

OPTICAL PROPERTIES OF NaK DROPLETS

Carsten Wiedemann⁽¹⁾, Stephan Erwig⁽¹⁾, Michael Oswald⁽¹⁾, Sebastian Stabroth⁽¹⁾, Heiner Klinkrad⁽²⁾,
Peter Vörsmann⁽¹⁾

⁽¹⁾*Institute of Aerospace Systems, Technische Universität Braunschweig, Hermann-Blenk-Str. 23, 38108 Braunschweig, Germany, Email: c.wiedemann@tu-bs.de*

⁽²⁾*ESA/ESOC, Robert-Bosch-Str. 5, 64293 Darmstadt, Germany, Email: heiner.klinkrad@esa.int*

ABSTRACT

Sixteen nuclear powered satellites of the type RORSAT launched between 1980 and 1988 activated a reactor core ejection system in Sufficiently High Orbits (SHO), mostly between 900 km and 950 km altitude. The core ejection causes an opening of the primary coolant circuit. The liquid coolant consists of eutectic sodium-potassium alloy (NaK-78) and has been released into space during these core ejections. The droplets are modeled as debris objects with an individually constant diameter. In this paper it is investigated, if the diameter can change due to evaporation. The evaporation rate depends on the temperature. The temperature is a function of the optical properties. The optical properties depend on the alloy composition and the droplet temperature. The alloy composition changes during the evaporation process, resulting in an enrichment of sodium in a droplet. A model is derived, which determines the optical constants as function of temperature and alloy composition. The results show that the evaporation rate depends strongly on the optical properties.

1. INTRODUCTION

NaK coolant releases are restricted to a very narrow region near 65° inclination. They contributed to the space debris environment in the centimeter and millimeter size regime. NaK droplets have been modeled before in ESA's MASTER Debris and Meteoroid Environment Model. The approach is revised for the MASTER 2005 upgrade. The next version of the MASTER model is developed by a consortium under ESA/ESOC contract. The consortium is led by the Institute of Aerospace Systems of the Technische Universität Braunschweig (Germany). MASTER 2005 is developed in cooperation with QinetiQ (UK), supported by Forschungsgesellschaft für Angewandte Naturwissenschaften FGAN (Germany) and Astronomisches Institut Universität Bern AIUB (Switzerland). According to today's knowledge of the reactor design and operation, the reactor core ejection is responsible for the opening of the primary coolant circuit. A model for the release of NaK droplets has been derived. The model considers a non-uniformly distribution of the directions of the droplet release velocities. The maximum additional droplet velocity is estimated to be

30 m/s. The mean velocity is 15 m/s. The most likely mechanisms for the droplet generation are the surface tension driven Rayleigh breakup and the effervescent atomization. Two orifice diameters are assumed. The droplet size distribution is expressed as volume distribution using the Rosin-Rammler equation. The Rosin-Rammler equation is applied to both orifice diameters, resulting in a bimodal size distribution. The largest droplet has a diameter of 5.67 cm. The smallest released droplet is assumed to be 0.5 mm in diameter. The size distribution model fits well with published measurement data of radar observations. It is estimated that altogether 208 kg of NaK-78 (13 kg per RORSAT) have been released on orbit. Simulation runs show that there are still 66,000 droplets with a total mass of 157 kg in orbit at the beginning of 2005, whereas the smallest droplet has a diameter of 5 mm (Wiedemann et al., 2004a, 2004b, 2004c).

Depending on the optical properties, NaK droplets can be liquid. Thus their lifetime is determined by the atmospheric drag and the evaporation rate. The investigations of this work shall give the bases for the estimation of the evaporation rate. According to the literature, there is only a limited number of available measurement data for NaK alloys. Furthermore the published data for pure sodium and potassium are controversial. Thus it is necessary to investigate the optical properties of alkali metals and their alloys. A bibliographical study concerning the theory and measurement results has been carried out. The results are reviewed in detail. Because of the limited number of measured data for NaK alloys, an investigation on the physical theory is necessary. Based on this, the optical constants of NaK alloys are estimated. The presented results consider the Drude-theory, which uses electrical properties to derive the optical constants of a metal. The theory is compared with measured data. The deviations are discussed. The aim of this work is the estimation of the correct order of magnitude of the optical properties of NaK alloys. This model will be used to determine the diameter reduction considering the different evaporation rates of the alloy compounds.

The optical properties have a significant influence on the temperature of a droplet. The droplet temperature is calculated using the solar absorptivity α and the infrared

emissivity ε . These coefficients depend on the temperature and on the alloy composition. Due to the lack of measured values for NaK alloys with varying composition, the optical constants shall be calculated here. Important basic work has been done by Grinberg et al. (1997) and Lambour et al. (1999), who used constant values for the optical properties (independent of temperature and alloy composition). Grinberg et al. (1997) calculated the droplet temperature and estimated the lifetime due to evaporation. Lambour et al. (1999) calculated the evaporation rate based on a thermal model of a droplet. The optical constants which have been used for these calculations are summarized in Tab. 1. Lambour et al. (1999) give a value for the absorptivity which correlates reasonable with the measured reflectivity R of larger orbiting NaK droplets. The value of the emissivity refers to unpublished Russian work. Grinberg et al. (1997) give a lower value for the absorptivity. Both use α/ε ratios close to "1". Because metals generally have $\alpha/\varepsilon > 1$, the choice of $\alpha/\varepsilon = 1$ means an estimation of a minimum temperature (minimum evaporation rate) for a droplet.

α	ε	α/ε	Reference
0.156	0.132	1.182	Lambour, 1999
0.050	-	1.000	Grinberg, 1997

Table 1. Optical constants of NaK droplets given in the literature.

R	Reference
0.84 - 0.89	Sridharan, 1999
0.85 \pm 0.07	Hall, 2004

Table 2. Measured reflectivity of orbiting NaK droplets.

Havstad et al. (1995) estimated the optical properties of liquid metal alloys using the Drude-theory. Based on the work of Lambour et al. (1999) and Havstad et al. (1995), the initial approach in this work was to combine the calculations of the evaporation rate with an improved estimation of the optical properties. But a detailed literature review revealed that the absorptivity of alkali metals is controversial, especially in the visible and the near infrared.

2. THE DRUDE-THEORY

A theory for the computation of the optical constants of metals is given in the literature which is suitable especially for the alkali metals. This theory was developed by Paul Drude and derives the optical constants from the electrical properties. Drude assumes that the valence electrons can be treated as free electrons. The band structure and the periodic structure of the potential of the crystal are neglected. The electrons behave like an ideal gas of uncharged particles. The model of free electrons is a very simple model for the description of the valence electrons in metals. This assumption is sufficient for alkali metals,

because they show a nearly free electron behavior. The Drude free electron parameters, and hence the optical constants and the radiant properties can be computed from the dependence of the Hall coefficient and direct current resistivity on alloy composition. The Hall coefficient gives the free electron density and the resistivity gives the average time between collisions. Havstad et al. (1995) have found that published values for the Hall coefficient and the electrical resistivity of liquid metal alloys can provide useful estimates of the optical constants of some groups of binary liquid metal and solid alloys.

The optical properties are expressed as complex dielectric constant ε' . The parameters and the used notation is given in the following.

$$\varepsilon' = \varepsilon_1 + i\varepsilon_2 \quad (1)$$

$$\varepsilon_1 = 1 - \frac{4\pi e^2 \tau^2}{m(1 + \omega^2 \tau^2)} = 1 - \frac{N_e e^2 \tau^2}{m\varepsilon_0(1 + \omega^2 \tau^2)} = n^2 - k^2 \quad (2)$$

$$\varepsilon_2 = \frac{4\pi e^2 \tau}{m\omega(1 + \omega^2 \tau^2)} = \frac{N_e e^2 \tau}{m\varepsilon_0 \omega(1 + \omega^2 \tau^2)} = 2nk \quad (3)$$

The following parameters are used: elementary charge e [1.6e10-19 C], extinction coefficient k , mass of an electron m [kg], index of refraction n , permittivity of free space ε_0 [8.85e-12 F/m], real part ε_1 and imaginary part of the complex dielectric function ε_2 [F/m], and angular frequency ω [1/s]. The Drude-model can be applied by computing the number density N_e [1/m³] and the average time between collisions τ [s] from measurements of the Hall coefficient R_H [m³/(As)] and the direct current resistivity ρ [Ω m] (Havstad et al., 1995).

$$N_e = 1/(R_H e), \quad \tau = m/(N_e e^2 \rho) \quad (4)$$

The monochromatic reflectivity R_λ can be calculated from the optical constants. (Sometimes the absorption index κ instead of the extinction coefficient is used.)

$$n = \left[\frac{1}{2} \left(\sqrt{\varepsilon_1^2 + \varepsilon_2^2} + \varepsilon_1 \right) \right]^{0.5} \quad (5)$$

$$k = \left[\frac{1}{2} \left(\sqrt{\varepsilon_1^2 + \varepsilon_2^2} - \varepsilon_1 \right) \right]^{0.5} = n\kappa \quad (6)$$

$$R_\lambda = \frac{(n-1)^2 + k^2}{(n+1)^2 + k^2} = \frac{n^2(1+\kappa^2) + 1 - 2n}{n^2(1+\kappa^2) + 1 + 2n} \quad (7)$$

The absorption α or emissivity ε can be derived by weighting $1 - R_\lambda$ with the solar spectrum or the black body temperature and integration over all wavelengths. Using a thermal model similar that of Lambour et al.

(1999), the thermal behavior (including evaporation) of a droplet can be calculated.

For the application of the Drude-model, the surface composition of a droplet has to be known. Concerning the alloy composition at the surface, there are different views in the literature. Meshcheryakov (1996, 1997) expects that the surface should consist of sodium, because potassium evaporates faster. In opposition to that most theoretical approaches predict a phase segregation. Surface segregation is the enrichment of the concentration of one component at the surface of an alloy. The driving force for the surface segregation is the reduction in the surface energy (Kiejna, 1990). According to this there should be an enrichment of potassium at the surface due to its lower surface tension, compared to sodium. Other investigations revealed only a weak tendency towards segregation, because alkali metals are well soluble in each other (DiMasi et al., 2000; Gonzales et al., 1996). Thus in this work, the alloy composition of bulk and surface is treated to be identical. Also the influence of impurities is important. Contamination tests have been made with oxygen. According to Tostmann et al. (2000) and Shpyrko et al. (2003) the exposure of NaK to large doses of oxygen did not change the surface. This lack of surface contamination is believed to be due to the high solubility of the oxides of these metals in the bulk liquid. Furthermore the low surface tension of liquid alkali metals does not support segregation of the oxides at the surface. The surface of the NaK alloy was found to remain automatically clean.

3. THE ANOMALOUS ABSORPTION

Experimentally determined measurement data of the monochromatic optical constants for sodium (Hietel, 1965) and potassium (El Naby, 1962) revealed an anomalous absorption in the visible and the near infrared. This absorption cannot be explained by the Drude-model. The results of these measurements initiated several theoretical and experimental studies. To follow the discussion on these studies and to find more measurement data, an extensive literature review was carried out. This bibliographical study comes to the result that the optical constants of the alkali metals are still controversial. Most of the work refers to monochromatic optical constants of sodium or potassium. Only one reference was found which investigated NaK alloys. Unfortunately the results of this reference are limited to one single wavelength.

The monochromatic optical properties of alkali metals have been investigated extensively. Theoretical and experimental results of sodium are given in the literature (Hietel, 1965; Chan et al., 1982; Hattori, 1984; Smith, 1967; Meshcheryakov, 1996; Helman, 1977; Foo, 1969; Appelbaum, 1966; Foo et al., 1968; Sturm et al., 1973;

Overhauser, 1967). Some references refer results of potassium (El Naby, 1962, 1963; Mayer et al., 1963a, 1963b; Harms, 1972; Kamiuto, 1986; Ching et al., 1973; Alouani et al., 1989; Overhauser et al., 1976; Cohen et al., 1964; Overhauser, 1964). Several papers give results of both, sodium and potassium (Monin et al., 1974; Smith, 1968; Ujihara, 1972; Mayer et al., 1966; Smith, 1969; Whang et al., 1972; Duncan et al., 1913; Palmer et al., 1971; Abeles, 1968; Hodgson, 1972; Animalu, 1967; Sievers, 1980; Stevenson, 1973; Bennett et al., 1972; Fragachan et al., 1985; Esposito et al., 1968; Young, 1969; Butcher, 1951; Meessen, 1961). Only one reference was found that deals with monochromatic optical properties of NaK alloys for one selected wavelength (Morgan, 1922).

The investigators who try to explain the absorption anomaly assume several effects. These are for example the intraband absorption (Drude-absorption), interband absorption (transfer of an electron into a higher band), collective electron interactions, and anomalous skin-effect (which is believed to be small). The experimentally observed absorption anomaly could not be reproduced by all investigators.

Due to the uncertainty concerning the optical constants, two approaches will be used here. One is the Drude-model, and the other one is the Drude-model plus additional absorption. The monochromatic additional absorption is derived, using data from Hietel (1965) and El Naby (1962). For NaK alloys the additional absorption is interpolated between the values of pure sodium and pure potassium. Both approaches are approximate solutions which may be used to estimate the minimum or maximum heating of a droplet. The monochromatic reflectivity of sodium, potassium, and NaK-78 is shown in Fig. 1 and Fig. 2.

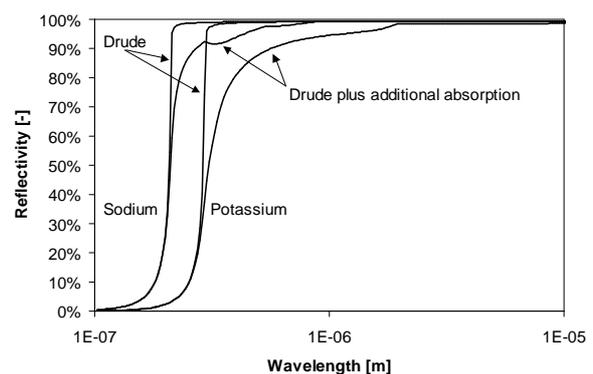


Figure 1. Monochromatic reflectivity of sodium and potassium at room temperature. The Drude-model is compared to measured data from Hietel (1965) and El Naby (1962) which both show additional absorption. The additional absorption reduces the reflectivity.

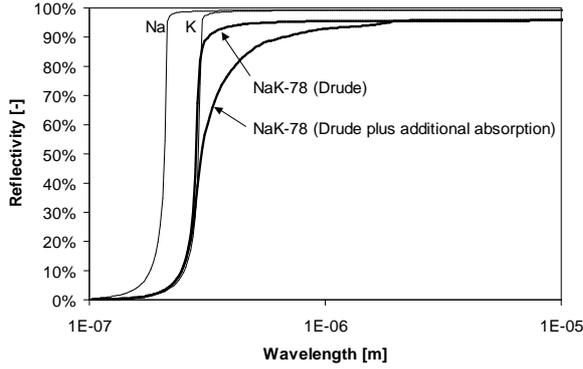


Figure 2. Monochromatic reflectivity of NaK-78 at room temperature. The Drude-model is compared with estimated values for the additional absorption, derived from interpolation. The reflectivity of the eutectic NaK alloy is lower, compared to pure sodium and potassium due to the higher electrical resistivity of NaK.

4. RESULTS

Using the optical properties with and without additional absorption, the thermal state of NaK droplets was calculated and compared. For the calculation, a thermal model similar to that of Lambour et al. (1999) was applied. The results are shown in Fig. 3 and 4. For the Drude-model, the α/ε ratios vary from 1.44 (340 K) to 1.58 (240 K). Considering an additional absorption, these values are significantly higher. The α/ε ratios vary from 3.11 (380 K) to 5.04 (240 K). Based on the thermal state, the long term behavior of a droplet with an initial diameter of 1 cm was computed. The results are shown in Fig. 5. In both cases potassium is evaporating more rapidly. This causes a fast diameter reduction, until the droplet consists only of sodium.

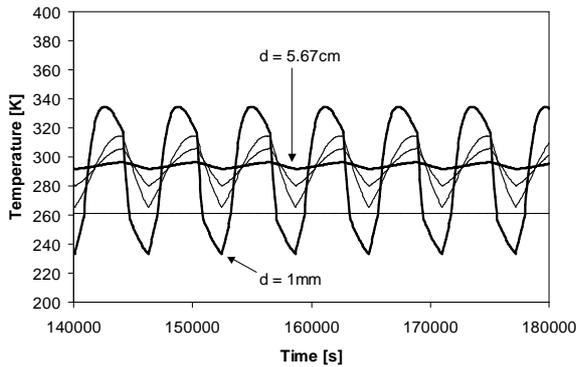


Figure 3. Thermal state of NaK-78 droplets with the diameters $d = 1 \text{ mm}$, 5 mm , 1 cm , and 5.67 cm using the Drude-model. The simulation shows the thermal behavior of droplets on a shadow orbit including evaporation at an orbital altitude of 900 km, 38.9 hours after the coolant release. (The straight line is the melting point of NaK-78 at 261 K.)

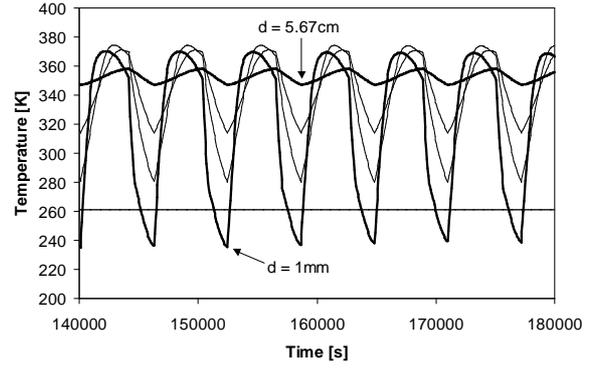


Figure 4. Thermal state of NaK-78 droplets with the diameters $d = 1 \text{ mm}$, 5 mm , 1 cm , and 5.67 cm considering additional absorption. (Other conditions are similar to Fig. 3.)

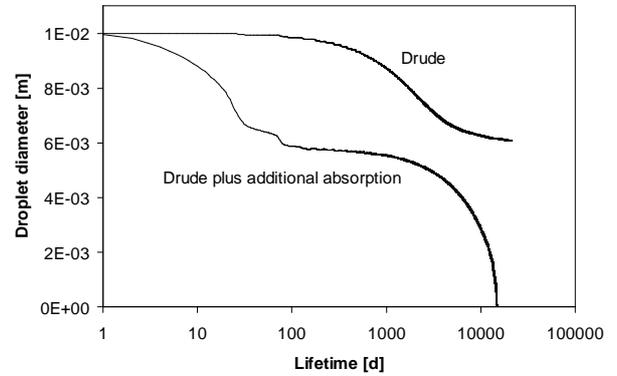


Figure 5. Long term simulation of the lifetime due to evaporation for a NaK droplet with an initial diameter of 1 cm at a constant orbital altitude of 900 km. The two different models for the optical properties are compared. The use of the Drude-model results in a significantly longer lifetime.

5. CONCLUSION

An extensive literature study concerning the optical properties of alkali metals was carried out. It was found that the optical properties of sodium and potassium are controversial. Thus two different approaches for the estimation of the monochromatic optical constants were selected for a sensitivity analysis. In both cases, the total optical properties result in higher α/ε ratios as given in the literature. This causes higher droplet temperatures. The comparison shows that the optical properties have a strong influence on the evaporation lifetime of a droplet.

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