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## Review article

## Flow behaviour of crystallising coal ash slags: Shear viscosity, non-Newtonian flow and temperature of critical viscosity

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### ABSTRACT

The crystallisation in coal ash slags plays a critical role in coal combustion and gasification. The stable operation of a boiler or entrained-flow gasifier is vitally dependent on the easy, continuous removal of combustion byproducts in the form of coal ash slags. Of the factors that govern the flow of a coal ash slag in a reactor, slag viscosity is the most important. Crystallisation affects overall slag behaviour and properties, but its most pronounced effect is on slag flow characteristics. It increases slag viscosity, causes Newtonian slag flow to transition to non-Newtonian, and yields the temperature of critical viscosity. In this paper, we systematically review and analyse the complex effect of crystallisation on these three factors.

#### 1. Introduction

Coal ash slags are multicomponent oxide mixtures commonly generated in coal combustion and gasification from incombustible coal ash residue [1–3]. Although most of the coal burned in a reactor (e.g. gasifier, boiler) forms syngas (a mixture of CO, CO<sub>2</sub>, H<sub>2</sub>), residual ash particles are entrained by the gas stream, settle on the reactor walls and flow down to a slag discharge facility. Coal ash slags must be fairly fluid at process temperatures and pressures to ensure their easy removal from the reactor. Slag flow characteristics, such as viscosity, play a critical role in stable gasifier operation, and need to be addressed by both experimental determination and modelling. Two other important features of slag flow in gasifiers – the temperature of critical viscosity and transition from Newtonian to non-Newtonian flow – also need to be considered.

Coal ash slags behave as Newtonian fluids under a wide range of conditions (e.g. different temperatures, pressures and atmospheres) encountered in the industry. If a Newtonian slag has a viscosity coefficient within acceptable limits (typically between 5 and 25 Pa·s) at operating temperatures (1200–1500 °C) [4], it provides steady operating conditions in an entrained-flow gasifier, because it continuously flows down to a tap hole in the gasifier. During steady gasifier operation, slags can either be homogeneous (i.e. only in the liquid phase) or heterogeneous (i.e. the liquid phase contains one or more other phases). The viscosity–temperature dependence of a typical coal ash slag is shown in Fig. 1 [5]. With decreasing temperature, slag viscosity increases steadily until lower temperatures, at which a sharp increase

may occur. The temperature at which this happens is usually referred to as the temperature of critical viscosity ( $T_{cv}$ ). Critical viscosity and  $T_{cv}$  are often associated either with the onset of slag crystallisation, or with non-Newtonian transition of slag flow.

The appearance of any heterogeneous inclusions (e.g. crystals, gas bubbles) in a liquid slag is generally caused by a phase transformation due to changing conditions (e.g. decreasing gasification temperature or pressure). Phase transformations have a pronounced and complex effect on slag behaviour and properties. The most frequently observed phase transformation in coal ash slags is crystallisation: i.e., the precipitation of solid crystals out of the liquid phase with decreasing temperature below the slag liquidus. The flow of partially crystallised slags can still remain Newtonian up to a certain fraction of solid phase in the liquid slag.

At higher degrees of crystallisation, slag flow turns into non-Newtonian: i.e., the slag viscosity can increase (shear thickening) or decrease (shear thinning) with an applied shear stress. Other complex effects (e.g. normal stresses, pseudo-plastic behaviour) can also occur during slag flow. At a certain level of crystal content in slag, its viscosity increases by orders of magnitude, and the viscous flow practically stops.

Crystallisation in coal ash slags can occur under a wide range of operating conditions, and it is difficult to foresee what variation of operating parameters (temperature, pressure or coal chemical composition) can cause problems in gasifier or boiler operation. For example, a slight decrease in temperature can sharply increase slag viscosity due to the onset of crystallisation, drastically affecting slag flow behaviour. Slag with excessively high viscosity can block tap holes, leading to

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Fig. 1. Viscosity-temperature dependence of a typical coal ash slag.

gasifier shutdown.

The gasifier temperature, pressure or coal chemical composition at which crystallisation may start in the coal ash slag, as well as the proportion and composition of crystallising solid phases, can easily be estimated using a thermochemical software package (e.g. [6,7]). This approach is sometimes referred to as the global equilibrium analysis [8], i.e. a gasifier is considered to be in the global chemical equilibrium that can be modelled using special software, which minimises the total Gibbs energy to obtain phases that are in equilibrium at a given temperature, pressure and bulk chemical composition. Equilibrium calculations are not an easy task. They can only be performed using numerical methods and powerful computers, and by constructing and using large thermodynamic databases in which thermodynamic properties of multicomponent slag phases are stored. The FactSage [9] and MTDATA [10] software packages appear to be most suitable for calculations of coal ash slags. Examples of equilibrium calculations for a geological melt [11] using the MTDATA and FactSage software, respectively, are shown in Fig. 2(a and b). Both predictions agree reasonably well with the experimental data. The first phase that crystallises out in equilibrium under given conditions is usually referred to as the primary phase. In Fig. 2, this is feldspar, which is a solid solution between anorthite and albite.

In the real-life operation conditions, the overall state of the gasifier can shift far from the theoretical global equilibrium, especially with temperature varying from 2000–2200 K in the combustion zone to 1000–1300 K at the inner surface of water-cooled walls. Nevertheless, a local chemical equilibrium in various parts of the gasifier (e.g. in the slag deposit flowing down the gasifier walls) can still be attained [12], because the corresponding chemical reactions' rates in selected regions are generally much higher than those of the concurrent physical phenomena. The local equilibrium concept is extensively used for studying physico-chemical phenomena (e.g. viscous flow, kinetics of crystallisation etc.) inside gasifiers that are linked with experimental data of real gasification processes.

The kinetics of crystallisation also affects slag behaviour. Crystallisation typically occurs in two stages: nucleation and crystal growth. In the classical nucleation theory, the homogeneous nucleation rate of solid nuclei in a liquid matrix depends on the difference in chemical potentials of the solid and liquid phases and the interfacial energy. The crystal growth rate is governed by the undercooling, diffusivity and/or viscosity. Crystallisation kinetics in many coal ash slags can be very fast (see e.g. [13,14]). Unfortunately, no ready-to-use computer tool such as FactSage is available to evaluate slag crystallisation kinetics. As a result, viscosity measurements of coal ash slags are often carried out under non-steady crystallising conditions, e.g. during continuous cooling with a constant cooling rate. Although such



**Fig. 2.** An example of the equilibrium calculation using FactSage and MTDATA: (a) phase proportions and (b) melt composition. Solid lines show MTDATA predictions, dashed lines show FactSage predictions, and experimental points from [10] are given as symbols.

tests can be in accordance with non-equilibrium gasification conditions, it might be difficult to evaluate the obtained experimental results due to simultaneous variation of many factors during experiments.

Sometimes, a thermodynamically stable phase cannot rapidly crystallise out of the liquid phase, due to kinetic or other restrictions (e.g. slow nucleation or growth rates, high cooling rate). In this case, a time lag in crystallisation can be expected, and viscosity measurements can show no presence of crystalline phases up to a certain time or temperature (e.g. [14]). This situation can be encountered in a laboratory if slag viscosity measurement is carried out at a constant cooling rate that is higher than a certain critical value for a given system.

Coal ash slags may also freeze without crystallisation, forming a glassy or amorphous phase. In this case, crystallisation is completely suppressed due to the slag's high glass-forming ability (GFA). This slag property is usually related to the lowest cooling rate at which the glassy phase can still form.

Exsolution of volatiles (light elements like sulphur or phosphorous) or liquid–liquid phase separation (appearance of another immiscible liquid phase with a different chemical composition) are other examples of phase transformations in the coal ash slags that can strongly affect slag behaviour. Viscous flow of slags containing small gas bubbles or immiscible liquid droplets is very complex due to high deformability of inclusions and is outside the scope of the current paper (for details cf. [5]).

In the present review, we discuss and critically analyse the complex effect of slag crystallisation on the important parameters used to Download English Version:

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