A R C H I V E S O F M E T A L L U R G Y A N D M A T E R I A L S
Volume 59 2014 Issue 1
DOI: 10.2478/amm-2014-0046

B. OLEKSIAK*, J. ŁABAJ*, J. WIECZOREK*, A. BLACHA-GRZECHNIK**, R. BURDZIK***

# SURFACE TENSION OF Cu – Bi ALLOYS AND WETTABILITY IN A LIQUID ALLOY – REFRACTORY MATERIAL – GASEOUS PHASE SYSTEM

# NAPIĘCIE POWIERZCHNIOWE STOPÓW Cu – Bi ORAZ ZWILŻALNOŚĆ W UKŁADZIE: CIEKŁY STOP – MATERIAŁ OGNIOTRWAŁY – FAZA GAZOWA

The study involved measurements of surface tension of liquid binary copper-bismuth alloys with respect to their chemical composition and temperature as well as investigations of the liquid alloy – refractory material – gaseous phase system wettability using usual refractory materials, i.e. aluminium oxide, magnesium oxide and graphite. The experiments were performed with the use of sessile drop method and a high-temperature microscope coupled with a camera and a computer was utilised.
*Keywords*: surface tension, contact angle, wettability, sessile drop method, high-temperature microscope, liquid Cu-Bi alloys

W ramach pracy przeprowadzone zostały pomiary napięcia powierzchniowego ciekłych dwuskładnikowych stopów miedzi z bizmutem, w funkcji składu chemicznego i temperatury oraz badania zwilżalności w układzie: ciekły stop- materiał ogniotrwały-faza gazowa z wykorzystaniem typowych materiałów ogniotrwałych, to jest tlenku glinu, tlenku magnezu oraz grafitu. Badania wykonano metodą kropli leżącej, przy użyciu mikroskopu wysokotemperaturowego sprzężonego z kamerą i komputerem.

## 1. Introduction

Surface tension of liquid metals and alloys as well as wettability on a liquid-solid phase contact surface affect, among others, courses of phenomena at interfaces during pyrometallurgical processes of metal production and refinement as well as during casting processes. In addition, an important factor here is presence of surface-active substances in metals and alloys. These properties are observed for several elements, e.g. oxygen, sulphur, lead, antimony or selenium. Surface-active substances accumulate at interfaces and most commonly affect surface tension and contact angle values. For pyrometallurgical processes, they can influence their rates, usually through inhibition; however, there are cases when they increase rates of processes at interfaces. Surface tension and wettability also highly affect refractory material corrosion as well as they are important factors for processes of metal coating application, casting and composite material production [1-10]. At present, when metallurgical and casting modelling as well as material property modelling become increasingly popular, such parameters as surface tension, contact angles, density or viscosity of the liquid phase are of particular importance [11-15].

The study involved measurements of surface tension of liquid binary copper-bismuth alloys with respect to their chemical composition and temperature as well as investigations of the liquid alloy – refractory material – gaseous phase system wettability using usual refractory materials, i.e. aluminium oxide, magnesium oxide and graphite.

## 2. Methodology

For the investigations of surface tension and wettability, copper-bismuth alloys, containing 1, 2, 3, 4, 5, 10, 20, 30, 40, 50 %mass Bi, were used. Samples of the above alloys were prepared to form cylinders of approximately 4 mm in diameter and height. For surface tension measurements, the samples were placed on 14 mm x 9 mm $Al_2O_3$ plates in the working chamber of a measuring device. For wettability investigations, MgO and graphite plates were also applied. As protective atmosphere during the experiments, argon 6.0 was utilised.

The investigations of surface tension and wettability were performed using the sessile drop method. A high-temperature microscope coupled with a camera and a computer was applied. The sample images were displayed on the screen and recorded by the computer, which ensured that measurements of relevant geometrical parameters of the liquid metal or alloy drop and determination of surface tension and contact angle

* SILESIAN UNIVERSITY OF TECHNOLOGY, FACULTY OF MATERIALS ENGINEERING AND METALLURGY, 40-019 KATOWICE, 8 KRASINSKIEGO STR., POLAND
** SILESIAN UNIVERSITY OF TECHNOLOGY, FACULTY OF CHEMISTRY, 44-100 GLIWICE, 9 STRZODY STR., POLAND
*** SILESIAN UNIVERSITY OF TECHNOLOGY, FACULTY OF TRANSPORT, 40-019 KATOWICE, 8 KRASINSKIEGO STR., POLAND

could be subsequently performed. A detailed description of the measuring equipment and measurement methods is presented in several already published papers [1, 2, 14-17].

The surface tension and wettability investigations were conducted at 1373, 1423, 1473, 1523 and 1573 K.

## 3. Study results

Results of the surface tension measurements for liquid copper-bismuth alloys of maximum 50 %mass Bi content are presented in Table 1. Results of the contact angle measurements for the liquid alloy – refractory material – gaseous phase system are presented in Tables 2-4.

TABLE 1

Results of the liquid Cu-Bi alloy surface tension measurements

| Bi content, %mass | Temperature, K | Density, $kg \cdot m^{-3}$ | Surface tension, $mN \cdot m^{-1}$ |
|---|---|---|---|
| 1 | 2 | 3 | 4 |
| 1 | 1373 | 8004 | 1295 |
| | 1423 | 7954 | 1285 |
| | 1473 | 7904 | 1274 |
| | 1523 | 7854 | 1268 |
| | 1573 | 7803 | 1255 |
| 2 | 1373 | 8008 | 1193 |
| | 1423 | 7956 | 1186 |
| | 1473 | 7906 | 1178 |
| | 1523 | 7858 | 1133 |
| | 1573 | 7808 | 1125 |
| 3 | 1373 | 8012 | 1107 |
| | 1423 | 7962 | 1099 |
| | 1473 | 7912 | 1086 |
| | 1523 | 7862 | 1071 |
| | 1573 | 7812 | 1059 |
| 4 | 1373 | 8016 | 994 |
| | 1423 | 7956 | 967 |
| | 1473 | 7916 | 961 |
| | 1523 | 7866 | 953 |
| | 1573 | 7816 | 936 |
| 5 | 1373 | 8021 | 791 |
| | 1423 | 7971 | 786 |
| | 1473 | 7920 | 774 |
| | 1523 | 7870 | 769 |
| | 1573 | 7820 | 773 |
| 10 | 1373 | 8044 | 738 |
| | 1423 | 7993 | 731 |
| | 1473 | 7943 | 722 |
| | 1523 | 7893 | 718 |
| | 1573 | 7842 | 713 |
| 20 | 1373 | 8095 | 670 |
| | 1423 | 8044 | 660 |
| | 1473 | 7993 | 616 |
| | 1523 | 7943 | 608 |
| | 1573 | 7892 | 606 |

cd. TABLE 1

| 1 | 2 | 3 | 4 |
|---|---|---|---|
| 30 | 1373 | 8156 | 481 |
| | 1423 | 8104 | 466 |
| | 1473 | 8053 | 459 |
| | 1523 | 8001 | 449 |
| | 1573 | 7950 | 432 |
| 40 | 1373 | 8228 | 454 |
| | 1423 | 8176 | 440 |
| | 1473 | 8124 | 444 |
| | 1523 | 8072 | 432 |
| | 1573 | 8019 | 430 |
| 50 | 1373 | 8315 | 453 |
| | 1423 | 8263 | 437 |
| | 1473 | 8210 | 420 |
| | 1523 | 8157 | 420 |
| | 1573 | 8104 | 415 |

TABLE 2

Results of the contact angle measurements for the liquid copper-bismuth alloy – aluminium oxide – gaseous phase system

| Bi content, %mass | Temperature, K | Contact angle, $^o$ |
|---|---|---|
| 1 | 2 | 3 |
| 1 | 1373 | 140 |
| | 1423 | 139 |
| | 1473 | 139 |
| | 1523 | 138 |
| | 1573 | 138 |
| 2 | 1373 | 139 |
| | 1423 | 138 |
| | 1473 | 137 |
| | 1523 | 137 |
| | 1573 | 136 |
| 3 | 1373 | 137 |
| | 1423 | 136 |
| | 1473 | 136 |
| | 1523 | 136 |
| | 1573 | 135 |
| 4 | 1373 | 137 |
| | 1423 | 136 |
| | 1473 | 136 |
| | 1523 | 136 |
| | 1573 | 135 |
| 5 | 1373 | 136 |
| | 1423 | 135 |
| | 1473 | 135 |
| | 1523 | 135 |
| | 1573 | 135 |
| 10 | 1373 | 134 |
| | 1423 | 134 |
| | 1473 | 133 |
| | 1523 | 132 |
| | 1573 | 132 |

cd. TABLE 2

| 1 | 2 | 3 |
|---|---|---|
| 20 | 1373 | 134 |
| | 1423 | 133 |
| | 1473 | 133 |
| | 1523 | 132 |
| | 1573 | 131 |
| 30 | 1373 | 134 |
| | 1423 | 134 |
| | 1473 | 134 |
| | 1523 | 133 |
| | 1573 | 132 |
| 40 | 1373 | 125 |
| | 1423 | 123 |
| | 1473 | 122 |
| | 1523 | 120 |
| | 1573 | 119 |
| 50 | 1373 | 123 |
| | 1423 | 121 |
| | 1473 | 120 |
| | 1523 | 118 |
| | 1573 | 116 |

TABLE 3

Results of the contact angle measurements for the liquid copper-bismuth alloy – magnesium oxide – gaseous phase system

| Bi content, %mass | Temperature, K | Contact angle, $^o$ |
|---|---|---|
| 1 | 2 | 3 |
| 1 | 1373 | 125 |
| | 1423 | 125 |
| | 1473 | 124 |
| | 1523 | 124 |
| | 1573 | 125 |
| 2 | 1373 | 123 |
| | 1423 | 123 |
| | 1473 | 122 |
| | 1523 | 120 |
| | 1573 | 120 |
| 3 | 1373 | 123 |
| | 1423 | 123 |
| | 1473 | 123 |
| | 1523 | 122 |
| | 1573 | 122 |
| 4 | 1373 | 123 |
| | 1423 | 122 |
| | 1473 | 122 |
| | 1523 | 121 |
| | 1573 | 120 |
| 5 | 1373 | 123 |
| | 1423 | 123 |
| | 1473 | 122 |
| | 1523 | 120 |
| | 1573 | 120 |

cd. TABLE 3

| 1 | 2 | 3 |
|---|---|---|
| 10 | 1373 | 123 |
| | 1423 | 123 |
| | 1473 | 123 |
| | 1523 | 122 |
| | 1573 | 122 |
| 20 | 1373 | 122 |
| | 1423 | 122 |
| | 1473 | 122 |
| | 1523 | 122 |
| | 1573 | 121 |
| 30 | 1373 | 122 |
| | 1423 | 122 |
| | 1473 | 121 |
| | 1523 | 121 |
| | 1573 | 121 |
| 40 | 1373 | 120 |
| | 1423 | 120 |
| | 1473 | 120 |
| | 1523 | 119 |
| | 1573 | 119 |
| 50 | 1373 | 119 |
| | 1423 | 119 |
| | 1473 | 118 |
| | 1523 | 117 |
| | 1573 | 117 |

TABLE 4

Results of the contact angle measurements for the liquid copper-bismuth alloy – graphite – gaseous phase system

| Bi content, %mass | Temperature, K | Contact angle, $^o$ |
|---|---|---|
| 1 | 2 | 3 |
| 1 | 1373 | 140 |
| | 1423 | 138 |
| | 1473 | 138 |
| | 1523 | 137 |
| | 1573 | 137 |
| 2 | 1373 | 141 |
| | 1423 | 141 |
| | 1473 | 140 |
| | 1523 | 140 |
| | 1573 | 140 |
| 3 | 1373 | 140 |
| | 1423 | 139 |
| | 1473 | 140 |
| | 1523 | 140 |
| | 1573 | 139 |
| 4 | 1373 | 138 |
| | 1423 | 138 |
| | 1473 | 138 |
| | 1523 | 138 |
| | 1573 | 138 |

cd. TABLE 4

| 1 | 2 | 3 |
|---|---|---|
| 5 | 1373 | 135 |
| | 1423 | 134 |
| | 1473 | 133 |
| | 1523 | 133 |
| | 1573 | 133 |
| 10 | 1373 | 135 |
| | 1423 | 135 |
| | 1473 | 132 |
| | 1523 | 132 |
| | 1573 | 132 |
| 20 | 1373 | 127 |
| | 1423 | 125 |
| | 1473 | 123 |
| | 1523 | 122 |
| | 1573 | 122 |
| 30 | 1373 | 120 |
| | 1423 | 120 |
| | 1473 | 120 |
| | 1523 | 119 |
| | 1573 | 118 |
| 40 | 1373 | 119 |
| | 1423 | 119 |
| | 1473 | 119 |
| | 1523 | 119 |
| | 1573 | 119 |
| 50 | 1373 | 118 |
| | 1423 | 118 |
| | 1473 | 116 |
| | 1523 | 115 |
| | 1573 | 114 |

## 4. Summary

The results of measurements of liquid Cu-Bi alloy surface tension have revealed a significant effect of the bismuth additive on the parameter values. In copper, bismuth acts as a surface-active substance like e.g. oxygen, sulphur or lead. A slightly increased Bi fraction in the Cu-Bi alloy leads to a markedly reduced surface tension, which is illustrated by the Cu-Bi surface tension values in Table 1. When the bismuth fraction in the alloy is 40 %mass and higher, the surface tension is near the value for pure bismuth. This may suggest accumulation of Bi at the liquid alloy-gaseous phase interface as for other known surface-active substances. The temperature rise causes a linear decrease in the surface tension of Cu-Bi alloys.

The wettability investigations for the liquid copper-bismuth alloy – refractory material – gaseous phase system show that this element causes reduction in the values of contact angles at $Al_2O_3$, MgO and graphite surfaces. For these alloys, the effect of temperature rise on the above material wettability improvement is also observed.

While analysing the results of wettability studies on liquid metal (alloy) – solid metal oxide obtained by Najdicz [5] and considering his wettability theory, it can be assumed that the investigated Cu-Bi alloys, when in contact with other thermodynamically stable oxides (e.g. CaO, $ZrO_2$, $SiO_2$), will show a similar behaviour to that observed for aluminium and magnesium oxides.

**Acknowledgements**

The study was conducted under the Research Project No. N N508 583639, financed by the Ministry of Science and Higher Education – Poland.

REFERENCES

[1] G. Siwiec, B. Oleksiak, A. Smalcerz, J. Wieczorek, Surface tension of Cu-Ag alloys, Archives of Metallurgy and Materials **58** (1), 193-195 (2013).
[2] G. Siwiec, B. Oleksiak, P. Folęga, T. Matuła, Wettability in the liquid Cu-Ag alloy – fireproof material – gas phase system, Metalurgija **52** (3), 334-336 (2013).
[3] J. Łabaj, G. Siwiec, B. Oleksiak, Surface tension of expanded slag from steel manufacturing in electrical furnace, Metalurgija **50** (3), 209-211 (2011).
[4] L. Blacha, G. Siwiec, B. Oleksiak, Loss of aluminium during the process of Ti-Al-V alloy smelting in a vacuum induction melting (VIM) furnace, Metalurgija **52** (3), 301-304 (2013).
[5] J.W. Najdicz, Kontaktnyie javlienija v mietallurgiczeskich rasplavach, Naukowa Dumka, Kijów (1971).
[6] L. Blacha, Untersuchungen der antimonentfernungsgeschwindigkeit aus blisterkupfer im prozess der vakuumraffination, Archives of Metallurgy and Materials **50** (4), 989-1002 (2005).
[7] L. Blacha, R. Burdzik, A. Smalcerz, T. Matuła, Effects of pressure on the kinetics of manganase evaporation from the OT 4 alloy, Archives of Metallurgy and Materials **58** (1), 197-201 (2013).
[8] A. Lisiecki, Welding of titanium alloy by Disk laser, Laser Technology 2012: Applications of Lasers, Proceedings of SPIE 8703 (2013).
[9] T. Węgrzyn, The influence of nickel and nitrogen on impact toughness properties of low alloy basic electrode steel deposits, 11th International Offshore and Polar Engineering Conference (ISOPE-2001) Stavanger, Norway (2001).
[10] J. Łabaj, G. Siwiec, L. Blacha, R. Burdzik, The role of a gas phase in the evaporation process of volatile components of the metal bath, Metalurgija **53** (2), 215-217 (2014).
[11] S. Amore, J. Brillo, I. Egry, R. Novakowic, Surface tension of liquid Cu–Ti binary alloys measured by electromagnetic levitation and thermodynamic modeling, Applied Surface Science **257**, 7739-7745 (2011).
[12] P. Fima, K. Wiater, M. Kucharski, The surface tension and density of the Cu-Ag-In alloys, Surface Science **601** (16), 3481-3487 (2007).
[13] M. Kucharski, P. Fima, The surface tension and density of liquid Ag-Bi, Ag-Sn, and Bi-Sn alloys, Monatshefte fur Chemie **136** (11), 1841-1846 (2005).
[14] G. Siwiec, J. Botor, M. Kucharski, Modeling and eksperimental measurements of surface tensions of Cu-Pb-Fe alloys, Archives of Metallurgy and Materials **54** (4), 1167-1172 (2009).

[15] J. W i l l n e r, G. S i w i e c, J. B o t o r, The surface tension of liquid Cu-Fe-Sb alloys, Applied Surface Science **256** (9), 2939-2943 (2010).

[16] G. S i w i e c, J. B o t o r, B. M a c h u l e c, The surface tension determination throught the estimation of the parameters of the sessile drop equation, Archives of Metallurgy **48** (2), 209-221 (2003).

[17] G. S i w i e c, J. B o t o r, The influence of antimony, lead and sulphur on the surface tension of liquid copper, Archives of Metallurgy and Materials **49** (3), 611-621 (2004).

*Received: 10 May 2013.*