosity, skeleton and fluid compressibilities ($M = \infty$ for both incompressibility), K_o the impregnated brick compressibility in drained conditions.

A reference pressure P_{nd} can be established by considering the undrained case¹²

$$P_{nd} = -\delta_{\nu} \left(\frac{1}{M} + \frac{(1+\nu)b^2}{3(1-\nu)K_o} \right)^{-1} \theta_{\max}$$
(3)

where θ_{max} is the amplitude of the thermal loading.

The study of a one-dimensional half space submitted to a harmonic surface temperature permits to characterize the effect of the prevalent parameters.¹² Indeed, this theoretical case allows underlining the effect of the cyclic thermal loading imposed by the steel refining process. It appears that the maximum dimensionless pressure P_{max}/P_{nd} and its dimensionless location X_{cr} only depend on the diffusivity ratio. These results are summarized in figure 2.

It is worth nothing that the characteristic length of a harmonic thermal loading of a frequency ω is¹²

$$L = \sqrt{\frac{2D_T}{\omega}} \tag{4}$$

The localization of the maximal interstitial pressure is linked to the frequency of the thermal loading. There is a real difference between a monotonic and a cyclic loading. The first one does not induce a localisation of the interstitial pressure.¹² But, even for a low level of interstitial pressure, the cyclic thermal loading should induce a fatigue of the material at a localized abscissa.

Because of the large range of fluctuation in porosity and viscosity with temperature and liquid composition, the bauxite based refractory presents a diffusivity ratio D_H/D_T which could vary from 10⁻⁴ to 10. Consequently, for a typical steel refining cycle, the interstitial liquid pressure during heating stage should vary from 0.2

MPa, up to 12 MPa and the critical abscissa x_{cr} from 0 up to 12 cm.⁶



Figure 2: Maximum dimensionless pressure and dimensionless location vs diffusivity ratio

For a given diffusivity ratio the pressure only depends on the thermal amplitude whereas the critical abscissa depends on the frequency of the loading.¹² So lower the frequency is so higher the critical abscissa. For the average diffusivity ratio a maximal liquid pressure of 8.8 MPa is reached at an abscissa of 1.5 to 3 cm, depending on the ladle rotation (130 min heating / 70 min and 300 min heating / 250 min cooling). This high interstitial pressure just behind the hot face should greatly favour spalling.

These estimates of the maximal pressure and its location is valid for an elastic skeleton. Be that as it may, it gives a qualitative understanding of what should happen and what the influences of the prevalent parameters are. To quantify this effect, we should account for a more realistic mechanical behaviour of the skeleton.

Asymmetric creep behaviour

If at low and medium temperatures ceramics exhibit an elastic quasi-brittle behaviour, time-dependent inelastic behaviour is the dominant thermo-mechanical feature at high temperature, typically above half of their melting point. Although considerable work on the creep of ceramics exists in the literature^{13,14,15} any model reproduces the behaviour of bauxite based refractory. Indeed, such type of refractory exhibit high viscoplastic behaviour with a significant difference in tension and compression due to the presence of creep-resistant oxide grains (i.e., alumina) and a glassy matrix (rich in silica) with a much lower creep resistance. Thus, a new model is proposed for the asymmetric creep behaviour of the refractory¹⁶. A simplified version of this model in which the asymmetry is only applied to the viscoplastic kinetic without yield stress is presented below.

The stress tensor $\boldsymbol{\sigma}$ is decomposed in the principal frame into a tensile part $\langle +\boldsymbol{\sigma} \rangle$ and a compressive one $\langle -\boldsymbol{\sigma} \rangle$.

The elasticity law is assumed not depending on the sign of load

$$\boldsymbol{\sigma} = \mathbf{C} : \left(\boldsymbol{\varepsilon} - \boldsymbol{\alpha} (T - T_o) \mathbf{I} - \boldsymbol{\varepsilon}_{vp} \right)$$
(5)

where C is the elastic stiffness tensor, ε the total strain, '.' the contraction with respect to two indices, α the coefficient of thermal expansion, T the current temperature, T_o the reference temperature, I the identity tensor and ε_{vv} the viscoplastic strain.

The kinetic law of the viscoplastic strain is the sum of two parts corresponding to viscoplastic strains developed under tensile and compressive stresses respectively

$$\dot{\boldsymbol{\varepsilon}}_{vp} = \frac{3}{2} \left(\frac{\sqrt{\frac{3}{2}} \boldsymbol{S}^+ : \boldsymbol{S}^+}{K_t} \right)^{n_t} \frac{\boldsymbol{S}^+}{\sqrt{\frac{3}{2}} \boldsymbol{S}^+ : \boldsymbol{S}^+} - \frac{3}{2} \left(\frac{\sqrt{\frac{3}{2}} \boldsymbol{S}^- : \boldsymbol{S}^-}{K_c} \right)^{n_c} \frac{\boldsymbol{S}^-}{\sqrt{\frac{3}{2}} \boldsymbol{S}^- : \boldsymbol{S}^-}$$
(6)

where $\dot{\boldsymbol{\varepsilon}}_{vp}$ is the viscoplastic strain rate, (K_t, n_t) the coefficients of Norton's law of tensile creep, (K_c, n_c) the coefficients of compressive creep, \boldsymbol{S}^+ and \boldsymbol{S}^- are the deviatoric part of the tensile and compressive part of the stress tensor respectively.

To identified the model compressive and three-point bend tests were performed for un-impregnated and bauxite-based refractory at impregnated high (1000°C, 1250°C temperature and 1400°C). Experimental set-up and results are developed in ref. 16. It is considered that $n_t = n_c = n$ since the tensile creep exponent in tension is nearly the same as in compression. The ratio K_c/K_t is identified at the temperature θ = 1250 °C and is assumed constant over the temperature range 1000 °C-1400 °C. The thermal expansion is measured after stabilisation of the microstructure. Results of identification are summarized in table 1.

Figure 3 shows the comparison between identification and experimental results for three points bending test under 1250°C. The model well reproduces the load rate sensibility and the high asymmetry between tension and compression. Simulation results close to the stress peak have to be takes with care since the model does not account for damage. Simulation results after the peak cannot be good for the same reason.



Figure 3: identification of the model for the impregnated refractory at 1250°C (3 point bending test)

Spalling scenario

Numerical simulations of the complete structure of vessels such as a steel ladle or a converter have been performed by several authors.^{3,18} Yet, the highly non-linear behaviour of refractories, the presence of joints between the layers and between the bricks in the working lining, the complex loadings make the computations very difficult. Thus, quite often, strong simplifications are made to run calculations and determine the stress state in the lining.

$E = max \{34.9 - 0.0249 \ \theta, 0.78\}$
<i>v</i> =0.2
$n = max\{8.43 - 0.0054 \ \theta, 1.03\}$
$K_c = \min\{0.25 \ e^{0.0077.\theta}, 9,535\}$
$K_c/K_t = 8.5$
$\alpha = 4.5 \ 10^{-6} + 2.0 \ 10^{-9} \ \theta$
$\phi = 0.12$
$k = 10^{-12} \mathrm{m.s^{-1}}$

Table 1: Thermomechanical properties of bauxitebased refractories in the temperature range 900- $1,650 \ ^{\circ}C \ (\theta \ expressed \ in \ ^{\circ}C)$

To study the possible scenario of spalling the ladle is reduced to a ring with an inner radius of 2 m and an outer radius of 2.15 m. It is assumed that the lining is a porous medium fully saturated with liquid slag over a 60 mm wide zone. The skeleton of the porous liquid saturated medium follows the asymmetric creep behaviour present above. Terzaghi's effective stresses⁵ σ^{eff} defined from the total stresses σ by $\sigma^{eff} = \sigma + P.I$ are related to effective strains ε^{eff} defined from the total strain ε by $\varepsilon^{eff} = \varepsilon + (P/K_g.I - \varepsilon_g^{th})/3$. The unimpregnated porous medium is described by the continuum homogeneous medium with the same behaviour than the skeleton of the porous medium. The heat transfer problem is computed first. Then, the coupled mass transfer and mechanical problem is solved by introducing the space-time temperature field as a bulk loading to compute the thermal strain, the fluid mass flow and the temperature-dependent material parameters. Mesh size and type, thermal and mechanical boundary conditions are developed in ref. 17.

The cyclic thermal loading depends on the refining process. The heating stage is fast during the filling of the vessel (thermal shock), then there is a long dwell (refining) and a low cooling stage after the emptying of the ladle (Fig. 4).



Figure 4: thermal loading of the simplified ladle

Figure 5 shows the evolution of the interstitial pressure with time at several depths.



Figure 5: pressure of interstitial liquid at several depths

The interstitial pressure *P* is positive during the beginning of the heating stages due to the relative bulk variation δ_v . As the permeability is low, the fluid mass flow is not sufficiently fast to relax the pressure. Moreover, as incompressible visco-plasticity is used, the pressure can not be relaxed by growing of voids. A maximal pressure in the region of 1 MPa is reached about 20 mm from the hot face. Then, it decreases due

to the high viscosity and even becomes negative with a minimum around of -0.1 MPa at the 50-mm zone. It could generate suction or gaseous phases. During the cooling stages, the pressure remains low because the solid skeleton is much softer at higher temperature and the thermal shock (i.e., temperature rate) lower. Negative pressure occurs with a minimum value around -0.2 MPa at the middle of the impregnated zone. It is obvious that the high viscosity of skeleton relax the pressure in comparison with the pressure predicted with poroelastic behaviour of skeleton.

During the heating steps, compressive axial and hoop stresses also occur (ring heating by its inner face); they are nearly identical. The "maximal" compressive biaxial stress is reached in the zone near the limit of impregnation during the heating and could favour the onset of microcracks parallel to the hot face in this zone. During the cooling stages biaxial tensile stresses occur but they remains low, typically lower than 0.5 MPa because of the asymmetric creep behaviour. The radial stress in the skeleton is equal to the pressure Pbecause of the whole momentum balance. During the beginning of the heating up, the working lining is subjected to a biaxial compressive stress state with a radial tensile stress. This stress field seems to be highly favourable for buckling.

Conclusion

A general coupling scheme has been proposed to study degradations of refractory structures where both chemistry, thermal and mechanic phenomena are involved. This coupling scheme underlines that coupling between chemistry and thermomechanics often needs a mass or species transfer phenomenon. The modelling of this latter is a key point to well understand and compute the scenario of degradation.

The mechanics of porous media⁵ is a powerful tool to model slag impregnation and its mechanical effects. In this first approach, only the steady state of impregnation is considered and the model is limited to the liquid saturated area. With these assumptions, it is established that the liquid which fill the porosity play a mechanical role in the spalling phenomenon. The influence of the thermal loading type (cyclic or monotonic) is also underlined.

To bypass the previous assumptions, it is necessary to model the transient stage of impregnation. It requires the computation of transient reactive impregnation. Today, it is already possible to compute transient nonreactive impregnation.⁵ The difficulty is to access to relevant experimental data such as sorption curves with liquid slag (at service temperature...) to supply the numerical simulations of multiphysical phenomena. However, the simulation of reactive impregnation requires further development on the numerical coupling between thermochemistry and finite element model. No matter what is the difficulty, the development of the model of reactive porous media seems to be a promising way for further development in refractory.

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