Spectrochimica Acta Part B 141 (2018) 94–98

Contents lists available at ScienceDirect

Spectrochimica Acta Part B

journal homepage: www.elsevier.com/locate/sab

Research note

Laser-induced breakdown spectroscopy measurement of a small fraction of rhenium in bulk tungsten

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ARTICLE INFO

Article history:
Received 14 June 2017
Received in revised form 18 December 2017
Accepted 24 January 2018
Available online 2 February 2018

Keywords:
LIBS
Rhenium
Tungsten-rhenium alloy
Boltzmann plot
Calibration-free

ABSTRACT

Laser-induced breakdown spectroscopy (LIBS) of bulk rhenium (Re) and tungsten (W)-Re alloy has been performed using a Q-switched Nd:YAG laser (wavelength = 1064 nm, pulse width ~4–6 ns, laser energy = 115 mJ). It is found that the electron temperature, T_e, of laser-induced Re plasma is lower than that of W plasma, and that T_e of W-Re plasma is in between Re and W plasmas. This indicates that material properties affect T_e in a laser-induced plasma. For analysis of W-3.3%Re alloy, only the strongest visible Re I 488.9 nm line is found to be used because of the strong enough intensity without contamination with W lines. Using the calibration-free LIBS method, the atomic fraction of Re, c_{Re}, is evaluated as a function of the ambient Ar gas pressure, P_{Ar}. At $P_{Ar} < 10$ Torr, LIBS-measured c_{Re} agrees well with that from EDX (energy-dispersive X-ray micro-analysis), while c_{Re} increases with an increase in P_{Ar} at >10 Torr due to spectral overlapping of the Re I 488.9 nm line by an Ar II 488.9 nm line.

1. Introduction

Tungsten (W) and its alloys are promising candidates for plasma-facing materials (PFMs) in future fusion reactors. In ITER, pure W will be used as the divertor PFM. The advantages of W as a PFM are the highest melting temperature of metals, high thermal conductivity, low physical sputtering yield, and low hydrogen isotope retention [1–4].

Deuterium (D)-tritium (T) fusion reactions produce 14 MeV neutrons, which irradiate W PFM. The high energy neutron irradiation can lead to the transmutation of W, producing neighboring atoms in the periodic table as well as hydrogen (H) and helium (He). The main product of W-transmutation is rhenium (Re), the concentration of which is calculated to be around 0.2 at.% after 2 years DD and 12 years DT ITER experiments and around 4 at.% after 5 years irradiation under first wall fusion power-plant conditions [5].

The ability of laser-induced breakdown spectroscopy (LIBS) for in situ diagnostics of PFMs in fusion reactors has been examined (see e.g. [6–8]), since post-mortem analyses of PFMs by removing PFM tiles will be largely restricted in future radioactivated reactors. In recent years, LIBS has been examined for analyses of both bulk W materials [9–12] and deposited W layers [13–16].

The purpose of this work is to assess whether LIBS can quantitatively measure a small fraction (a few at.%) of Re in W, which mimics W PFMs irradiated by high-energy neutrons in fusion reactors. First, laser-induced Re plasmas produced from a pure Re sample are characterized. It should be noted that laser-induced Re plasmas have not been extensively studied so far compared to W. Next, a W sample containing a small fraction of Re is analyzed. The fraction of Re is quantitatively measured based on the calibration-free LIBS method [17], and then compared with EDX (energy-dispersive X-ray micro-analysis) of the sample.

2. Experimental setup

The LIBS experimental setup is briefly described below. The details can be found in Refs. [10,11].

A Q-switched Nd:YAG laser (Continuum Surelite III-10) was used in this experiment. The output laser wavelength and pulse width are 1064 nm and ~4–6 ns, respectively. The output laser energy was set to 115 mJ. A laser pulse was focused onto a target surface with a plano-convex lens (focal length = 250 mm). A target was placed inside a vacuum chamber, which is pumped down with a rotary pump. Thus, the minimum ambient gas pressure was ~5 mTorr, and Ar gas was injected to investigate the effect of the ambient gas pressure, P_{Ar}.

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https://doi.org/10.1016/j.sab.2018.01.013
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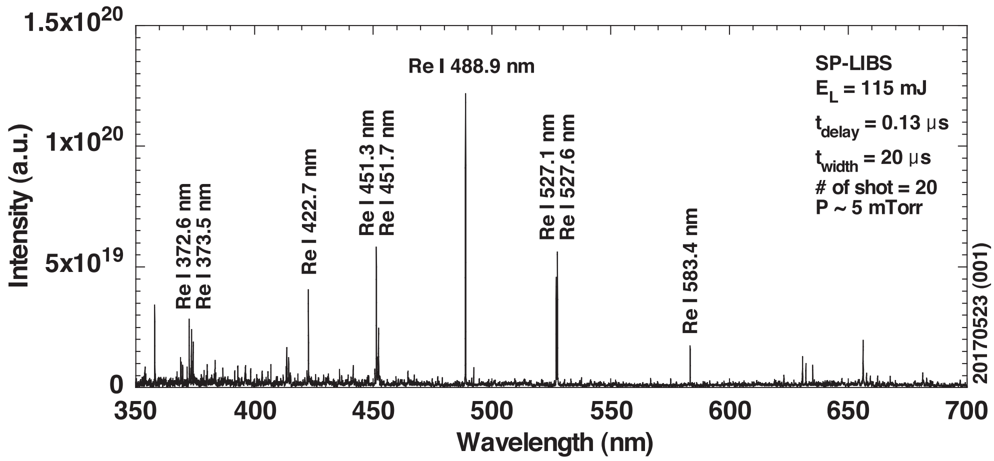

Fig. 1. Typical single-pulse (SP) LIBS spectrum of laser-induced Re plasma at $P \sim 5$ mTorr. Only major peaks of Re I lines are labeled. The spectrum is corrected for the spectral response.

Laser-induced plasma light was collected with a lens at the front of the target surface. An optical fiber was coupled to the lens, and the other end of the fiber was connected to the entrance slit of an Echelle type spectrometer (Andor ME5000). An ICCD camera (Andor iStar DH334T) was attached to the exit port of the spectrometer. The spectral sensitivity of the entire spectroscopic system including a vacuum window was calibrated with an integrating sphere (Gooch & Housego OL 455-12) at a wavelength, λ, range of $350 \le \lambda \le 1000$ nm, while the spectrometer can simultaneously cover a wider range of $200 \le \lambda \le 1000$ nm. Since the vacuum chamber is not large enough to accommodate the integrating sphere, the sphere was placed outside the chamber. Then, the calibration of the spectroscopic system was done by putting the vacuum window in between the exit port of the sphere and the lens. In this experiment, 20 plasma emission spectra were accumulated for better accuracy.

The ICCD camera was triggered with a TTL signal from the laser. The ICCD delay time, t_{delay}, and the gate width, t_{width}, were set to 0.13 μs, to exclude strong continuum emission, and 20 μs, to cover most of the plasma lifetime, respectively. Thus, the measured quantities presented here are averaged over the gate width. Note that t_{delay} is relative to the timing when a laser pulse hits the target surface.

3. Laser-induced pure Re plasma

In this section, properties of laser-induced Re plasmas are explored. Fig. 1 shows a typical laser-induced Re plasma spectrum with no Ar gas injection, i.e. $P \sim 5$ mTorr. In the figure, only major Re I peaks are labeled. The Re I 488.9 nm line is found to be strongest in the presented wavelength range, which is one of the resonance transitions. The P_{Ar} dependence of the Re I 488.9 nm and another resonance transition 527.6 nm line emission intensity is plotted in Fig. 2. As P_{Ar} increases, the intensity increases and peaks at $P_{Ar} \sim$ 10–30 Torr. Then, the intensity drops at $P_{Ar} > 30$ Torr. This behavior as well as the peak P_{Ar} are very similar to laser-induced W plasmas with Ar background gas.

For Boltzmann plot analysis to obtain the electron temperature, T_e, 21 Re I transitions were carefully selected from laser-induced Re plasma spectra. Since necessary atomic data of Re I are not currently available in the NIST database [18], we took them from another database [19]. This database compiles Re I data from Refs. [20,21]. Spectroscopic parameters of selected 21 Re I transitions are listed in Table 1.

A Re I Boltzmann plot for $P \sim 5$ mTorr is presented in Fig. 3 (a), which is obtained from the spectrum shown in Fig. 1 and the spectroscopic parameters listed in Table 1. The data points are well fit to the straight line, giving $T_e = 0.44 \pm 0.03$ eV.

Since the electron density was not measured in this study, we cannot judge whether if the plasma is in LTE (local thermodynamic equilibrium) or not based on the McWhirter criterion [22]. However, the plasma is expected to be close to LTE, because, as demonstrated in Fig. 3, the population well obeys the Boltzmann distribution [17].

It is seen from Fig. 3 that the intensity of the resonance transitions (488.9 nm and 527.6 nm) becomes gradually lower than the fitted lines as P_{Ar} increases. This behavior is similar to W I lines, which suffer from self-absorption [10]. Thus, it is considered that self-absorption of the resonance transitions may occur in the laser-induced Re plasmas at higher P_{Ar}, while the effect of self-absorption on T_e seems to be small.

Here, we compare the database [19] that we employed here with another data set [23], which was recently reported. In the newer data set [23], $A_{ki} = 1.16e + 7\,s^{-1}$ was given for the 488.9 nm line,

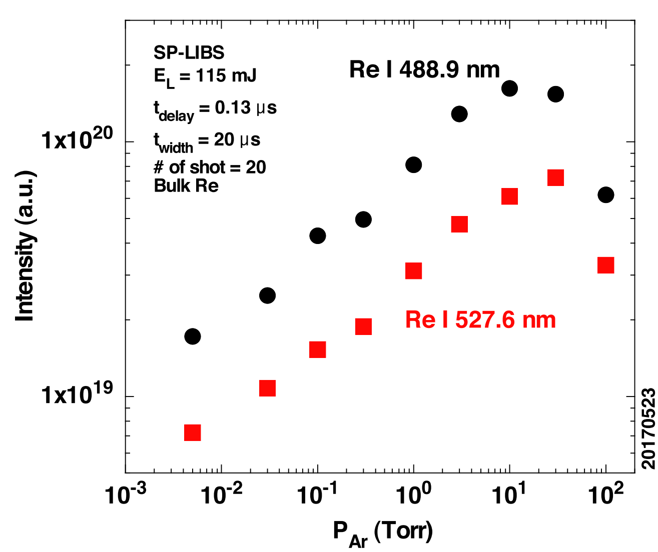

Fig. 2. Ar pressure dependence of the line emission intensity of the Re I resonance transitions 488.9 nm and 527.6 nm. Note that the intensity is corrected for the spectral response.