

An application of numerical simulation of multiphase flow for the redesign of a mixer agitator in Pb refining process

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Abstract. The paper presents numerical simulation of multiphase turbulent flow in a mixing crucible unit. Results of simulation were used for redesign of mixer agitator to achieve better performance of the Pb refining process. The simulation is based on Euler-Lagrange description of turbulent multiphase flow with the one way coupling, due to low concentration of solid state particles and significant differences in density of coexisting phases, base metal and particles. Dispersions of solid particles are traced using stochastic-deterministic approach. The developed construction of an agitator has been tested in the industrial Pb refining factory, giving very promising results in comparison with long term statistical data. Duration of unit operations of removal copper and tin was reduced of about 40% together with even better removal efficiency and less energy and reagents consumption.

1. Introduction

Pyro-metallurgical refining of production lead is a dominant process for separation of impurities from the metal to get pure commercial refined lead. Due to high solubility of several metals in liquid lead, raw lead contains numerous impurities. Depending of a technology used for production of raw lead the content of elemental Pb varies between 90% and 98%. The rest are impurities where primary are the following metals: cooper, tin, arsenic, antimony and others.

Refining is carried out in a batch manner. The refining plant consists of a series of batch reactors in which consecutive recovery of dissolved metals takes place by means of adding chemical reactants to produce a phase that is insoluble in base metal, and enriched with certain impurity materials. The single unit operation starts when feeding with „bullion” (molten raw lead) of the mixer ladle is finished. After feeding with the „bullion” the agitator is gradually accelerated to attain predefined angular velocity, at which the melt is intensively mixed in the pot. During mixing the refining compound is supplied on the bath surface in the vicinity of mixer shaft.

Depending on the agitator shape, the certain melt circulation pattern can be observed [1]. When the agitator is of the turbine type, pumping the melt downward the pot (classical Pb refining case), the funnel sucking the refining compounds is formed on the melt surface around the agitator shaft. The particles of refining compounds (solid state loose material) are sucked into the melt and transported around the mixing ladle volume. In a contact with base material and dissolved impurities the oxidation (or sulphurization) heterogenic reactions take place. It is commonly believed that the reactions are

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controlled by the diffusion process at the surface of refining compound particles, therefore high intensity of mixing and high turbulence is promoting the chemical process. As a result of chemical reactions compounds insoluble in the parent metal are formed. Insoluble products flow out on base metal surface and can be removed automatically from the boiler. In the pyro metallurgical process of refining lead highly adverse phenomenon of a skull buildup on the side walls of a ladle occurs relatively often. The skull buildup is observed in these boilers, in which oxidation processes are carried out. Such a phenomenon is unfavourable due to the necessity of hammering or dissolving these buildup that sometimes significantly reduce the volume of the boiler. Periodic cleaning of the walls of the boiler is a long and tedious operation and at high frequencies significantly infers with the production cycle.

1.1. Motivation for research project

The main impulse for research on the process of refining production lead was to reduce the frequency of required cleaning operations of the boiler walls. As a result of the chemical analysis of the skull, which held a composition similar to that resulting from the refining of waste products, it was considered that the most effective limiting factor in their formation and growth will be intensification of the flow in the upper zone of the boiler. This is in harmony with the settled view that the most important phenomenon, which governs the mixing process in the refining ladle, is the molten base metal recirculation. The recirculation pattern depends on numerous parameters of the process but the most important ones are: the mixer agitator shape, its rotational speed and positioning in the pot [1]. The numerical simulation of the base metal flow seems to be an economical and powerful tool for the investigation of optimal shape of the mixer agitator. Thus, preliminary computations of fluid flow induced by the stirrer has shown that intensive bath circulation area is focused around the agitator and shaft, but in the upper zone, near the walls of the boiler, there are areas with low flow velocities. It was supposed that this is a result of very simple design of originally used mixer, consisted of two rectangular stirrer blades inclined at an angle of 35 degrees to the axis of rotation. The agitator redesign had to be done in order to increase the flow speed of the metal near the walls of the boiler by reducing the radial component of the velocity in favour of vertical component. The design process has two fundamental restrictions that need to be fulfilled: the drive energy consumption and the maximum rotational speed of agitators were to be limited to the level of the previously used, and provide suction of the refining salts from the surface into the bulk fluid in the vicinity of the shaft.

2. Mathematical model development

In this section we will formulate the mathematical model of the stirring process using fundamental equations of fluid motion. We will consider multiphase flow in the refining ladle, where the molten raw lead is a carrier phase and the refining compounds (loose solid material) is called dispersed phase. With the intense circulation of carrier phase the refining compound particles fed at the surface are entrapped and thinly scattered in the volume (bulk flow). This phenomenon contributes to an increase in the intensity of the heterogeneous chemical processes as a result of the reduction of barriers to mass transport. Optimum circulation of carrier phase, therefore, is a key issue in the process of refining. Numerical analysis of multiphase phase flow distribution can be taken in many ways [2,3,4,5]. However, one is most favourable from the point of view of clarity of the chemical process description and a minimum number of assumptions required for its application. This is called the mathematical formulation of the Euler–Lagrange multi-phase flow with scattered phases [6]. The flow of carrier phase is described by the classical turbulent flow equations formulated in the Euler’s coordinate system (the observer), and dispersed phase flow equations are expressed in Lagrange frame of reference (moving along with elements of distributed phase in their traffic). The application of such a flow model is confined to the case of a small enough mass participation of dispersed phase in overall mass of fluid, to be able to neglect the dispersed phase volume in the area occupied by the carrier fluid. The movement of a single particle of dispersed phase (of mass m_p and velocity vector \mathbf{v}) is then determined by the momentum balance equation in the form:

$$\frac{d(m_p \mathbf{v})}{dt} = \beta(\mathbf{u} - \mathbf{v}) + \mathbf{F}_c \quad (1)$$

where the forces acting on the particle:

- the drag force proportional to the difference in the speed of the particles \mathbf{v} and mean velocity \mathbf{u} of the carrier phase in turbulent motion, where β is a momentum exchange rate coefficient of a rigid spherical particle [5],
- sum of other forces \mathbf{F}_c acting of the particle (only buoyancy force has been included here).

Integrating particle momentum equation yields the trajectory of the particle in the Eulerian frame of reference. As the momentum balance equation velocity vector \mathbf{u} component express the mean flow velocity of carrier phase (mean in the view of turbulent description of the flow), the traced particle trajectory is called to be deterministic (or mean trajectory).

Simultaneous flow simulation of coexisting phases can include the interplay between them by means of the momentum exchange due to interfacial drag forces (and mass and energy exchange in the extended non-isothermal and chemical reaction model). This is usually implemented by the evaluation of local in space and time momentum (and mass and energy) sources in the carrier phase flow model equations [6]. This is called as two way coupling of the multiphase flow. The back coupling of momentum balance equations of coexisting phases is usually considered essential for the case when the considerable amount of dispersed phase, with relatively large density, is mixed with carrier phase thus having effect on the mixture flow. In our consideration the back coupling has been neglected due to quite adverse conditions, the dispersed phase has low density and overall mass of it is much less than the carrier phase.

2.1. Geometry

The refining cylindrical pot of inner diameter $\Phi=2.36$ m is rounded at the bottom and partially filled with raw lead up to the free surface located approximately at $H=1.85$ m over the pot base. The pot cavity is closed by the cap, where propeller electric engine is mounted. At the beginning of the refining campaign the pot is fed by the raw melted lead and the agitator is immersed into the bath to a depth h , figure 1.

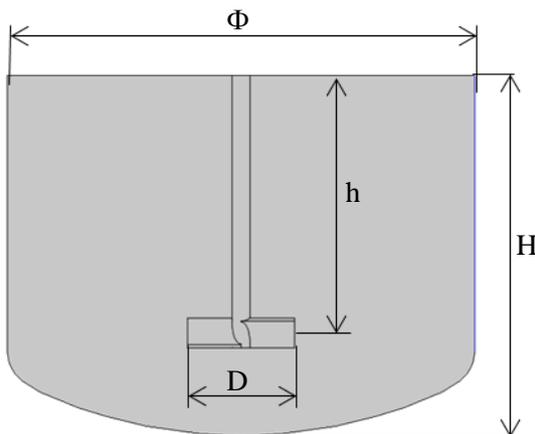


Figure 1. Schematic diagram of the ladle.

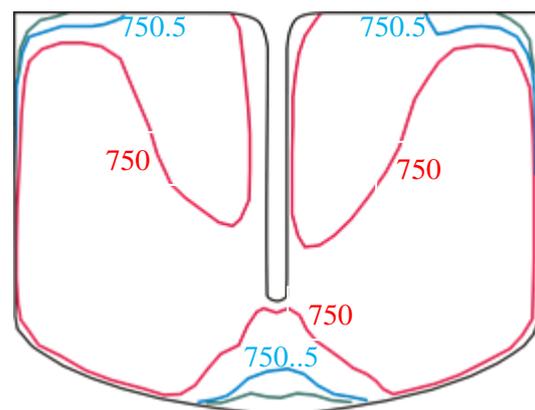


Figure 2. Temperature distribution, K.

Then the cap is mounted, and the melt agitation starts. During whole campaign the pot content is maintained at nearly constant temperature using gas burner system arranged around the external surface of the pot. The 3D model of the mixer ladle was created incorporating melt volume confined by the inner surface of the pot walls, surface of the agitator, surface of the shaft and free top surface of

the melt. The form and overall dimensions of the pot has been maintained constant in consecutive simulations, but the shape and immersion depth of the agitator has been modelled each time according to the guidelines from the analysis of previously obtained simulation results and predetermined objectives of the rotor design. The reconstruction procedure outlined above started from the agitator design, following the plant established practice (two rectangular blades mounted at acute angle slope on opposite sides of the shaft). Due to rotating shaft and agitator blades, the whole interior solution domain was divided into two parts: rotating domain encapsulating agitator end exterior domain around the pot walls. This was dictated by the solution procedure requirements and necessity of defining proper boundary conditions of the velocity field.

2.2. Physical properties

The temperature dependence of physical properties of molten lead has been used in the form of piecewise approximation. Initially conducted non-isothermal computations have revealed negligible temperature variation within the melt, therefore subsequent simulations were made for isothermal conditions at 750 K, at which the density of lead is $\rho=10480 \text{ kg/m}^3$ and molecular dynamic viscosity is $\mu=0.0018 \text{ Pa}\cdot\text{s}$.

2.3. Computational grid

The computational grid was created using tetrahedral elements, refined considerably near the surface of solid pot walls, the agitator blades and the shaft. An application of standard high Reynolds $k-\varepsilon$ turbulence model was taken into account by the incorporation of near the wall elements fulfilling the condition of first to the wall node distance, validating use of logarithmic velocity distribution as a boundary condition [7,8]. A posteriori inspection of dimensionless distance from the wall was made, resulting in the boundary grid refinement in the case of offence of mentioned above condition. Overall number of grid elements varies around 160 000 elements, depending on the stirrer shape.

2.4. Boundary conditions

At the solid walls of the pot and the propeller agitator, the wall function boundary condition has been applied. To resemble the actual free surface (top surface of the melt) the equilibration of normal stresses at the both sides of a surface were posed, taking into account surface tension of lead ($\sigma=0.35 \text{ N/m}$), and at every simulation time step the curvature of the surface was calculated and the computational grid reconstruction algorithm was used to follow new surface shape. Additional continuity conditions were used at common boundary of rotating domain and outer region of the melt.

2.5. Mathematical model specifications for the carrier fluid flow

The process of lead refining is conducted under nearly constant operating conditions, some minor variations of rotational frequency takes place during feeding of the refining compounds to get better conditions for the immersion of solid particles from the melt surface by means of the funnel formation at higher frequencies. This would lead to the conclusion that the steady state form of the mathematical model could be used, but the rotation of complex propeller agitator necessitates to apply unsteady form of the model equations.

Next step in the mathematical model development is to decide whether the flow is laminar or turbulent. For the mixing processes the criterion of the flow regime (turbulent or laminar) is specified by the critical value of Reynolds number defined as [1]:

$$\text{Re}_{\text{mixing}} = \left(\frac{\rho n d^2}{\mu} \right) \quad (2)$$

where n stands for rotational frequency 1/s of propeller agitator of diameter d . The molten lead is a very dense liquid with moderate viscosity, so one could expect domination of inertia forces over viscosity forces. In fact the value of $\text{Re} > 5 \times 10^6$ obtained even for moderate rotational frequency 80 rpm and adequate to operational conditions diameter d indicates turbulent motion of the melt.

Thus, assuming isothermal conditions, the flow of the melt has been considered incompressible and turbulent. The mathematical model consists of the set of equations:

- the continuity equation
- the momentum balance equation
- the turbulence closure model equations (high Reynolds number $k-\varepsilon$)
- the energy equation (conditionally included in the preliminary simulations)

These fundamental equations are not included here due to the volume of the text limitations as their form is obvious for all interested readers.

Preliminary computations were conducted for the non-isothermal conditions to estimate temperature variations in the melt and the importance of the natural convection for the circulation pattern. In this case an energy equation was appended to the model, and functional dependence on temperature of density and viscosity of the melt were used. Since the results of these non-isothermal simulations indicate significant alignment of the temperature in the boiler volume (figure 2), further simulations were conducted for isothermal conditions.

2.6. Transport of diluted refining compounds

The deterministic approach described above does not take into account the fact that the dispersed phase particles interacts with the carrier phase, which remains in the turbulent flow. Actually, in the momentum equation for the particle (1), the carrier phase instantaneous velocity should be used (instead of mean velocity value) to precisely describe the velocity and trajectory of a single particle. But, in such a formulation, all of the particles of the dispersed phase need to be traced. This would be very time consuming task and even impossible to accomplish in practice. Therefore, the concept of tracing “clouds” of particles proposed by Baxter and Smith [9] seems to be very promising and effective technique for the simulation of a diluted phase flow in turbulent conditions. This is called herein as stochastic-deterministic approach. Fundamental difference between strictly deterministic and stochastic-deterministic approach becomes visible in the description method of the spatial particles distribution. The stochastic-deterministic approach exploits the concept of mean trajectory (according to equation (1)) but it does interpret it as the trajectory of the particles ‘cloud’ centre rather than the single particle trajectory. The ensemble of particles with identical diameter, material and thermodynamic properties, called traced particles ‘cloud’, enters the model domain with the initial conditions defined by its initial spatial and time position and initial dispersion in space. The mean trajectory derived from the equation (1) using mean carrier phase velocity vector distribution is interpreted as the solution of ensemble average of equations of motion for the particles represented by the “cloud”. In this way the instantaneous particles distribution $g(x,y,z,t)$ is expressed by the statistical distribution function (3D probability density Gaussian distribution) about a mean trajectory

$$g(x, y, z, t) = \frac{\exp\left\{-\frac{1}{2}\left[\frac{(x - \xi_1(t))^2}{\sigma_{11}(t)} + \frac{(y - \xi_2(t))^2}{\sigma_{22}(t)} + \frac{(z - \xi_3(t))^2}{\sigma_{33}(t)}\right]\right\}}{(2\pi)^{3/2} \sigma_{11}(t) \sigma_{22}(t) \sigma_{33}(t)} \quad (3)$$

- $\xi_1(t), \xi_2(t), \xi_3(t)$ – position of the center of the multivariate distribution (average trajectory),
- $\sigma_{11}(t), \sigma_{22}(t), \sigma_{33}(t)$ – square roots of multivariate Gaussian distribution variances, where due to an isotropy of the $k-\varepsilon$ turbulence model of the carrier phase we use single value of the distribution variance $\sigma_{part}(t) = \sigma_{11}(t) = \sigma_{22}(t) = \sigma_{33}(t)$.

The variance $\sigma_{part}(t)$ of the postulated distribution, describing the spatial dimension of the cloud, should be estimated along the trajectory, as a cloud is expanding in time due to particles interaction with eddies of turbulent motion of the carrier phase. This can be approximated using Kampě de Feriět formula for the transverse diffusion in a turbulent flow [10]:

$$\sigma_{part}^2(t) = \sigma_{part}^2(t=0) + 2 \int_0^t \overline{v'(t_1)v'(t_1)} \int_0^{t_1} R_{part}(t_2) dt_2 dt_1 \quad (4)$$

where:

- $\sigma_{part}^2(t=0)$ - initial variance of the particle distribution for the traced “cloud”,
- $\overline{v'(t)v'(t)}$ - the mean square fluctuation component of the particle velocity,
- $R_{part}(t)$ - the auto-correlation Lagrange function of particles.

Direct application of the relationship equation (4) encounters basic difficulty in the fact that the parameters appearing under integral concern the parameters describing turbulent flow of the particles, and unless the turbulent motion of the particles is solved, these parameters are unknown. Therefore, the relation would be applicable for the strictly stochastic method of solution, what in fact we are going to avoid in the stochastic-deterministic approach using the ensemble average trajectory of the ‘cloud’ by integrating equation (1). The concept of the stochastic-deterministic approach is based on the approximation of equation (4), proposed by Baxter and Smith, who expressed the particle distribution variance using statistical parameters of the fluid turbulent motion in the vicinity of the particle. These parameters are available from the turbulent model applied for the gas phase flow. Interested readers are redirected to the original paper [9] for the details.

3. Solution procedure

Each simulation was executed starting from the propeller agitator acceleration period, when the rotational frequency was slowly raised to prevent computations divergence. After an accomplishment of desired value of rotation frequency computations were continued to attain steady values of several controlled parameters: propeller torque, pressure distribution on the free surface of the melt, maximum velocity magnitude, etc. It usually required relatively large number (dozens) of propeller turnovers to be simulated (namely 30 or even more). Then, the tracing of particle “clouds” were carried out to observe the dispersion of the particles in the bath.

Simulations carried out for a number of different agitators led to selecting the best of them. It proved to be a stirrer having 2 level sets of blades. The lower agitator with a diameter of D consists of three profiled blades twisted of 45° angle along its length. The blade base is mounted to the shaft at 45° angle to horizontal plane of rotation. The upper agitator of a similar design, but with a diameter of $0.75D$, was mounted on a shaft in a position rotated by 60° relatively to the lower agitator. The optimal distance between both agitators was established as $0.7D$. The level of a rotation plane of the lower agitator was set at $0.3H$ from the bottom of the ladle. Selection criterion for the positioning was to attain an appropriate depth of a sucking funnel formed on the surface of a bath in simulations.

4. Simulation results

The simulation results for optimal design of a stirrer are presented below. The velocity distribution of a carrier phase, shown for the central vertical cross section of a ladle is presented on figure 3, and for the horizontal cross section at the level of middle distance between propellers is presented on figure 4.

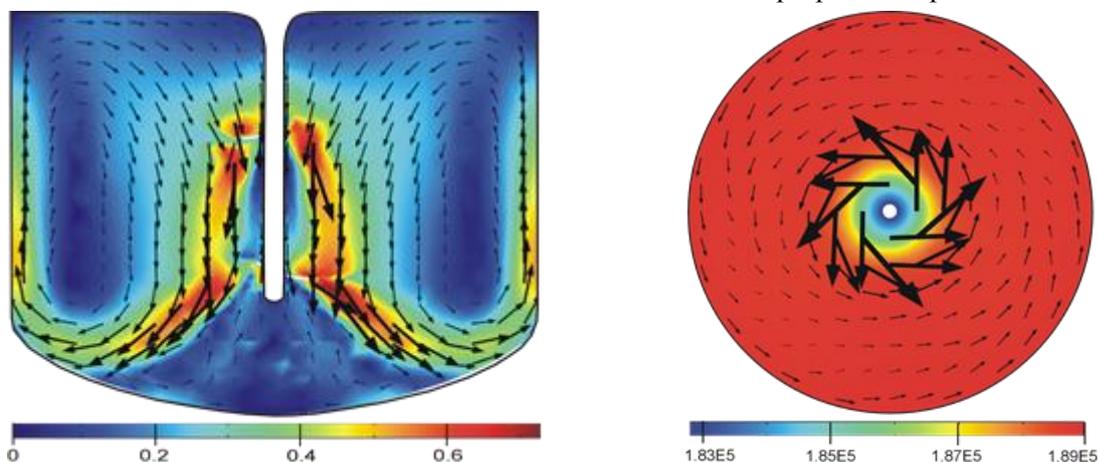


Figure 3. Velocity of carrier phase distribution, in plane components only, m/s.

Figure 4. Velocity of carrier phase (vectors), color map of pressure distribution, Pa.

The instantaneous streamlines are presented on figure 6, showing that there is no possibility to create two vortices in the flow, which suggested the literature of the subject, when double impeller is in use. It is interesting to observe nonhomogeneous distribution of turbulent kinetic energy (see figure 5), where its maximum values are distributed around the shaft over the upper agitator. This is very important observation, because an application of upper agitator not only creates a sucking funnel of required depth but also produce favorable conditions for rapid dispersion of refining compounds particles just at the beginning of their way through the base metal.

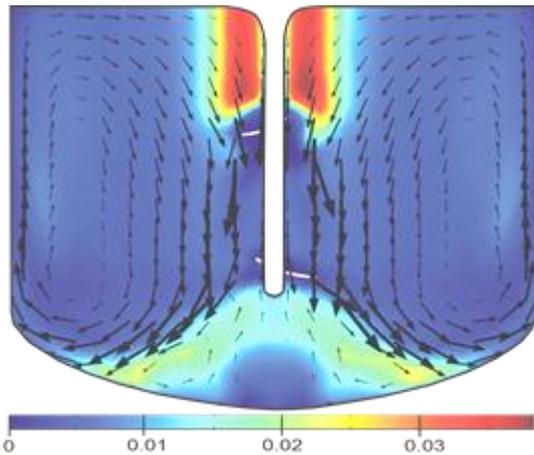


Figure 5. Turbulent kinetic energy distribution, m^2/s^2 .

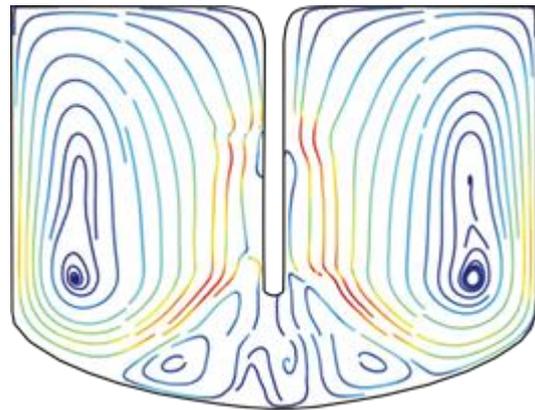


Figure 6. Instantaneous streamlines in a cross section of a mixer, warm colors – higher velocity.

On figure 7 we present the simulated evolution of the particles “clouds” along their way through the carrier fluid. A couple of “clouds” has been presented here for the clarity of the graph. It can be observed how the sucking funnel formed on the top of the bath pulls in particles, then disperses them gradually in the bath.

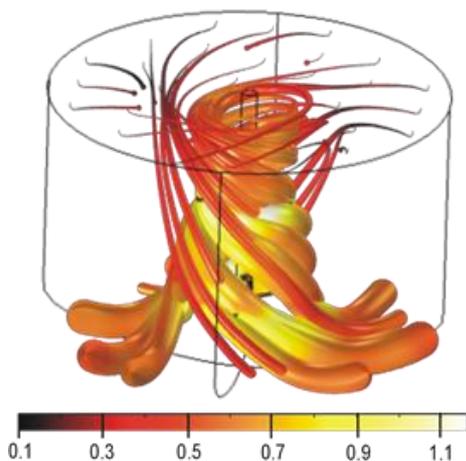


Figure 7. Refining particles “clouds” tracing (7sec since feeding), color express particles velocity m/s.

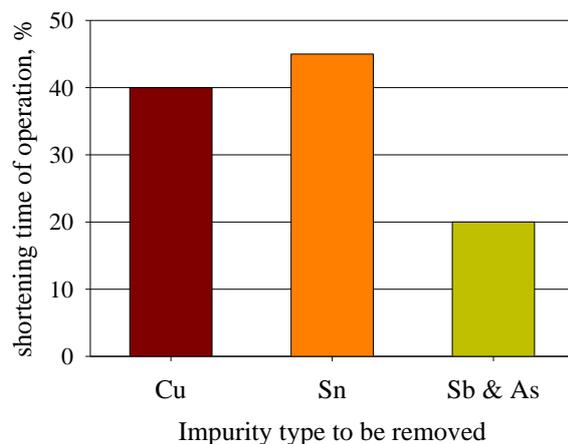


Figure 8. Time shortening of unit operation as a result of application of redesigned propeller for different impurities.

5. Verification

Direct verification of the simulation results, obtained by means of measurements of parameters of the simulated movement, seems to be virtually impossible due to the nature of the process and the type of carrier fluid. However, since the basic objectives of the utilitarian design of technological nature, it was decided to execute industrial trials that were supposed to answer the question of whether inflicted objectives have been obtained.

The stirrer of new design, consisting of doubled triple wing blades of different diameters has been produced from mild steel, using precision-casting technique. In order to verify the anticipated effects of application of a new agitator, a set of industrial test has been conducted. Trials were carried out in typical conditions for refining process before modification of the stirrer. Research objectives have been identified as: intensification of the process of refining, reducing energy consumption, reducing the formation of metal-oxide buildup at the side walls at the top of the boiler. Observations are subject to the following parameters: duration of the unit process in refining process to remove specific additives (copper, tin, antimony and arsenic), the final concentration of additives that are deleted, mass waste refining process. Process parameters observed have been confronted with the appropriate values on the record prior to the modification of the agitator. The most spectacular result of application of the new paddle was a significant time shortening of removing impurities. The percentage of time shortening for removing impurities after modification of the stirrer for each unit operation are shown on figure 8. An additional positive aspect of the intensification of the process is the weight reduction of the waste product: 26% for unit operation of removing tin and 10% for removing antimony and arsenic. In the case of removal of antimony and arsenic, there is also a deeper expulsion of impurities (60% less final concentration of impurities).

6. Conclusions

The presented results of the project show a great potential of numerical analysis of mixing processes. Precise design of agitator plays a decisive role in such processes, where intense dispersion of distributed phase is required for increasing the efficiency of the process. Obtained results of simulations have been confirmed by experiments carried out in industrial environments. Further subjects should take into account the analysis of chemical kinetics, which in the absence of specific studies could not be included in this work.

7. References

- [1] Kamieński J 2004 *Mieszanie układów wielofazowych* Warszawa: WNT)
- [2] Löhrner R 2001 *Applied CFD techniques* (Chichester: John Wiley & Sons)
- [3] Oran E, Boris J 1987 *Numerical simulation of reactive flow* (NY: Elsevier)
- [4] Stewart H, Wendroff B 1984 Two-phase flow: models and methods, *J. Comp. Phys.*, **56**, 263
- [5] Crowe C, Sommerfeld M, Tsuji Y 1988 *Multiphase flows with droplets and particles* (Boca Raton: CRC Press)
- [6] Crowe C, Sharma M, Stosk D 1977 The particle-source-in cell (PSI-CELL) model for gas - droplet flows *J. of Fluids Engineering* 329-332.
- [7] Launder B, Spalding D 1972 *Mathematical models of turbulence* (London: Academic Press)
- [8] Bennet C, Myers J 1962 *Momentum, Heat, and Mass Transfer* (NY: McGraw-Hill)
- [9] Baxter L, Smith P 1993 Turbulent dispersion of particles: the STP model *Energy&Fuels* **7** 852
- [10] Kampé de Fériet J 1939 Les fonctionés aleatoires stationnaires at la théorie statistique de la turbulence homogene *Ann. Soc. Sci. Bruxelles Ser. I* **59** 145-210.