THE ANALYSIS OF THE INTERFACIAL INSTABILITY CONDITIONS IN SYSTEMS LIQUID STEEL- REFINING SLAG

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Abstract: *As consequences of the increasing needs of high quality steel products, besides the improvements of the actual technologies, applied in steel refining process using slugs, a special attention is paid to new possibilities of quality improvement in liquid state in different stages of processing. A practical modality to apply actions to improve the quality, by preserving the beneficial contributions via limiting, avoiding or suppression of undesired effects, is to act at interface of the active involved phases, steel and slag. Both beneficial and deleterious effects originate from turbulence and mixing developed at interface in adequate/inadequate moments and as intensities. Taking into consideration the complex interaction at interface, based on the solutal contributions, it is presented a comparative analysis of the threshold of instability in individual phases, according to one layer model, also of their interface. The conditions for turbulent convection by solutal Marangoni effect are also evaluated.*

Keywords*; Instability threshold, characteristic length, critical solutal Marangoni number, slag, steel.*

1. Introduction

Instability state at interface between two fluids consists in the modification of the interfacial tension (also of the stress state) due the variation of the parameters influencing the interfacial tension (temperature, composition, electric potential), followed by a instantly starting specific flow when an overcoming of a threshold value of the considered parameter takes place. This situation is produced by processes taking place at interface or by external applied actions. The term interfacial tension will be also understood as referring to the surface tension of a fluid. Slugs and steels are totally different fluids as nature and properties, including those properties which influence the evolution and the efficiency of the specific treatments applied to steels for advanced refining. The systems slag-steel are also very different, compared with other systems of two liquids, where the problem of instability is searched for academic or industrial purposes. This makes that the principles of instability in systems consisting in two immiscible fluids (liquid-liquid or gas-liquid) first established not to be valid for refining slag–steel system. It is encouraging that many other situation, generating interfacial instabilities, were found in different other systems of fluids [1], but even so, the interfacial instability at reefing slugs-steel interface is insufficiently approached. The aim of the paper is to put into evidence the conditions of onset instability, by specific threshold values of relevant quantities, in steel and in slag at the most usual technological temperature of 1873K, in order to evaluate the trends possible to be used, in order to exploit them to improve the quality of refined steels under slugs.

2. The Marangoni solutal effect

The surface tension (σ) of a substance in liquid state, containing surface active component, depends upon the concentration of this(c), temperature (T) and the electric potential (ψ). Their variations on a direction(x), along the free surface or of the interface between two immiscible fluids lead to arising of a tangential surface (or interfacial). If the electric surface or interfacial potential is constant or can be neglected and the system is in a thermal equilibrium state, the main factor affecting the surface and interfacial tension is the cariation of the content of the surface active component and the resulting tangential stress can be given by the following relation:

$$\tau_s = \frac{d\sigma}{dx} = \frac{\partial\sigma}{\partial c}\,\frac{dc}{dx} \quad (1)$$

In what it will follow in this paper the term interfacial tension will be used for both because the surface tension is a particular case of the interfacial tension.

The gradient of the interfacial tension due to the variation of the solute at interface acts as a tension on the surrounding liquid called frequently as Marangoni tension and will generate flow or will modify the state of the already existing flow, this being called as Marangoni effect. The solutal Marangoni effect can act:

- at macroscale, as the consequence of the external applied actions;
- at microscale, as a consequence of local concentration differences, produced by certain specific processes, also due to some microinhomogeneities of the surface active component concentrations.

In both cases, but especially at microscale which is more sensitive, there is a particular coupling and fitting of several factors acting in producing of the Marangoni effect and they are connected to the state of the critical instability and the onset of convection.

In order to evaluate these complex actions of factors acting in the Marangoni effect it was introduced [2] a dimensionless number called Marangoni number Ma, which initially was used for thermocapillary action. The form for the solutocapillary action is the following:

$$Ma=|\partial\sigma/\partial c|\cdot\Delta c\cdot L/\rho\cdot\nu\cdot D =|\partial\sigma/\partial c|\cdot\Delta c\cdot L/\eta\cdot D \quad (2)$$

where:

$|\partial\sigma/\partial c|$ - concentration coefficient of the interfacial tension; it is taken as absolute value because of its dependence with concentration;

Δc - characteristic difference of the surface active of the solute concentration along the interfacial surface or across the interface, on the normal direction on the interface at the considered point; in many cases, according to the specific situation instead of Δc, a characteristic concentration c_0 is considered

L - characteristic length, taken into account in the action of the Marangoni stress and the resulting associated Marangoni effect;

ρ - density of the fluid in considered layer at interface;

D - mass diffusion coefficient of the surface active solute;

$\eta=\rho\cdot\nu$- dynamic viscosity of the considered fluid;

ν – kinematics viscosity of the considered fluid.

It is obvious that for the case of a liquid containing a certain surface active solute, the Marangoni number is strongly dependent on the value of the product existing at numerator of the relation (2). Generally, the concentration coefficient of the interfacial tension is enough well known as values and as a general relation, especially for thermocapillar effect. Frequently, for it the Bussinesq approximation is used. It follows that it must be evaluated the value of the characteristic length, corresponding to necessities. The minimal critical solutal Marangoni number was found to be $Ma_c^s=48$ according to ref.[1], in the conditions of absence of differences of densities along and across the interface(dimensionless number Rayleigh Ra=0) and also absence of mass transfer (Biot dimensionless number Bi=0) and the absence of absorption and adsorption. It is obvious that such cumulated conditions exist only locally and temporarily and such fields have not strict rules of occurrence at interface.

3. The establishing of the characteristic length L.

The solutocapillary action is in a tight competition as occurrence with the actions of gravity and mass transfer, this last being evaluated by the diffusion and viscosity as specific quantities. While solutocapillarity acts as a destabilizing factor, gravity and viscous diffusion act as stabilizing factors. Gravity acts to flatten the surface and the viscous diffusion acts to wreck the differences of concentration of the surface active solute. In order to be dominant, the solutocapillary action must be the faster among all and this means it must to have the shortest characteristic time of action. The characteristic time of the mentioned actions is scaled using the following relations:

- characteristic time scale of the gravity:

$$t^2_{grav}= L/g \tag{3}$$

- characteristic time scale of the viscous diffusion:

$$t^2_{diff}=t_{diff}.t_{visc}=(L^2/D)(L^2/\nu)=L^4/\nu.D \tag{4}$$

- characteristic time scale of the solutocapillarity:

$$t^2_{solutocap}=\rho.L^3/|\partial\sigma/\partial c|\Delta c \tag{5}$$

In order to have instability and onset of Marangoni convection as short-wavelength waves, the value of the dimensionless number Galilei [1][3] must have a value enough high, overcoming the minimal value:

$$Ga=t^2_{diff}/t^2_{grav} \geq 3/2Ma_c^s \tag{6}$$

This means, for $Ma_c^s=48$, it results:

$$Ga\geq 72 \tag{7}$$

According to rel (6) it results:

$$Ga=t^2_{diff}/t^2_{grav}=gL^3/\nu D \tag{8}$$

From relations (2)(6)(8) it results the following relations :

$$Ma_s^c=2/3Ga^c=2/3gL^3\nu D \tag{9}$$

$$L=(Ga.\nu.D.g^{-1})^{1/3}=(3/2Ma_s^c\nu.D.g^{-1})^{1/3} \tag{10}$$

For $Ma_c^s=48$ and acceleration of gravity g= 9,81m/s^2 results:

$$L=1.943366\ (\nu.D)^{1/3} \tag{11}$$

It results that at known acceleration of gravity, on Earth and in space, at low gravity, the characteristic length scale L for the minimal solute critical number Marangoni depends only upon the relevant physical quantities for mass transfer, mass diffusion coefficient of the surface active solute in the considered liquid phase and the kinematic viscosity of the liquid phase, at chemical composition in bulk. A special attention must be paid to the accuracy and precision of the computations because. Approximations and rounding of any kind of the numerical results of steps in computations are not recommended.

4. Values of the instability threshold and onset Marangoni convection

It results from relation (2) that as threshold values is important the following derived quantities:

1. the threshold of the solute concentration difference, representing the difference of concentration of the surface active component Δc for which the instability occurs steeply in minimal conditions, that means in this case for value $Ma_c^s=48$:

$$\Delta c_{crit}=Ma_s^c\cdot\rho\cdot\nu\cdot D\cdot L^{-1}|\partial\sigma/\partial c|^{-1} \tag{12}$$

2. the critical value $(\Delta\sigma)_{crit}$ of the difference of the interfacial tension at which the instability occurs steeply in minimal conditions, that means in this case for value $Ma_c^s=48$:

$$\Delta\sigma_c = [|\partial\sigma/\partial c|\Delta c]_{critic} =Ma_s^c\rho.\nu.D.L^{-1} =24.699.\rho.(\nu.D)^{2/3} \tag{13}$$

In the preliminary moments, just before the steeply rising of the values of the involved parameters up to level of instability threshold, all necessary conditions regarding the mass transfer, adsorption and eventual absorption, mentioned at the end of paragraph 3. are fully fulfilled.

Based on the before statements the conditions of the direct evolution to the turbulent flow by Marangoni effect can be established. The turbulent Marangoni convection means conditions of chaotic flow where the convection under the form of rolls and hexagons is strongly alterated, these patterns being either mixed and strongly deformed or even they are not at all identifiable. This kind of convection occurs at higher values of the Marangoni number, starting with values corresponding to the relation [8]:

$$\varepsilon = (Ma_s - Ma_s^c)/Ma_s^c > 6.3 \quad (14)$$

5. Specific parameters of instability and convection in the CaO-Al_2O_3 slag and in steel.

The removal of sulphur from steel into slag is described chemically by the reaction:

$[S]+(O^{2-})\Leftrightarrow(S^{2-})+[O]$

The reaction takes place during steel refining under slugs. Usually these slags belong to the complex system CaO-MgO-Al_2O_3-SiO_2 and low contents of MgO (< 8 mass %) and SiO_2 (< 5 mass %), when a performing treatment must be realized. For an advanced desulphurization, also for an advanced removal of non-metallic inclusion, slugs composition includes contents high levels of CaO (45-55%). The contents of Al_2O_3 are in the range of 30-40% for reasons of fluidity.

Table 1. Relevant properties, direct and derived quantities at 1873K for threshold of instability, in a CaO -Al_2O_3 slag and in a low carbon, aluminium killed steel.

Phase Property, quantity	Slag 50% CaO-50%Al_2O_3	Steel C=0.1% ;Mn=1,8% ; P=0,02% ;S=0,015%; Al_{sol}=0.01%
Sulphur solubility, mass%	1.466 [6]	Very high
Density , Kg/m^3	2710 [4]	6970
Dynamic viscosity, η, $N.s\cdot m^{-2}$	0.197 [5]	0.00408 [7]
Sulphur mass diffusion coefficient D_S , m^2/s	2.9×10^{-10}	4.4×10^{-9}
Surface tension, mN/m	$\sigma=1.4781(\%S)^2-22.159(\%S)+546$	1714-1800
Interfacial tension, mN/m	$\sigma_i=0.7928(\%S)^2-31.548(\%S)+1316.5$	
Characteristic length L, m	53.685×10^{-6}	25.972
Critical variation of tension, $\Delta\sigma_{crit.}$, $\times10^{-3}$ mN/m	51.08	30.748
Critical variation of sulphur, $\Delta(S)_{crit.}$, mass%	(23.05-28.66) 10^{-4} %	4.58×10^{-6} %
Critical variation of sulphur concentration for ε=6,3 (onset of turbulent Marangoni convection) $\Delta(S)^t_{crit.}$, mass%	$(145.22-180.56)\cdot10^{-4}$ %	$28.85\cdot10^{-6}$%

A comparison between quantities and parameters giving the threshold of instability between such slag and a treated low carbon steel is presented in the table1 where a slag containing CaO=50mass% and Al_2O_3=50% mass is considered because of specific available data for computations. It is remarkable that despite of all structural and major differences of properties between the liquid steel and the slugs, the phenomenon of instability due to solutocapillary effects of sulphur in slag takes place at much closed values of the parameters and especially of the difference of the interfacial tension $\Delta\sigma_{crit.}$

From table 1 it results that the minimal level of critical concentration of sulphur $\Delta(S)_{crit}$, for onset short wavelength waves Marangoni convection (Ma_s^c=48) is very low in slag, also in steel, being in the range of errors and accuracies of methods and analyses used in industrial practice and in many laboratories. The concentration conditions for sulphur $\Delta(S)^t_{crit}$, for the onset instability with turbulent Marangoni convection in minimal conditions Ma_s^c=48, are also low, despite that are 6.3 times higher than that those corresponding to the initial stage of Marangoni convection. These data are useful to evaluate the extent of the convection by solutal Marangoni effect and its relation with other forms of convection, including the other forms due to the Marangoni effect, in order to manage and lead the processes in the desired direction.

6. Conclusions

It was shown that instability and convection by solutal Marangoni effect take place in desulphurizing and refining slugs at low levels of the critical difference of sulphur content in slag $\Delta(S)_{crit}\approx 25\cdot 10^{-4}$mass%, values situated below the contents usually determined and errors of methods and devices. The equivalent critical variation of the interfacial tension is about $\Delta\sigma_{crit}\approx 50\cdot 10^{-3}$ mN/m.A simple examination of the same parameters in a low carbon, aluminium killed steel shows similar conditions but levels of the critical concentrations lower with two order of magnitude. Values of $\Delta(S)_{crit}$ in slugs are about 500 times higher than in steel. On these data, the emulsifying of steel and slag during desulphurization, especially in the early stages can be better explained, on the base of the turbulent Marangoni convection through solutal effect, which seems to be possible in common conditions at industrial scale.

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