UDK: 66.021.3-936:546.21-72

Milena KNEŽEVIĆ, Marija KICOŠEVIĆ, Milena STOJIČIĆ, Jana VIDAKOVIĆ, Dragan POVRENOVIĆ[1]

UTICAJ KARAKTERISTIKA ČESTICA I FLUIDA NA PRENOS MASE KISEONIKA IZ GASNE U TEČNU FAZU I DISTRIBUCIJU MEHUROVA U TROFAZNOM SISTEMU

INFLUENCE OF CHARACTERISTICS OF PARTICLES AND FLUID ON GAS-LIQUID OXYGEN MASS TRANSFER AND BUBBLE DISTRIBUTION IN THREE-PHASE SYSTEM

IZVOD

U ovom radu je proučavana distribucija mehurova i zapreminski koeficijent prenosa mase, k_La, u trofaznom sistemu, sa sferičnim česticama od stakla i keramike i nesferičnim česticama od keramike i zeolita. Eksperimenti su vršeni u cilindričnoj koloni prečnika 64 mm i visine 2000 mm i 2D koloni dimenzija 278 x 20,4 x 500 mm. Brzine gasa su se kretale u intervalu 0,03-0,09 m/s, a brzine tečnosti 0-0,1 m/s. Kao tečna faza korišćena je česmenska voda i rastvor amonijum hlorida različitih koncentracija. Rezultati su pokazali da prisustvo čestica u sistemu utiče na povećanje zapreminskog koeficijenta prenosa mase, ali koliko će biti to povećanje zavisi od karakteristika samih čestica. Takođe je pokazano da prisustvo rastvorenog amonijum hlorida u vodi znatno doprinosi smanjenju vrednosti k_la, u odnosu na vrednosti koje su dobijene u sistemu sa čistom vodom.

Ključne reči: zapreminski koeficijent prenosa mase, trofazni sistem, čestice od stakla, keramike i zeolita.

EXTRACT

This paper presents the study of bubble distribution and volumetric mass transfer coefficient, k_La, in a three-phase system with spherical particles of glass and ceramic and non-spherical particles of ceramic and zeolite. Experiments were carried out in a cylindrical column with 64 mm diameter and height of 2000 mm and 2D column (278 x 20.4 x 500 mm). Gas velocity varied in the range of 0.03 to 0.09 m/s and liquid velocity was 0-0.1 m/s. Tap water and ammonium chloride solution of different concentrations were used as the liquid phase. Results showed that the presence of particles in the system increases volumetric coefficient of mass transfer, but it depends on the characteristics of the particles. It was shown that the presence of dissolved ammonium chloride in water contributes significantly to the reduction of the k_la value, compared to values obtained in pure water system.

Key words: volumetric mass transfer coefficient; three-phase system; glass, ceramic and zeolite particles.

1. UVOD

Trofazni sistemi, gas-tečnost-čestice, imaju široku primenu kod operacija multifaznog kontakta u hemijskoj, petrohemijskoj, biohemijskoj industriji i metalurgiji. Ovi sistemi se sve više ispituju i koriste za mikrobiološke procese jer omogućavaju veliku specifičnu površinu za rast mikroorganizama kao i intezivan prenos mase. Zbog svojih pogodnosti trofazni sistemi se primenjuju i za aerobne i anaerobne procese prečišćavanja otpadnih voda. Široku primenu u prečišćavanju komunalnih otpadnih voda ima aerobna filtracija, kod koje su akteri prečišćavanja, mikroorganizmi imobilisani na inertnim nosačima. Međutim, zbog mogućnosti primene širokog opsega različitih inertnih nosača i mogućnosti u kombinovanju geometrije i fluido-mehanike sistema, može se doći do različitih izvedbi trofaznih sistema sa ciljem odvijanja što efikasnijeg procesa u njemu.

Da bi se postigao cilj procesa prečišćavanja, mikroorganizmima, odgovornim za uklanjanje biorazgradivih zagađujućih materija iz otpadne vode, je neophodno

1. INTRODUCTION

Three-phase systems, gas-liquid-particle, are widely used in operations of multiphase contact in the chemical, petrochemical, biochemical and metallurgical industry. These systems are increasingly investigated and used for microbiological processes because they provide a large specific surface area for microbial growth and intensive mass transfer. Due to its convenience three-phase systems are used for aerobic and anaerobic wastewater treatment. Aerobic filtration has wide application in the treatment of municipal wastewater, with microorganisms immobilized on inert carriers. However, because of the possibility of application of a wide range of different inert carriers and opportunities in combining geometry and fluid mechanics of the system, one can come up with different versions of three-phase system with the aim of designing process as efficient as possible.

In order to achieve the goal of treatment process it is necessary to provide the required operating conditions for micro-organisms responsible for the removal

1 Univerzitet u Beogradu, Tehnološko-metalurški fakultet, Karnegieva 4, Beograd, Srbija

obezbediti potrebne radne uslove. Limitirajući faktor u podešavanju optimalnih uslova sredine za rast mikroorganizama u aerobnim reaktorima je prenos mase kiseonika kao posledica slabog rastvaranja kiseonika u vodi i različitim fermentacionim medijumima. Efikasnost prenosa mase kiseonika iz gasne u tečnu fazu, se najbolje opisuje preko vrednosti zapreminskog koeficijenta prenosa mase, k_La, [1-3].

Izvršena ispitivanja trofaznih sistema sa pokretnim slojem čestica, su pokazala da zapreminski koeficijent prenosa mase kiseonika zavisi od distribucije i veličine mehurova u sistemu [7] kao i da ponašanje mehurova u sistemu, koje je uglavnom uslovljeno načinom disperzije, ima direktan uticaj na prenos mase i hidrodinamiku sistema [8]. Na prenos mase kiseonika, odnosno na distribuciju mehurova utiču distributor gasa, protok gasa, protok tečnosti, osobine tečnosti i osobine različitih tipova ispune [4,6,9-16].

Pregledom literature, može se uočiti dosta suprostavljenih rezultata i različitih tumačenja uticaja ispitivanih parametara na prenos mase kiseonika u trofaznim sistemima. Zbog toga je nemoguće osloniti se na postojeće analize, pa je neophodno izvršiti eksperimentalne provere za svaki ispitivani sistem sa ciljem detaljnijeg utvrđivanja interakcija čestica i mehurova vazduha u trofaznom sistemu.

Osnovni cilj ovog rada je ispitivanje mogućnosti pri-

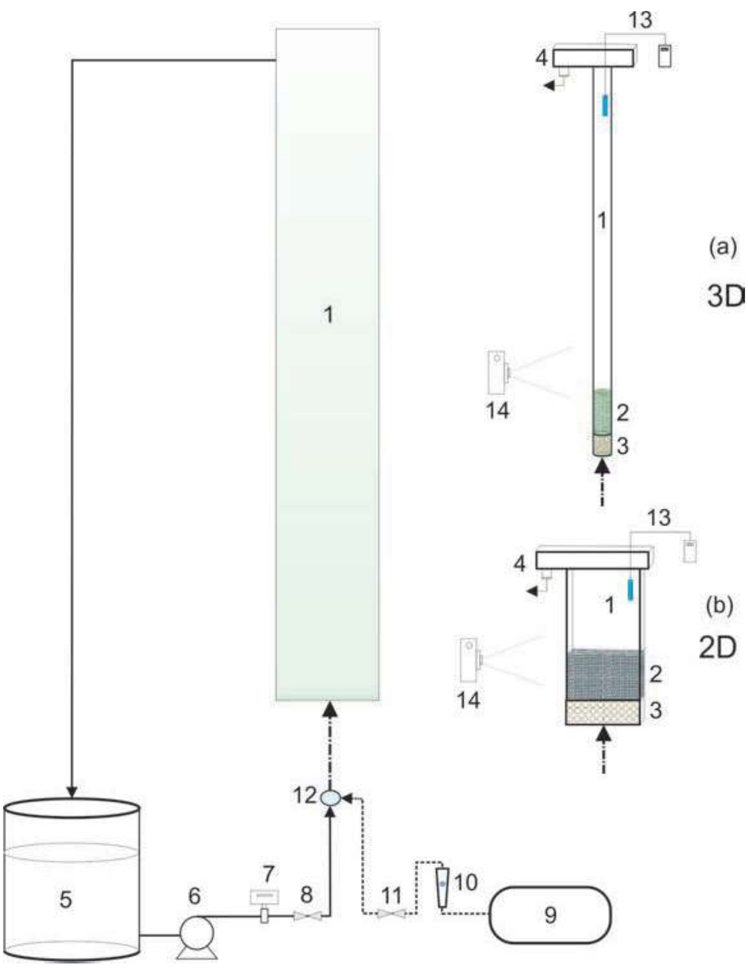
of biodegradable pollutants from wastewater. Limiting factor in setting the optimum operational conditions for the growth of microorganisms in the aerobic reactor is oxygen mass transfer as a result of low oxygen dissolution in water and various fermentation media. Efficiency of gas-liquid oxygen mass transfer is best described through the values of volumetric mass transfer coefficient k_La, [1-3].

Observed three-phase system with a movable layer of particles, showed that volumetric mass transfer coefficient of oxygen depends on the distribution and size of bubbles in the system [7], as well as the behavior of bubbles in the system, which is mainly conditioned by the dispersion, has a direct impact on the mass transfer and the hydrodynamics of the system [8]. Oxygen mass transfer and the distribution of bubbles are affected by gas distributor, gas flow, liquid flow, fluid properties and characteristics of different types of beds [4,6,9-16].

Reviewing the literature, it can be seen a lot of conflicting results and different interpretations of the examined parameters on oxygen mass transfer in three-phase systems. Therefore, it is impossible to rely on existing analysis, so it is necessary to carry out experimental testing of each test system with the aim of determining the detailed interactions of particles and air bubbles in three-phase system.

Main objective of this paper is to examine the application possibilities of three-phase systems for nitrification, as a first degree in the process of removing nitrogen compounds from wastewater. Therefore, it is necessary to understand the effect of fluid-mechanical characteristics of the oxygen mass transfer in these systems, especially the impact of different types of particles and the chemical composition of water on the oxygen mass transfer efficiency, i.e. the value of the volumetric mass transfer coefficient, and, k_La.

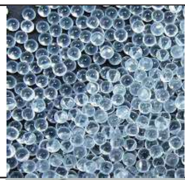
Limiting parameter of the nitrification pro-



Slika 1. Šeme eksperimentalnih aparatura: a) Cilindrična 3D kolona; b) 2D kolona. 1.kolona, 2.nasuti sloj čestica, 3.raspodeljivač, 4.prelivnik, 5.rezervoar za vodu, 6.pumpa, 7.elektromagnetni merač protoka vode, 8.ventil, 9.komprimovani vazduh, 10.rotametar, 11.ventil, 12.injektor vazduha, 13.oksimetar, 14.kamera.

Figure 1 Scheme of the experimental apparatus: a) 3D cylindrical column b) 2D column. 1. column, 2. Fluidized bed, 3. distributor, 4. spillway, 5. water tank, 6. pump, 7. electromagnetic flow meter, 8. valve, 9. compressed air, 10. rotameter, 11. valve, 12. air injector, 13. oximeter, 14. camera.

Tabela 1. Karakteristike čestica.
Table 1 Particle characteristics

Tip čestice / Type of particles	Prečnik / Diameter, mm	Gustina / Density, kg/m3	Izgled čestica Particles
Sferične staklene čestice Spherical glass particles	3	2500	
Sferične staklene čestice Spherical glass particles	6	2500	
Sferične keramičke čestice Spherical ceramic particles	6	2311	
Keramičke perle Ceramic beads	6,5	2500	
Zeolit Zeolite	<6	1915	

mene trofaznih sistema za nitrifikaciju, kao prvog stepena u procesu uklanjanja azotnih jedinjenja iz otpadne vode. Stoga je potrebno poznavati uticaj fluido-mehaničkih karakteristika na prenos mase kiseonika u ovim sistemima, a pre svega uticaj različitih tipova čestica i hemijskog sastava vode na efikasnost prenosa mase kiseonika, odnosno na vrednost zapreminskog koeficijenta prenosa mase, $k_L a$.

Limitirajući parametar procesa nitrifikacije je koncentracija rastvorenog kiseonika, odnosno prenos mase kiseonika na koga najveći uticaj imaju karakteristike čvrste faze. Pravilnim odabirom čvrste faze, odnosno inertih nosača za nitrifikacione bakterije, aktere nitrifikacionog procesa, može se uticati na povećanje efikasnosti procesa uklanjanja azota iz otpadnih voda, što i jeste konačni cilj ovih istraživanja.

2. EKSPERIMENTALNI DEO

Za eksperimentalna ispitivanja su se koristile dve kolone od pleksiglasa. Jedna kolona je bila 3D, visine 2000 mm i prečnika 64 mm, a druga kolona je bila 2D, dimenzija 278 x 20,4 i visine 500 mm. eksperimentalni sistem je prikazan na slici 1.

Na dno kolone uvođeni su vazduh i voda. Kod obe

cess is the concentration of dissolved oxygen and oxygen mass transfer, which are mostly affected by the characteristics of the solid phase. Proper selection of the solid phase or inert carrier of nitrification bacteria can cause an increase in the effectiveness of nitrogen removal from wastewater, which is the ultimate goal of this research.

2. EXPERIMENTAL PART

Two columns of Plexiglas were used for the experimental study. One column was 3D, height 2000 mm and a diameter of 64 mm, and the second column was 2D, with dimension of 278 x 20.4 mm and height of 500mm. Experimental system is shown in Figure 1.

Air and water were injected at the bottom of the column. In both columns common distributor of air and water was a packed layer of glass spherical particles with 4 mm diameter. Air velocity through the column ranged from 0.03 to 0.09 m/s, and the air flow was measured by rotameter. Water flow was measured by electromagnetic meter and the water velocity through the column ranged from 0 to 0.09 m/s. Glass, ceramics and zeolite particles were used, and their characteristics are shown in Table 1. Height of examined fluidized bed of particles amounted 10, 20, 30 and 40 cm.

kolone zaednički distributor za vazduh i vodu je predstavljao pakovani sloj staklenih sferičnih čestica prečnika 4 mm. Brzina vazduha kroz kolonu se kretala od 0,03 do 0,09 m/s, a protok vazduha je meren rotametrom. Protok vode je meren pomoću elektromagnetnog merača, a brzina vode kroz kolonu se kretala u intervalu od 0 do 0,09 m/s. Kao ispuna koristile su se čestice stakla, keramike i zeolita, a njihove karakteristike su prikazane u tabeli 1. Visina pakovanog sloja čestica u koloni, pri kojima su vršena ispitivanja su iznosila 10, 20, 30 i 40 cm.

Ispitivanja prenosa mase kiseonika su rađena sa česmenskom vodom, na sobnoj temperaturi, iz koje je pomoću natrijum sulfita prethodno uklanjan rastvoren kiseonik do koncentracija ispod 1 mg/l. Nakon uklanjanja kiseonika, u vodu je uvođen vazduh a promena koncentracije rastvorenog kiseonika u koloni sa vremenom merena je pomoću oksimetra, WTW Oxi 340i. Uvođenje vazduha je vršeno do postizanja najmanje 90% koncentracije zasićenja. Pod pretpostavkom da je u kolonama uspostavljeno idealno mešanje, za izračunavanje zapreminskog koeficijenta prenosa mase korišćena je jednačina 1:

$$\frac{dC}{dt} = k_L a(C^* - C_t) \qquad (1)$$

Takođe u eksperimentu je praćena distribucija i veličina mehurova pomoću analize slika. Na poleđinu kolona je bio zalepljen paus papir, na koji je usmeren reflektor, što je omogućilo jasnije uočavanje mehurova, a potom su prečnici mehurova na slikama određivani pomoću programa „Sigma Scan".

Eksperimenti su vršeni i sa vodom u koju je dodavan amonijum hlorid, koji u ovom slučaju simulira azotno zagađenje prisutno u otpadnim vodama, kako bi se ispitale i karakteristike tečnog medijuma na formiranje mehurova, odnosno prenos mase kiseonika u trofaznim sistemima.

3. REZULTATI I DISKUSIJA

Uticaj brzine gasa na $k_L a$

Uvođenjem vazduha u kolonu, bez protoka tečnosti (U_L=0), zapaža se da sa povećanjem protoka gasa raste i zapreminski koeficijent prenosa mase, slika 2.

Ovo se može pripisati povećanju obe veličine, specifčne površine a i koeficijenta prenosa mase k_L povećanjem protoka gasa. Velika brzina gasa, odnosno njegov fluks (m³/s*m²), utiče na povećanje „hold-up"-a gasa u sistemu, odnosno zapreminu zadržanog gasa u sistemu koja ima veliki uticaj na vrednost $k_L a$ jer je sa povećanjem zapremine zadržanog gasa omogućeno da veća količina kiseonika pređe iz gasne u tečnu fazu. Takođe, velika brzina gasa utiče na

Examinations of oxygen mass transfer were made with tap water, at room temperature, which was previously treated by sodium sulfite in order to remove dissolved oxygen to concentrations below 1 mg/l. After removing the oxygen, air was injected and a change of dissolved oxygen concentration in the column with time was measured using the oximeter, WTW Oxi 340i. Air injection was conducted to achieve at least 90% saturation. Assuming ideal mixing, equation 1was used to calculate the volumetric mass transfer coefficient:

$$\frac{dC}{dt} = k_L a(C^* - C_t) \qquad (1)$$

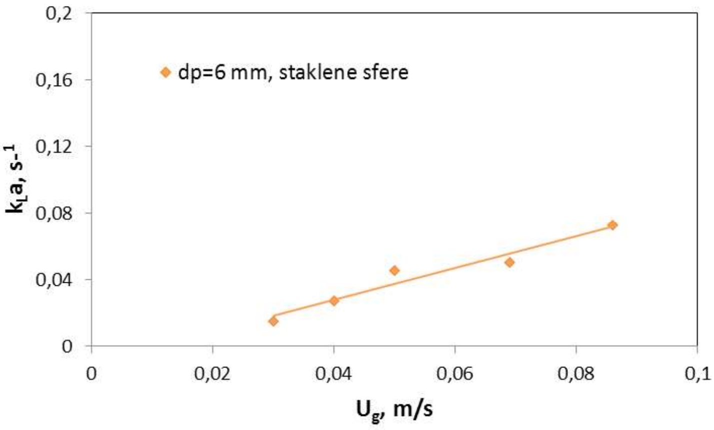
Experiment also included the monitoring of bubble distribution and size using image analysis. On the back of the column was taped tracing paper, which focuses on the projector, allowing clear identification of a bubble, and then the diameters of bubbles in the pictures were determined using Sigma Scan program.

Experiments were carried out with water to which was added ammonium chloride, which in this case simulates nitrogen pollution present in wastewater, in order to examine the characteristics of the liquid medium and the formation of bubbles and oxygen mass transfer in three-phase systems.

3. RESULTS AND DISCUSSION

Influence of gas velocity on $k_L a$

Injecting air into the column without liquid flow (U_L=0), it can be noted that the volumetric mass transfer coefficient increases with the gas flow, Figure 2.



***Slika 2.** Uticaj brzine gasa na zapreminski koeficijent prenosa mase u sistemu sa staklenim sferama prečnika 6 mm, (UL=0 m/s, H_{sloja}=30 cm)*

***Figure 2** Influence of gas velocity on the volumetric mass transfer coefficient in the system with glass spheres of 6 mm diameter (UL=0 m/s, H_{layar}=30 cm)* smanjenje prečnika mehura, što dovodi do povećanja međufazne površine, a time i do boljeg prenosa mase, [9]. Da povećanje brzine gasa utiče na povećanje „hold-up"-a gasa i k_la potvrđeno je i od strane drugih istraživača koji su ispitivali i druge tipove čestica [4,6,9,16,17,18].

Porast brzine gasa u trofaznom sistemu dovodi do drobljenja mehurova i smanjenja njihovog prečnika, pa iako bi se očekivao suprotan efekat, da sa smanjenjem prečnika mehura njegova brzina dizanja opada, usled povećanja turbulencije dolazi do povećanja brzine dizanja mehura u koloni, a time i povećanja turbulencije tečnosti oko mehurova. U tom slučaju otpor prenosu mase od strane filma tečnosti opada, što utiče na povećanje vrednosti koeficijenta prenosa mase, k_L, a što je i eksperimentalno potvrđeno u rezultatima [9].

Uticaj brzine tečnosti na k_La

Uvođenjem tečnosti u sistem dolazi do povećanja turbulencije u njemu, koja doprinosi da film tečnosti postane tanji a otpor prenosu kiseonika manji, što utiče na povećanje koeficijenta prenosa mase od strane tečnosti k_L. Na slici 3. je prikazan uticaj brzine tečnosti na zapreminski koeficijent prenosa mase.

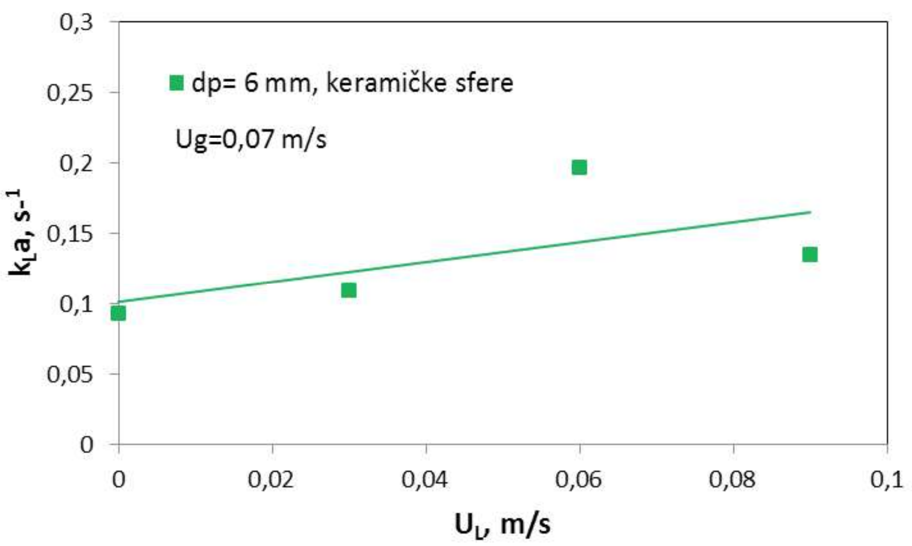
Povećanje brzine proticanja tečnosti u koloni, odnosto povećanje turbulencije, takođe doprinosi cepanju mehurova i smanjivanju njihovog prečnika što doprinosi povećanju specifične površine za prenos mase kiseonika iz gasne u tečnu fazu. U cilju određivanja dimenzija mehorova, praćena je promena u broju i veličini mehurova u 2D koloni (sl. 1b), snimanjem po-

This can be attributed to the increase in specific surface area a and mass transfer coefficient k_L with increasing gas flow. High gas velocity and its flux (m3/s•m2), increases the gas "hold-up" in the system, i.e. the volume of gas retained in the system, which has a major impact on the value of k_la, because the increase of retained gas volume enables the greater oxygen transfer from the gas to the liquid phase. Also, high gas velocity impacts on reducing gas bubble diameter, which leads to greater interfacial area, and hence to better mass transfer [9]. Increase of the gas velocity impacts the greater gas "hold-up" and k_la, was confirmed by other researchers who have examined other types of particles [4,6,9,16,17,18].

Increase in gas velocity in three-phase system leads to crushing bubbles and reduction of their diameter, and although we expected the opposite effect that with a reduction in the diameter of the bubble its speed decreases, due to the increased turbulence it leads to an increase in the ascending velocity of the bubble in the column, and thus increases the turbulence around the bubbles. In this case, the resistance to mass transfer of the liquid decreases, which increases the mass transfer coefficient, k_l, which was experimentally confirmed by the results [9].

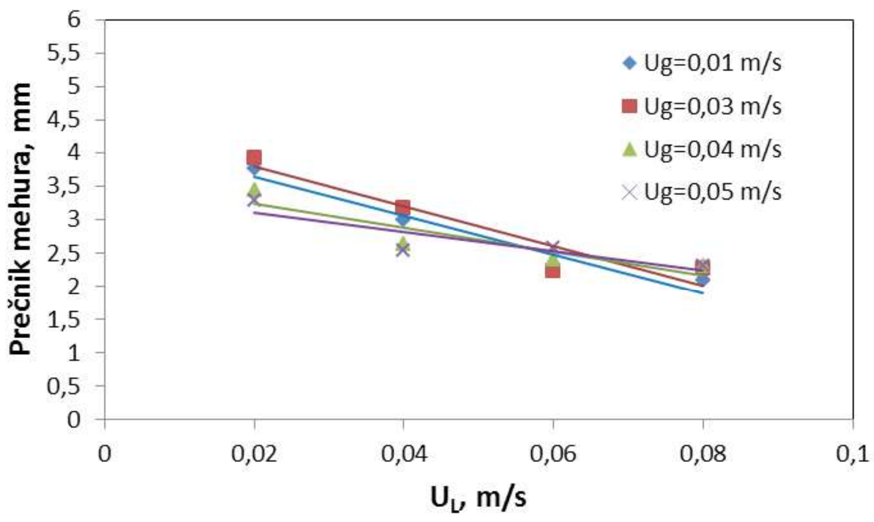
Influence of liquid velocity on k_La

Injection of liquid into the system leads to an increase of turbulence in it, which contributes to the thinner liquid film and less oxygen transfer resistance, which increases the mass transfer coefficient of the liquid k_L. Figure 3 shows the effect of liquid velocity on the volumetric mass transfer coefficient.



***Slika 3.** Uticaj brzine tečnosti na zapreminski koeficijent prenosa mase u sistemu sa keramičkim sferama prečnika 6 mm, (Ug=0,07 m/s, H_{sloja}=40 cm)*
***Figure 3** Influence of the liquid on the volumetric mass transfer coefficient in the system of ceramic spheres with a 6 mm diameter (Ug=0,07 m/s, H_{layar}=40 cm)*

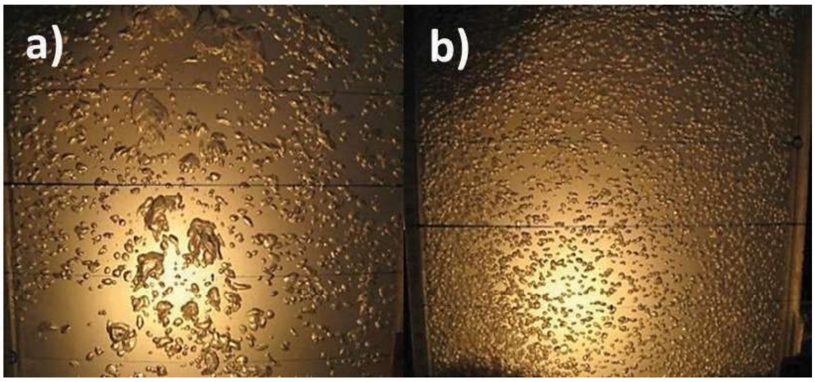
Increasing liquid flow in the column, i.e. the increasing turbulence, also contributes to fragmentation of bubbles and reduction of their diameter, which contributes to the increase of specific surface area for mass transfer of oxygen from the gas to the liquid phase. In order to determine the bubble size, changes in the number and size of bubbles in a 2D column (Fig. 1b) were monitored by recording camera, after which the resulting images were scanned and analyzed using "SigmaScan" software package for image analysis. Figure 4 shows the dependence of the diameter of the bubble and liquid velocity under certain operating conditions.



Slika 4. Uticaj brzine tečnosti na prečnik mehura, pri konstantnim brzinama gasa u sistemu sa staklenim kuglicama prečnika 6 mm.
Figure 4 Influence of the liquid on the bubble diameter, at constant gas speed in the system with glass beads with a 6 mm diameter.

Higher value of liquid velocity contributes to the formation of a large number of more uniformed bubbles (homogeneous bubble flow regime), while at lower values of liquid velocity their coalescence is emphasized, which leads to the formation of heterogeneous bubble flow regime, as shown in Figure 5.

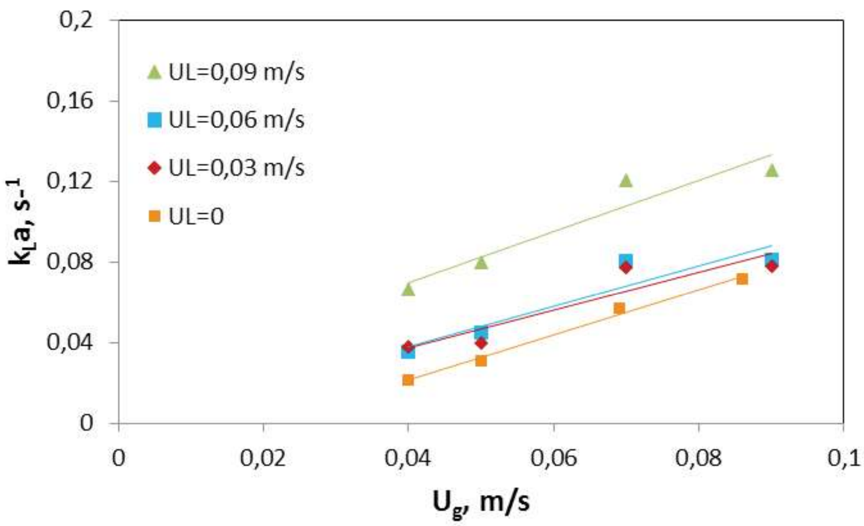
Results of performed experiments, as shown in Figure 6, show synergistic effect of air and water flow on oxygen mass transfer. Figure shows that the value of the volume coefficient increases with greater velocity of both fluids.



Slika 5. Veličina mehurova u koloni sa keramičkim česticama od 6 mm pri istoj brzini gasa a pri različitim brzinama tečnosti od a) 0,02 i b) 0,1m/s.
Figure 5 Bubbles size in the column with ceramic particles of 6 mm diameter at the same gas velocity and different liquid velocities of a) 0.02 b) 0.1 m/s.

Influence of particles characteristics on k_La

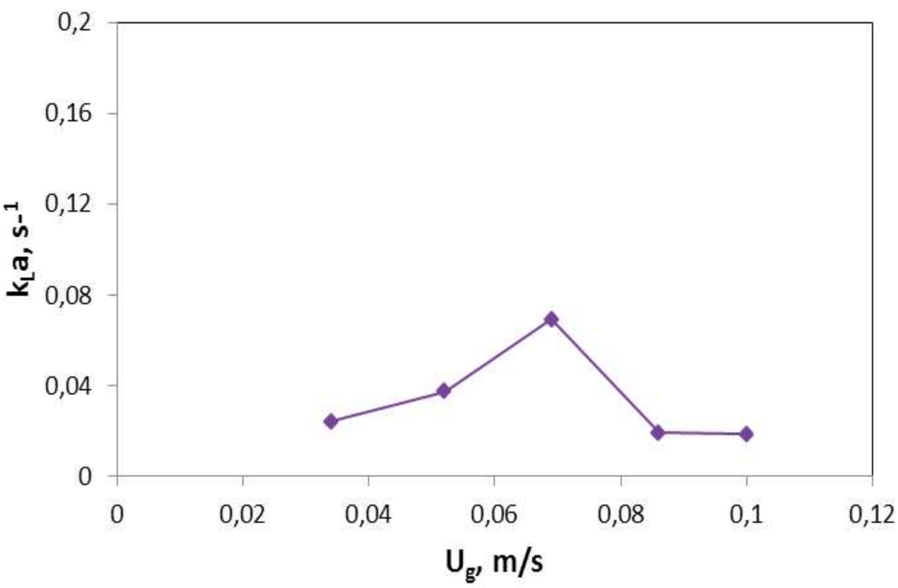
In two-phase systems in the absence of particles, the better mass transfer of oxygen is achieved at low gas velocities, when the so-called homogeneous bubble flow regime is formed, while at higher speeds there is coalescence, the merger of smaller into larger bubbles, [7]. This was confirmed by the results of our experiments performed on two-phase system, without the presence of particles, shown in Figure 7. It shows that with the increase of gas flow, the value of volumetric mass transfer coefficient increases to the certain value of gas flow, after which there is a decline in its value. Reason is related to the formation of coalescence bubbles, which reduces the interfacial area for transfer, at high gas flow rates.



Slika 6. Uticaj brzine gasa i tečnosti na zapreminski koeficijent prenosa mase u sistemu sa staklenim sferama *dp=6 mm*
Figure 6 Influence of the gas and the liquid velocity on volumetric mass transfer coefficient in the system with glass spheres *dp=6 mm*

However, a large amount of air is necessary in aerobic microbial processes due to the promoću kamere, nakon čega su dobijene slike skenirane i analizirane u u softverskom paketu za analizu slika „SigmaScan". Na slici 4 prikazana je zavisnost prečnika mehura od brzine proticanja tečnosti pri određenim operativnim uslovima.

Veća vrednost brzine tečnosti doprinosi formiranju većeg broja uniformnijih mehurova (homogen režim strujanja mehurova), dok je pri manjim vrednostima brzine tečnosti izraženije njihovo srastanje, pa dolazi do formiranja heterogenog režima proticanja mehurova, što se vidi na slici 5.



Slika 7. Dvofazni sistem. Uticaj brzine gasa na zapreminski koeficijent prenosa mase (U_L=0)

Figure 7 Two-phase system. Influence of gas velocity on the volumetric mass transfer coefficient (U_L=0)

Rezultati izvršenih eksperimenata, prikazanih na slici 6, pokazuju sinergetsko delovanje protoka vazduha i vode na prenos mase kiseonika. Sa slike se vidi da sa povećanjem brzine proticanja oba fluida, vrednost zapreminskog koeficijenta raste.

Uticaj karakteristika čestica na k_La

U dvofaznim sistemima bez prisustva čestica, bolji prenos mase kiseonika, se postiže pri nižim brzinama gasa, kada se obrazuje tzv. homogeni režim protoka mehura, dok pri većim brzinama dolazi do koalescencije, spajanja sitnijih u krupnije mehurove, [7]. Ovo su potvrdili i rezultati naših eksperimenata vršenih u dvofaznom sistemu, bez prisustva čestica, prikazani na slici 7., gde se vidi da sa povećanjem protoka gasa, vrednost zapreminskog koeficijenta prenosa mase raste do određenje vrednosti protoka gasa, nakon koje dolazi do opadanja njegove vrednosti. Razlog je u formiranju povezanih koalescentnih mehurova, koji doprinose smanjenju međufazne površine za prenos, pri velikim protocima gasa.

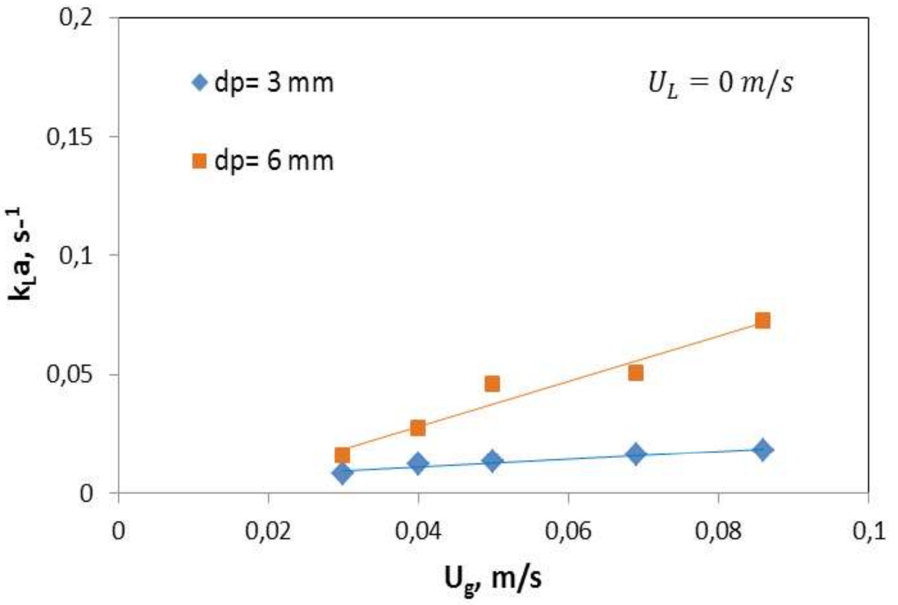
Međutim velika količina vazduha je neophodna u aerobnim mikrobiološkim procesima zbog presudne uloge rastvorenog kiseonika na efikasnost odvijanja procesa. Istraživanja su pokazala, da odgovarajuća prisutna čvrsta faza u sistemu ima mogućnost da pri visokim protocima gasa utiče na formiranje homogenog režima mehurova. Na osnovu ovoga je istaknuto da važnu ulogu na formiranje mehurova imaju osobine čestica koje čine ispunu. Uticaj osobina čestica potrebno je detaljno ispitati jer, pri određenim uslovima, čestice mogu da suzbiju koalescenciju, ali isto tako, u drugom slučaju, mogu i da je pospeše.

minent role of dissolved oxygen on the process efficiency. Studies have shown that the corresponding solid phases present in the system has the ability to influence the formation of homogeneous bubble regime at high gas flows. Based on this, it was pointed out that the characteristics of the particles have an important role in the formation of bubbles. It is necessary to examine in detail the influence of particles because, under certain conditions, particles can suppress coalescence, but also in other, they can enhance it.

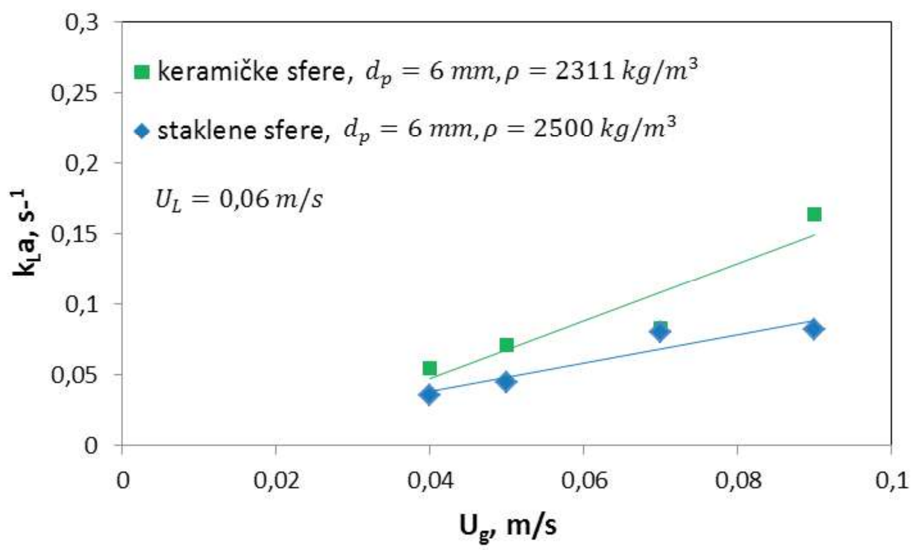
Diameter of the particle affects the efficiency of oxygen mass transfer in the system. With increasing particle diameter increases the amount of gas retained in the column, which is consistent with published results [6,10,19,20]. Fine particles are increasing coalescence, compared to systems with larger particles. At the same level of the fluidized bed and the same values of fluid flow, with smaller particles increased pressure drop causes the compression of air in the bottom layer, which causes the coalescence of bubbles and thus the air "by-pass" through the layer. Large bubbles have a lower specific surface area, greater ascending velocity and the shorter retention time, which leads to reduced mass transfer. Increasing diameter of the particles forces improved tearing of bubbles, which leads to an increase in the gas content in the column as well as an increase in the value of volumetric mass transfer coefficient, k_La, [6,11,12]. Influence of particle diameter on volumetric mass transfer coefficient in the system with glass spheres of 3 mm and 6 mm diameter, without fluid flow is shown in Figure 8. Same results were obtained in a system where there was a constant liquid flow.

Another important feature of a particle that affects the

Jedna od osobina čestica koja utiče na efikasnost prenosa mase kiseonika u sistemu je njihov prečnik. Sa porastom prečnika čestica raste količina zadržanog gasa u koloni, što je u skladu sa literaturnim rezultatima [6,10,19,20]. Sitne čestice utiču na povećanje koalescencije, u odnosu na sisteme sa krupnijim česticama. Pri istoj visini nasutog sloja i istim vrednostima protoka fluida, kod sitnijih čestica povećani pad pritiska uslovljava kompresiju vazduha na dnu sloja, što izaziva srastanje mehurova, a time i „by pass" vazduha kroz nasut sloj. Veliki mehurovi imaju manju specifičnu površinu, veće brzine dizanja kroz kolonu i kraće vreme zadržavanja, što dovodi do smanjenog prenosa mase. Povećanje prečnika čestica utiče na bolje cepanje muhurova, što dovodi do povećanja sadržaja gasa u koloni kao i do povećanja vrednosti zapreminskog koeficijenta prenosa mase, $k_L a$, [6,11,12]. Uticaj prečnika čestica na zapreminski koeficijent prenosa mase u sistemu sa staklenim sferama prečnika 3 i 6 mm, bez proticanja tečnosti, prikazan je na slici 8. Isti rezultati su dobijeni i u sistemu kada je postojao protok tečnosti.



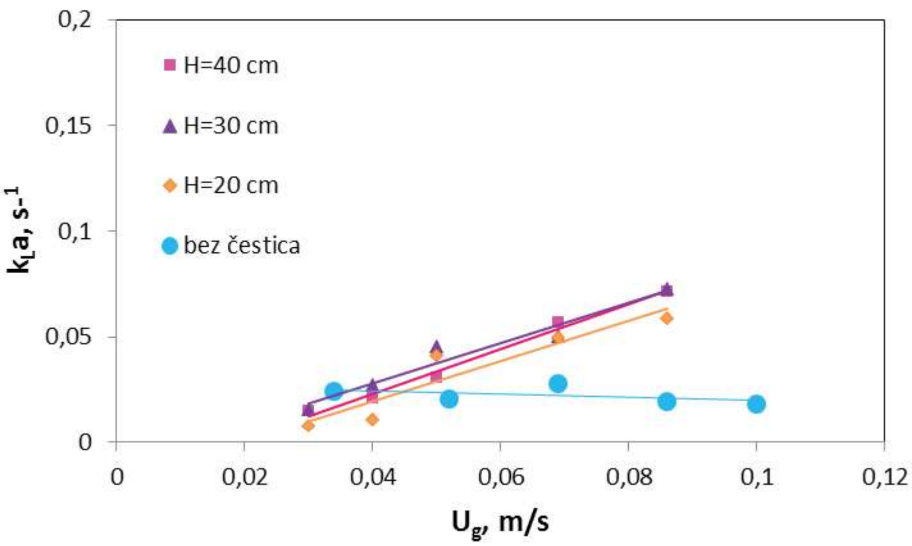
Slika 8. Uticaj prečnika staklenih čestica na $k_L a$ u zavisnosti od U_g, U_L=0 m/s, H_{Layer}=30 cm
Figure 8 Influence of the glass particle diameter on the $k_L a$ depending on U_g, U_L=0 m/s, H_{Layer}=30 cm.



Slika 9. Uticaj gustine čestice na zapreminski koeficijent prenosa mase.
Figure 9 Influence of particle density on the volumetric mass transfer coefficient

Druga bitna osobina čestica koja utiče na efikasnost prenosa mase kisenika je gustina čestica. Rezultati eksperimentalnih ispitivanja, prikazani na slici 9, pokazuju da su vrednosti zapreminskog koeficijenta prenosa mase veće u sistemu sa keramičkim sferama prečnika 6 mm i gustine $2311\ \mathrm{kg/m^3}$2311 kg/m³ od vrednosti dobijenih u sistemu sa staklenim sferama istog prečnika, 6 mm a gustine $2500\ \mathrm{kg/m^3}$2500 kg/m³, pod istim operativnim uslovima. Ovako mala razlika u gustinama čestica dovela je do značajne razlike u vrednostma $k_L a$ $k_L a$ u ova dva ispitivana sistema. Freitas i Teixeira [13] koji su ispitivali čestice prečnika 2,1 mm, gustine 1023 i $1048\ \mathrm{kg/m^3}$1048 kg/m³, došli su do istog zaključka, da mala razlika u gustini čestica ima uticaj na vrednosti zapreminskog koeficijenta prenosa mase.

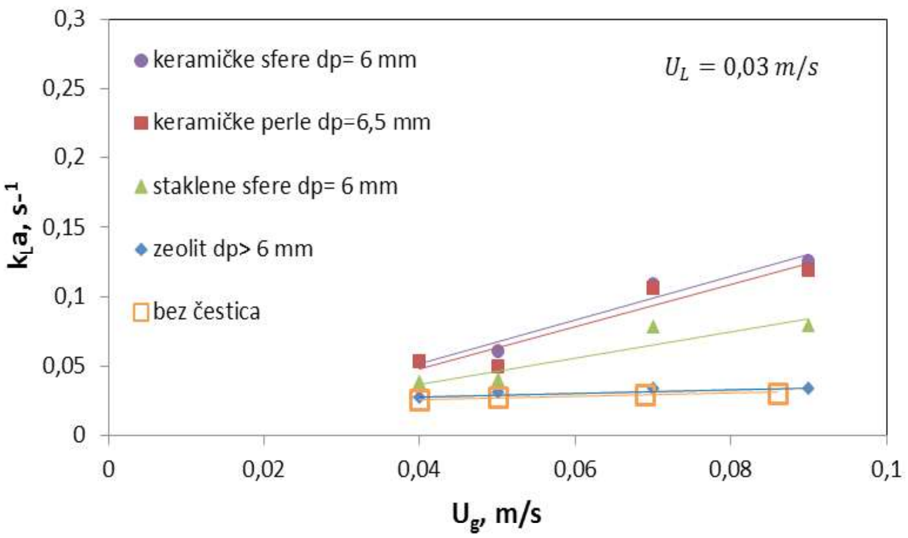
efficiency of oxygen mass transfer is the density of particles. Obtained experimental results, shown in Figure 9, show greater values of volumetric mass transfer coefficient in the system with ceramic spheres with a diameter of 6 mm and a density of 2311 kg/m3, from the values obtained in the system with glass spheres of the same diameter, 6 mm and density of 2500 kg/m3, under the same operating conditions. Such a small difference in particle densities led to significant differences in $k_L a$ values in the two examined systems. Freitas and Teixeira [13] who investigated the particle diameter of 2.1 mm and density of 1023 and 1048 kg/m3, came to the same conclusion, that the small differences in the density of the particles affects the values



Slika 10. Uticaj visine pakovanog sloja čestica na zapreminski koeficijent prenosa mase ($U_L = 0$)

Figure 10 Influence of fluidized bed height of particles on the volumetric mass transfer coefficient ($U_L = 0$

Pored prečnika i gustine čestica, na veličinu i distribuciju mehura u sistemu utiče i količina čestica u sistemu, odnosno visina pakovanog sloja čestica u koloni. Rezultati prikazani na slici 10, pokazuju da vrednost koeficijenta prenosa mase raste sa porastom visine sloja. Razlog je što mehur od svog „izvora" krećući se naviše kroz kolonu, sa povećanjem visine sloja čestica, nailazi na sve više prepreka, odnosno čestica koje utiču na njegov oblik i dimenzije.



Slika 11. Poređenje uticaja ispitivanih čestica na vrednost zapreminskog koeficijenta prenosa mase, pri određenim operativnim uslovima.

Figure 11 Comparison of the influence of examined particles on the value of volumetric mass transfer coefficient, under certain operating conditions.

U eksperimentima pored ispitivanja uticaja sferičnih čestica stakla i keramike na formiranje mehurova i njihovu distribuciju, ispitivale su se i čestice nesferičnog oblika, keramičke perle i zeolit, (tabela 1). U ovakvim sistemima uočena su nepredvidiva ponašanja pakovanog sloja čestica. U zavisnosti od vrednosti protoka fluida nesferične čestice su imale mogućnost da

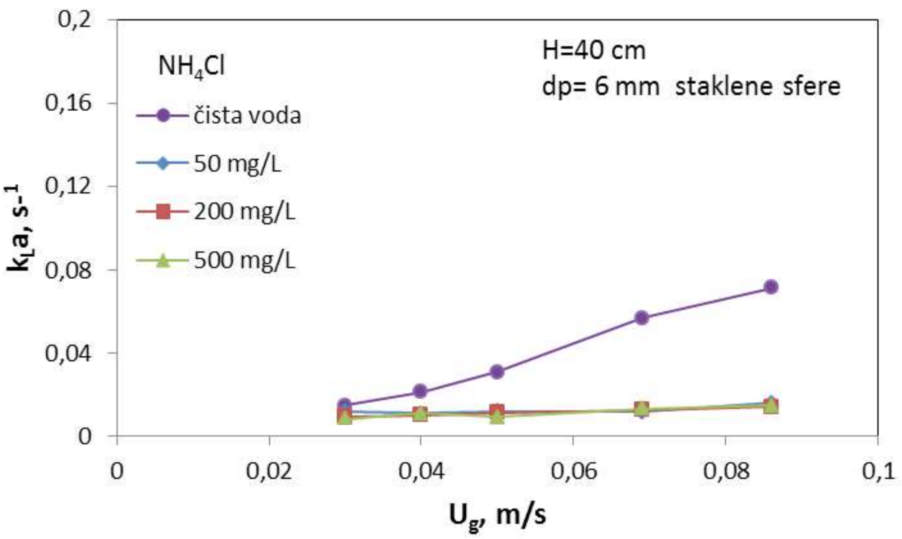
of volumetric mass transfer coefficient

Besides the diameter and density of the particles, the size and distribution of bubbles in the system is affected by the amount of particles, or the height of packed layer in the column. Results shown in Figure 10, show that the mass transfer coefficient increases with increasing height of the layer. This occurs because bubble moving upward from its "source" through the column faces more barriers or particles that affect the shape and dimensions.

This study, beside the examination of the influence of spherical particles of glass and ceramic on the formation of bubbles and their distribution, included the examination of non-spherical particles, ceramic beads and zeolite (Table 1). These systems showed unpredictable behavior of particles. Depending on the flow values non-spherical particles were able to form a non-uniform packed bed that allows fluid to flow in a stream. On the other hand, the results show that under certain operating conditions better values of volumetric coefficient of mass transfer in the system are achieved with ceramic beads than in a system with spherical glass particles.

Figure 11 shows that the lowest values of volumetric mass transfer coefficient are obtained in the system with zeolite. This can be attributed to the polydispersity of zeolite particles and their non-uniform shape, which lead them to orient particles in a fluid to provide minimum resistance to its flowing. In this flow reformiraju neuniforman pakovan sloj koji omogućava fluidima da protiču u obliku mlazeva. Sa druge strane, rezultati pokazuju da se pod određenim operativnim uslovima ostvaruje bolja vrednost zapreminskog koeficijenta prenosa mase u sistemu sa keramičkim perlama nego u sistemu sa staklenim sferičnim česticama.

Sa slike 11. se vidi da su najniže vrednosti zapreminskog koeficijenta prenosa mase dobijene u sistemu sa zeolitom. Ovo se može pripisati polidisperznosti čestica zeolita kao i njihovom neuniformnom obliku, koji dovode do toga da se čestice orjentišu u fluidu tako da pružaju najmanji otpor njegovom proticanju. Pri ovakvom režimu proticanja mehurovi se slabije drobe, što kao posledicu ima lošiji prenos mase kiseonika iz gasne u tečnu fazu.



Slika 12. Uticaj rastvorenog amonijum hlorida na vrednost zapreminskiog koeficijenta prenosa mase u trofaznom sistemu, sa staklenim česticama prečnika 6 mm.
Figure 12 Influence of dissolved ammonium chloride on the value of volumetric mass transfer coefficient in three-phase system, with glass particles of 6 mm diameter.

Uticaj karakteristika tečnosti na k_La

Poznato je da prisustvo rastvorenih i suspendovanih materija u vodi negativno utiče na koncentraciju rastvorenog kiseonika u njoj, a izražajnost ovog negativnog efekekta zavisi od sastava i koncentracija prisutnih materija u vodi.

Da bi se simuliralo amonijačno zagađenje u otpadnim vodama, u našim eksperimentima je u česmensku vodu dodavan amonijum hlorid. Uticaj rastvorenog amonijum hlorida u vodi na vrednost zapreminskog koeficijenta prenosa mase, prikazan je na slici 12, sa koje se vidi da vrednosti k_La značajno opadaju u prisustvu rastvorenog amonijum hlorida, u odnosu na vrednosti dobijene za čistu vodu, a da sa porastom koncentracije NH_4Cl on neznatno nastavlja da opada.

Pored uticaja karakteristika čvrste faze, potrebno je poznavati i druge karakteristike tečne faze, poput površinskog napona i viskoziteta koje takođe značajno utiču na prenos mase kiseonika u trofaznom sistemu, pa je za svaki konkretan slučaj potrebno uraditi nova istraživanja.

4. ZAKLJUČAK

U ovom radu je ispitivan uticaj karakteristika različitih tipova čestica na distribuciju mehurova i zapreminski koeficijent prenosa mase u trofaznom sistemu. Takođe su se poredile vrednosti zapreminskog koefi-

gime, bubbles are less crushed, which has the effect of reduced mass transfer of oxygen from the gas to the liquid phase.

Influence of liquid characteristics on k_La

It is known that the presence of dissolved and suspended solids in the water affects the concentration of dissolved oxygen in it, and expression of this negative effect depends on the composition and concentration of substances present in the water.

In order to simulate ammonia pollution in wastewater, in our experiments ammonium chloride was added into tap water. Influence of dissolved ammonium chloride on the value of the volumetric mass transfer coefficient is shown in Figure 12, and it can be concluded that the k_La values have a significant decrease in the presence of dissolved ammonium chloride, compared to the values obtained for pure water, and that it continues to decline slightly with the increase of NH_4Cl concentration.

Besides the impact of the solid phase characteristics, it is necessary to know the other characteristics of the liquid phase, such as surface tension and viscosity, which also significantly affect the oxygen mass transfer in three-phase system, and each individual case demands new research.

4. CONCLUSION

This paper presents the effect of the characteristics of different types of particles on the bubble distribution and volumetric mass transfer coefficient in three-phase system. It compared the values of volumetric mass transfer coefficient obtained in the three-phase cijenta prenosa mase dobijene u trofaznom sistemu sa čistom vodom i u sistemu sa rastvorom amonijum hlorida različitih koncentracija.

Pokazano je da prisustvo čvrste faze u sistemu doprinosi znatnom povećanju vrednosti zapreminskog koeficijenta prenosa mase u odnosu na vrednosti koeficijenta koje su dobijene u dvofaznom sistemu. Koliko će ono biti zavisi od karakteristika samih čestica, prečnika, gustine, oblika i njihove količine u sistemu. Sa povećanjem prečnika čestice raste i zapreminski koeficijent prenosa mase. Poređenjem čestica istog prečnika uočeno je da i malo smanjenje u gustini čestice utiče na povećanje k_La. Na prenos mase kiseonika utiče i ponašanje pakovanog sloja nesferičnih čestica koje je u direktnoj vezi sa oblikom čestica i režimom strujanja fluida. Zbog nepredvidivog načina pakovanja, nesferične čestice mogu da utiču na poboljšanje prenosa mase kiseonika ali i na njegovo smanjenje u trofaznom sistemu. Povećanje brzine vazduha i tečnosti doprinose povećanju prenosa mase kiseonika iz gasovite u tečnu fazu. Pokazano je i da pored karakteristika čestica na vrednost zapreminskog koeficijenta prenosa mase značajno utiču i karakteristike tečne faze. Značajno smanjenje vrednosti zapreminskog koeficijenta prenosa mase je dobijeno su sistemu sa rastvorom amonijum hlorida u odnosu na vrednosti dobijene u sistemu sa čistom vodom.

Dobijeni rezultati predstavljaju osnov za dalja istraživanja procesa nitrifikacije u trofaznim disperznim sistemima i razvoja novog tipa reaktora za prečišćavanje otpadnih voda.

Zahvalnica

Ovaj rad je urađen u okviru projekta OI-172022, finansiranog od strane Ministarstva prosvete, nauke i tehnološkog razvoja Republike Srbije.

system with clean water and the system with a solution of ammonium chloride of various concentrations.

It has been shown that the presence of the solid phase in the system contributes to a significant increase in the value of volumetric mass transfer coefficient compared to the values of the coefficient obtained in the two-phase system. Amount of increase depend on the characteristics of the particles diameter, density, shape and their quantity in the system. Increase in diameter of the particles increases the volumetric mass transfer coefficient. Comparing the same particle diameter showed that a small reduction in the density of particles increases the k_La. Oxygen mass transfer is affected by the behavior of packed layer of non-spherical particles which is directly related to the shape of the particles and fluid flow regime. Due to the unpredictable behavior of non-spherical particles they can affect the improvement of the mass transfer of oxygen, but also its reduction in a three-phase system. Increasing air and liquid velocity contribute to the increase of oxygen mass transfer from the gaseous to the liquid phase. It was shown that the characteristics of the liquid phase significantly affect the value of volumetric mass transfer coefficient, in addition to particle characteristics. Significant reduction of the volumetric mass transfer coefficient was obtained by the system with a solution of ammonium chloride in relation to the values obtained in pure water.

Obtained results represent the basis for further study of the nitrification process in three-phase disperse systems and development of a new type of reactor for wastewater treatment.

Acknowledgements

This paper was done as part of the OI-172022 project, funded by the Ministry of Education, Science and Technological Development of the Republic of Serbia.

Simboli / Symbols

k_La zapreminski koeficijent prenosa mase, s^{-1}

C^* koncentracija zasićenja kiseonikom, mg/l

C_t koncentracija rastvorenog kiseonika u vremenu t, mg/l

d_p prečnik čestice, mm

gustina čestice, kg/m^3

U_g brzina gasa kroz kolonu, m/s

U_L brzina tečnosti kroz kolonu, m/s

H visina nasutog sloja, m

LITERATURE

1. M.Y. Chisti, Airlift Bioreactors, Elsevier Applied Science, London, UK, 198.
2. J.C. Merchuk, M.H. Siegel, Airlift reactors in chemical and biological technology, J. Chem. Technol. Biotechnol. 41 (1988) 105–120.
3. F. Benyhaia, L. Jones, D. Plantaz, Mass transfer studies in pneumatic reactors, Chem. Eng. Technol. 19 (1996) 425–431.
4. Chia-Min Chen, Lii-Ping Leu, A highly elevated mass transfer rate process for three-phase, liquid-continuous fluidized beds, Chemical Engineering Journal 81 (2001) 223–230.
5. Marupatch Jamnongwong, Karine Loubiere, Nicolas Dietrich, Gilles Hébrard, Experimental study of oxygen diffusion coefficients in clean water containingsalt, glucose or surfactant: Consequences on the liquid-side mass transfer coefficients, Chemical Engineering Journal 165 (2010) 758–768.
6. Hideki Miura, Takashi Katoh, Yoshinori Kawase, Gas–liquid mass transfer in co-current three-phase fluidized beds with non-Newtonian fluids: Theoretical models based on the energy dissipation rate, Chemical Engineering Journal 185–186 (2012) 337–346.
7. E. Camarasa, C. Vial, S. Poncin, G. Wild, N. Midoux, J. Bouillard, Influence of coalescence behaviour of the liquid and of gas sparging on hydrodynamics and bubble characteristics in a bubble column, Chemical Engineering and Processing 38 (1999) 329–344.
8. W.-D. Deckwer, A. Schumpe, Improved tools for bubble column reactors design and scale-up, Chem. Eng. Sci. 48 (1993) 889–911.
9. Weiguo Yang, Jinfu Wang, Tiefeng Wang, Yong Jin, Experimental study on gas–liquid interfacial area and mass transfer coefficient in three-phase circulating fluidized beds, Chemical Engineering Journal 84 (2001) 485–490.
10. D.H. Lee, J.O. Kim, S.D. Kim, Mass transfer and phase holdup characteristics in three-phase fluidized beds, Chem. Eng. Commun. 119 (1993) 179–196.
11. A. Schumpe, W.-D. Deckwer, K.D.P. Nigam, Gas–liquid mass transfer in threephase fluidized beds with viscous pseudoplastic liquids, Can. J. Chem. Eng. 67 (1989) 873–877.
12. V. Sivasubramanian, Gas–liquid mass transfer in three-phase inverse fluidized bed reactor with Newtonian and non-Newtonian fluids, Asia-Pacific J. Chem. Eng. 5 (2010) 361–368.
13. C. Freitas, J.A. Teixeira, Oxygen mass transfer in a high solids loading three-phase internal-loop airlift reactor, Chemical Engineering Journal 84 (2001) 57–6.
14. M. Herskowitz, J.C. Merchuk, A loop three-phase fluidized bed reactor, Can. J. Chem. Eng. 64 (1986) 57–61.
15. D.B. Bukur, S.A. Patel, J.G. Daly, Gas holdup and solids dispersion in a threephase slurry bubble column, AIChE J. 36 (1990) 1731–1735.
16. Imran Hamdad, Shahrzad Hashemi, Dano Rossi, Arturo Macchi,Oxygen transfer and hydrodynamics in three-phase inverse fluidized beds, Chemical Engineering Science 62 (2007) 7399 – 7405.
17. Y.X. Guo, M.N. Rathor, H.C. Ti, Hydrodynamics and mass transfer studies in a novel external-loop airlift reactor, Chemical Engineering Journal 67 (1997) 205–214.
18. Jung Suk Lee, Hae Ryong Jin, Ho Lim, Dae Ho Lim, Yong Kang, Sang Done Kim, Ki Won Jun, Interfacial area and liquid-side and overall mass transfer coefficients in a three-phase circulating fluidized bed, Chem. Eng. Sci. (2013), http://dx.doi.org/10.1016/j.ces.2013.02.012i
19. H.M. Jena, B.K. Sahoo, G.K. Roy, B.C. Meikap, Characterization of hydrodynamic properties of a gas–liquid–solid three-phase fluidized bed with regular shape spherical glass bead particles, Chem. Eng. J. 145 (2008) 50–56.
20. H.M. Jena, B.K. Sahoo, G.K. Roy, B.C. Meikap, Characterization of hydrodynamic properties of a gas–liquid–solid three-phase fluidized bed with regular shape spherical glass bead particles, Chemical Engineering Journal 145 (2008) 50–56.