PHOTOSTIMULATED LUMINESCENCE TECHNIQUE APPLICABLE TO PRE-SCREENING OF K-RICH MATERIALS IN CHONDRITES. Tatsunori Yokoyama[1] , K. Misawa[2], O. Okano[3], H. Minowa[4], T. Fukuoka[5]. [1]National Museum of Nature and Science, 4-1-1 Amakubo, Tsukuba, 305-0005, Japan, [2]National Institute of Polar Research, Tachikawa, 190-8518, Japan (misawa@nipr.ac.jp), [3]Okayama Univ., Okayama, 700-8530, Japan, [4]Jikei Univ. School of Medicine, Tokyo, 105-8461, Japan, [5]Rissho Univ., Kumagaya, 360-0194, Japan.

Introduction: Relative to the Sun's photosphere, alkaline elements are more or less depleted in the inner solar system bodies (Earth, Moon, Mars and Vesta) and all meteorites except CI-chondrites [1]. This depletion implies that volatility-controlled elemental fractionations could have occurred in the early solar nebula (e.g. [2]). In relatively few cases, very alkali-rich materials have been observed in chondritic breccias: K-Rb-Cs-rich igneous rock fragments in Bhola (LL3–6), Krähenberg (LL5), Y-74442 (LL4), Siena (LL5) and Acfer 111 (H3–6) [3–7], and halite and sylvite in the Zag (H3–6) and Monahans (1998) (H5) chondrites [8,9] as well as in returned samples from 25143 Itokawa [10,11]. Cosmochemical studies on these alkali-rich materials provide good opportunities to understand fractionation processes of moderately volatile elements that occurred on their parent bodies or at an early stage of the solar system evolution [3,5,7,8,12]. Moreover, the alkali-rich materials might have caused a large alkali/alkaline earth fractionation, which, in turn, could be possible more precise age dating utilizing the parent-daughter pairs of long-lived radionuclides (^{87}Rb-^{87}Sr and ^{40}K-^{40}Ca isotopic systems [3,7,13]).

For such purpose, nondestructive identification and separation of small-sized alkali-rich materials prior to chemical and isotopic analyses are preferred because the alkali enrichments are often limited within very small spots, say usually less than a few mm across. In order to discriminate and separate alkali-rich fragments from others, we have conducted autoradiography experiments on small rock fragments from the brecciated chondrite Y-74442 using an imaging plate (IP). The IP is composed of a plastic plate coated with photostimulable phosphor crystals (BaFBr:Eu^{2+}) which absorbs incoming radiation and emits photostimulated luminescence (PSL) by a He-Ne laser scan [14].

Experimental: Control samples (KCl crystals and KCl solution dried down on teflon sheets) put in heat-sealed polyethylene bags were prepared. These samples were laid on IPs (Fujifilm, model BAS III-2040) and were put in a plastic cassette case. They were stored in a lead-armored chamber at the Radioisotope Research Center, Jikei University School of Medicine for one to three weeks at room temperature.

Potassium-rich fragments in Y-74442 were identified by autoradiography using a similar technique described by [15] with resultant improved sampling of material for this study. The Y-74442 sample was coarsely crushed in a boron carbide mortar and small fragments were examined under a binocular microscope. Dark grey-colored fragments of Y-74442 were weighing and photographed. We have designed a sample holder made of acrylic material (3 mm thick) which can also hold the IP of 96 x 129 mm in size. Individual sample and KCl standard were stored in doubly wrapped, heat-sealed polyethylene sheets (sample bags) and were laid on an acrylic plate holding by double-stick tapes.

The exposed IPs were run through an LD 650 nm laser scanner with an IP filter (GE Healthcare, model Typhoon FLA7000) that read and digitized the images with a 25 µm pixel-size resolution at the Radioisotope Research Center, Jikei Univ. School of Medicine. PSL-value (IP volume) was derived from the background corrected, averaged blackening of a certain area of the resulting image.

After the radiography experiments the rock samples were taken out from the bags and were decomposed in a mixture of HF and $HClO_4$, and then combined with mixed ^{87}Rb-^{84}Sr and ^{40}K-^{48}Ca spikes. Calcium, Sr and (K + Rb) were separated using a cation exchange column. The K, Ca, Rb and Sr abundances were determined by isotope-dilution thermal ionization mass spectrometry (ID-TIMS) [7,13].

Results and Discussion: We used acrylic IP holders and polyethylene sample bags. As a result, background noises during exposure were reduced down to 0.4 volume/pixel/day. In addition to radiations from radionuclides contained in the samples, natural background radiations (cosmic-rays and natural γ-rays) also contribute to further ionization of some Eu^{2+} to Eu^{3+} in the phosphor screen. Thus, for samples with K in the low µg range, which will require several weeks to generate an image, it is necessary to store the IP in a shielded area.

Figure 1 shows correlations between K (in µg) and PSL-value (IP volume, a.u.) for different exposure times. Fading of the stored images on the IP could have occurred after several weeks' exposure [16]. Exposure time depends on the concentration of the radioactive emitters in the sample. Total incident is expected to be ~30 decay events of ^{40}K per day for the sample containing 10 µg K. In order for samples containing several µg K to be visible, a longer exposure

time is necessary. Finally, we have undertaken radiography experiments for fifty days under a nitrogen purge to reduce particles from decay of radium at the Low-level Radioisotope Measurement Facility, Institute for Cosmic Ray Research, Univ. of Tokyo, which is located in a basement 23 m from the surface. Among twenty-seven rock-fragments weighing 0.23 to 17.3 mg, we have regarded at least eighteen as K-rich (Fig. 2).

The autoradiography method cannot discriminate between different types of radioactivity (α-, β- and γ-radiations). A certain level of radioactivities could have come from radionuclides other than ^{40}K, such as ^{87}Rb and decay series of ^{235,238}U and ^{232}Th. Alkali-rich rock fragments in LL-chondritic breccias, Rb and U contents are 100 ppm and 0.02 ppm, respectively [5,7]. Thus, total amounts of Rb, U and possibly Th in the samples become 1 µg, 0.2 ng and 1 ng at most, respectively, suggesting that the samples do not contain sufficient amounts of radionuclides, except ^{40}K, that produce PSL on IP. Polyethylene films reduce α-particles from the samples. Moreover, γ-rays only weakly interact with the radiosensitive screen. These facts suggest that the obtained PSL are mainly come from decay of ^{40}K. We futher take into account self-shielding within the sample material.

Conclusions: The autoradiography using IPs are useful to identify not only K-rich rock fragments in chondritic breccias but also K-rich chondrules as well as sylvite in chondrites without serious contamination if appropriate sample preparation and shielding from natural radiations are conducted. Detection limits to identify K-rich materials are considered to be ca. 10 µg of K in mm-sized samples.

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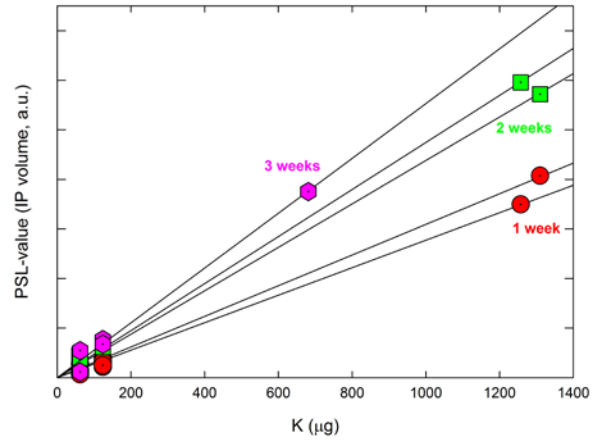

Fig. 1. Correlations between K (in µg) and PSL-value for different exposure times.

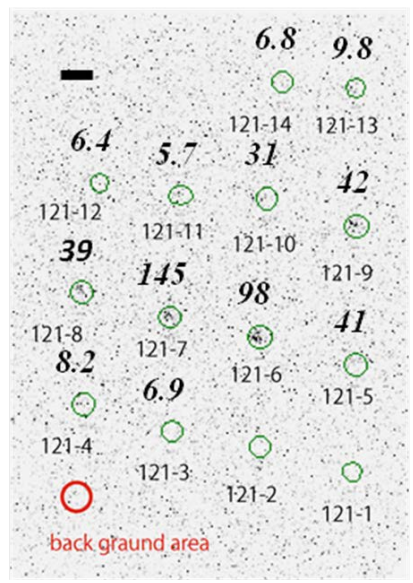

Fig. 2. Radiographic image showing the intensity of the PSL. Rock fragments separated from the Y-74442 LL breccia were exposed for fifty days. ***Italics*** = K (in µg) in rock samples determined by ID-TIMS [7,13]. Scale bar = 10 mm.