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Observed changes in electrical materials properties of ZrO_2 in Megapie leak detectors during irradiation

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ABSTRACT

Reliable leak detection is of high importance for liquid metal targets. For the PSI Megapie target two novel approaches had been pursued: a thermocouple-based leak detector and one employing capacity sensors with impedance read out. Whereas the temperature based leak detector performed according to expectations, severe degradation of the impedance probes occurred during the irradiation period of Megapie. The electrical characteristics of the chosen insulator material ZrO_2 stabilized by 1% Y_2O_3 showed massive changes over time and they exhibited strong dependence on the prevailing environment conditions. Three different contributions to effective electrical conductivity in the supposed insulator could be identified: a prompt radiation induced, a temperature induced, and most probably a gas induced change in conductivity. These components, both increases and decreases in bulk conductivity of the insulating slabs in the impedance probes, demonstrated some cross-dependence on each other. The alterations in conductivity and the mutual influence of the identified factors changed with the irradiation time and the probable exposure to gaseous radiolysis products at the location close to the beam entrance window of the liquid metal target. A phenomenological and qualitative description of the observed effects as they evolved during Megapie operation is presented.

1. Introduction

Short response times and the harsh radiation environment in liquid metal targets of spallation neutron sources pose stringent requirements and demanding boundary conditions for reliable liquid metal leak detection. For the liquid metal target of Megapie, which was irradiated at the Paul Scherrer Institute, PSI, in 2006, two completely different approaches had been followed and implemented to guarantee that the proton beam would be interrupted in case lead–bismuth leaked out of the primary liquid metal container [1,2]. Whereas a thermocouple-based leak detector made no problem at all, stripe sensors showed unexpected behavior right from the beginning.

2. Stripe sensor layout

Three individual stripe-pairs were made from tungsten and arranged around the bottom end of the Megapie liquid metal container in a groove formed by the lower target enclosure. These stripes were held in place by insulator slabs produced from ZrO_2 stabilized by 1% Y_2O_3. This material was chosen after extensive research because of its superior thermal properties, in particular because of its tolerance to thermal shocks and gradients. Each stripe pair formed an open capacitor. The electrical impedance of the devices was continuously monitored by dedicated circuitry with respect to Ohmic conductance and high frequency response at 20 kHz. Wheatstone bridges were optimized for triggering an alarm with any imbalance between a stripe pair and its reference. The electronics were not designed for calibrated measurement of Ohmic conductivity or AC-impedance. Resistive and AC-channel responses were not independent of each other.

In a precursory experiment a fully representative stripe sensor was subjected to irradiation with a very similar spectrum stemming from spallation processes in Lisor 7. In half a bar helium atmosphere and at an overall radiation level in the order of 1% of the value later encountered in the Megapie target, not the slightest dependence of any reading on the actual radiation level or on the temperature was observed. Gammas were the dominated the radiation environment; during the full active operation period of the Megapie target a dose of approximately 100 MGy was accumulated in each joint made from three ZrO_2 slabs. Fig. 1 showed mechanical layout of a stripe sensor. One-sector prototype stripe sensor is shown in its dedicated test set-up before irradiation at Lisor (left). Ceramic slabs kept the stripes (and thermocouples) in place (above and right).

3. Observations during operation

In following, the most striking findings with the stripe sensors during the actual irradiation period of Megapie are reported.

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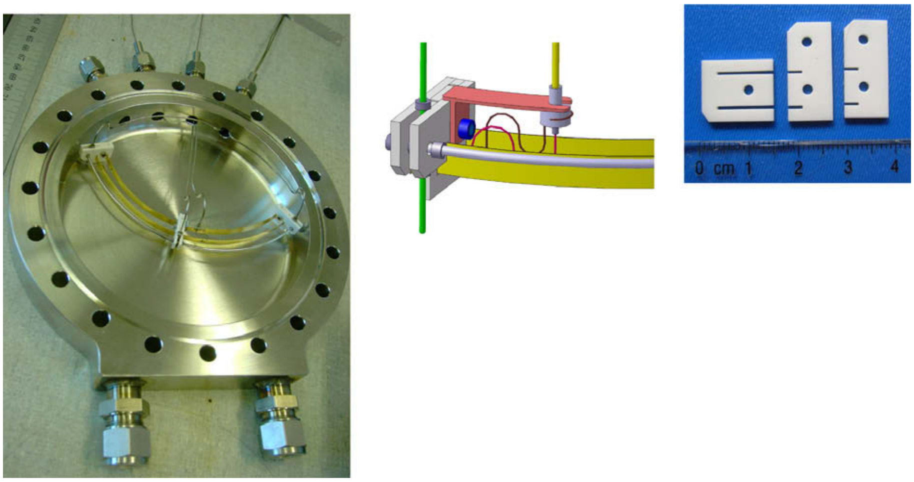


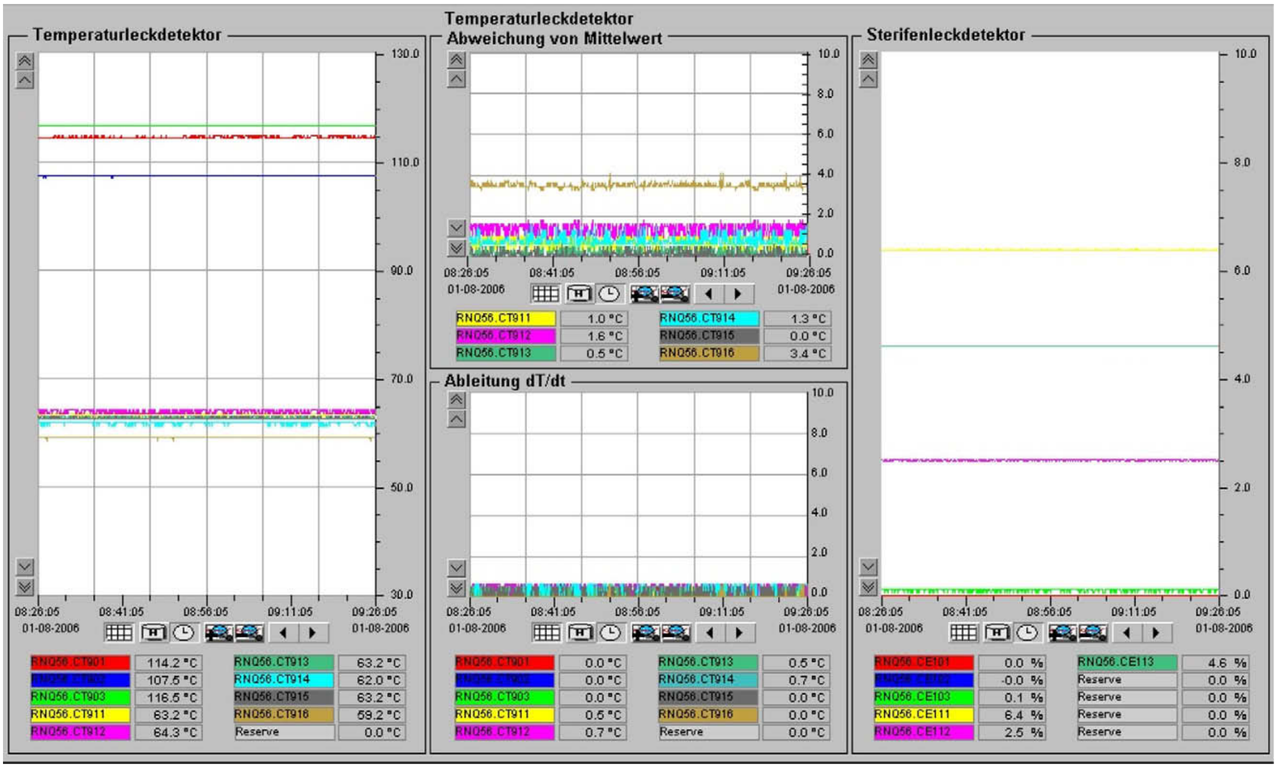
Fig. 1. Mechanical layout of a stripe sensor.

Figures and figure captions present the major developments as they occurred over the operational time the leak detectors. Non-expected behavior occurred practically immediately with the first protons. Although no real quantitative numbers were obtained, the reported qualitative observations were clear enough to arrive at a firm conclusion, which would have been nice to know at the outset of the leak detector development for the Megapie spallation environment.

Fig. 2 shows snapshot from control system screen with very stable conditions on 1 August 2006 shortly before the first protons were impinging on the target. Thermocouple readings (left panel) as well as deviations from mean and derivatives of temperatures (center panel) demonstrate very quiet conditions. Ohmic and AC-impedance signals from three stripe sensors are given in the rightmost panel. The very small temperature fluctuations are due to the action of the temperature control system in the target, they have no impact on impedance signals.

Fig. 3 shows the proton beam on the Megapie target initially ramped up in small steps (0, 33 and 40 μA) on 15 August, i.e. the second day of Megapie irradiation; panels are the same as in Fig. 2. Radiation induced conductivity in the stripe sensors lead to much faster responses in both the Ohmic as well as in the AC-impedance channels compared to the tracing of the transients in the temperatures of the thermocouples shown on the left and in the center. There was some decay in the observed stripe sensor signals with time. Noise at this point in time already was much larger than at the start.

Fig. 4 shows that at about 100 μA beam, still on the second day of irradiation, the rising transients of the impedance signals appeared to follow a thermal time course, whereas the trailing edge



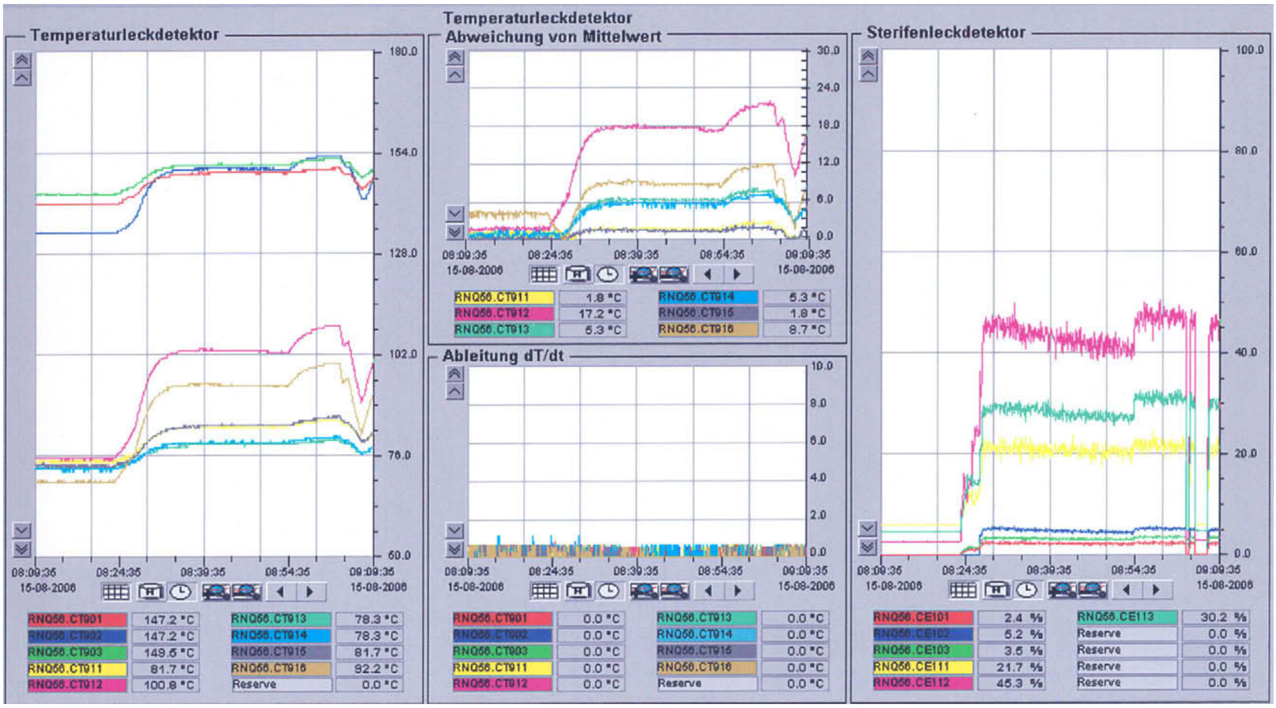
RNQ56.CT901	114.2 °C	RNQ56.CT913	63.2 °C
RNQ56.CT902	107.5 °C	RNQ56.CT914	62.0 °C
RNQ56.CT903	116.5 °C	RNQ56.CT915	63.2 °C
RNQ56.CT911	63.2 °C	RNQ56.CT916	59.2 °C
RNQ56.CT912	64.3 °C	Reserve	0.0 °C

RNQ56.CT911	1.0 °C	RNQ56.CT914	1.3 °C
RNQ56.CT912	1.6 °C	RNQ56.CT915	0.0 °C
RNQ56.CT913	0.5 °C	RNQ56.CT916	3.4 °C

RNQ56.CT901	0.0 °C	RNQ56.CT913	0.5 °C
RNQ56.CT902	0.0 °C	RNQ56.CT914	0.7 °C
RNQ56.CT903	0.0 °C	RNQ56.CT915	0.0 °C
RNQ56.CT911	0.5 °C	RNQ56.CT916	0.0 °C
RNQ56.CT912	0.7 °C	Reserve	0.0 °C

RNQ56.CE101	0.0 %	RNQ56.CE113	4.6 %
RNQ56.CE102	-0.0 %	Reserve	0.0 %
RNQ56.CE103	0.1 %	Reserve	0.0 %
RNQ56.CE111	6.4 %	Reserve	0.0 %
RNQ56.CE112	2.5 %	Reserve	0.0 %

Fig. 2. Snapshot from control system screen with very stable conditions on 1 August 2006 shortly before the first protons were impinging on the target.



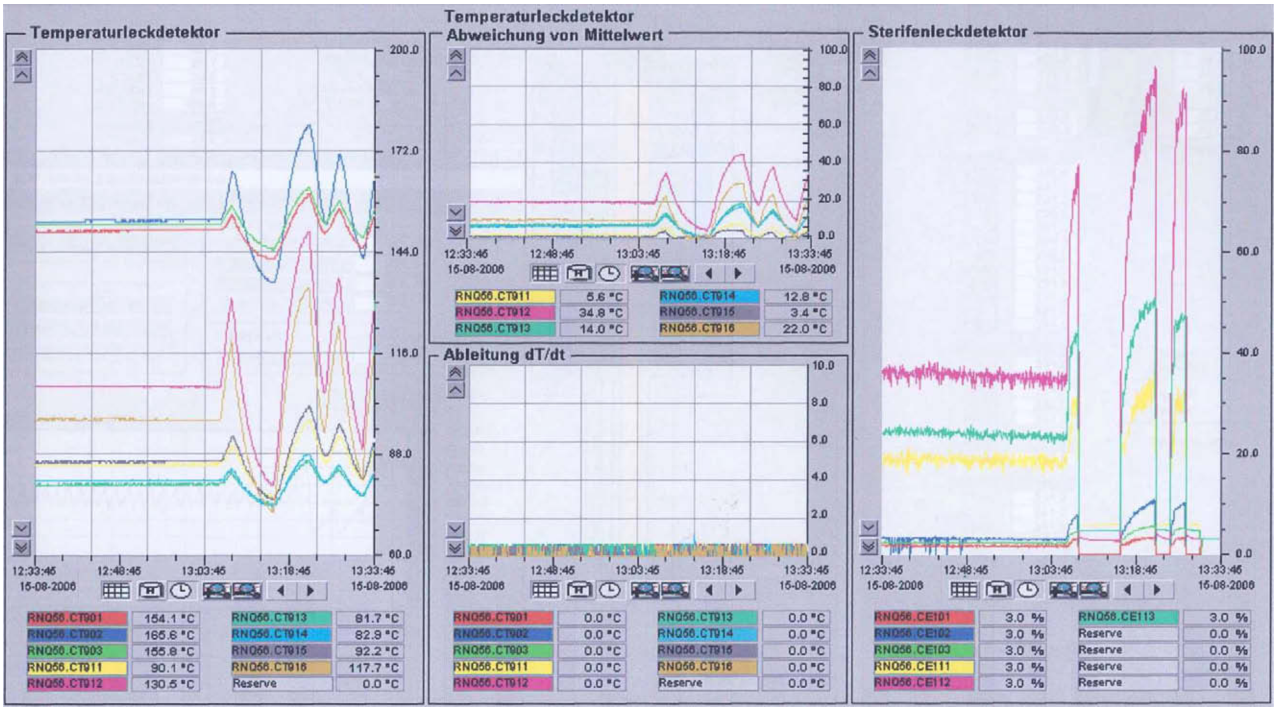
RNQ56.CT901	147.2 °C	RNQ56.CT913	78.3 °C
RNQ56.CT902	147.2 °C	RNQ56.CT914	78.3 °C
RNQ56.CT903	149.5 °C	RNQ56.CT915	81.7 °C
RNQ56.CT911	81.7 °C	RNQ56.CT916	92.2 °C
RNQ56.CT912	100.8 °C	Reserve	0.0 °C

RNQ56.CT911	1.8 °C	RNQ56.CT914	5.3 °C
RNQ56.CT912	17.2 °C	RNQ56.CT915	1.8 °C
RNQ56.CT913	5.3 °C	RNQ56.CT916	8.7 °C

RNQ56.CT901	0.0 °C	RNQ56.CT913	0.0 °C
RNQ56.CT902	0.0 °C	RNQ56.CT914	0.0 °C
RNQ56.CT903	0.0 °C	RNQ56.CT915	0.0 °C
RNQ56.CT911	0.0 °C	RNQ56.CT916	0.0 °C
RNQ56.CT912	0.0 °C	Reserve	0.0 °C

RNQ56.CE101	2.4 %	RNQ56.CE113	30.2 %
RNQ56.CE102	5.2 %	Reserve	0.0 %
RNQ56.CE103	3.5 %	Reserve	0.0 %
RNQ56.CE111	21.7 %	Reserve	0.0 %
RNQ56.CE112	45.3 %	Reserve	0.0 %

Fig. 3. The proton beam on the Megapie target initially ramped up in small steps (0, 33 and 40 μA) on 15 August, i.e. the second day of Megapie irradiation; panels are the same as in Fig. 2.



RNQ56.CT901	154.1 °C	RNQ56.CT913	81.7 °C
RNQ56.CT902	165.6 °C	RNQ56.CT914	82.9 °C
RNQ56.CT903	155.8 °C	RNQ56.CT915	92.2 °C
RNQ56.CT911	90.1 °C	RNQ56.CT916	117.7 °C
RNQ56.CT912	130.5 °C	Reserve	0.0 °C

RNQ56.CT911	5.6 °C	RNQ56.CT914	12.8 °C
RNQ56.CT912	34.8 °C	RNQ56.CT915	3.4 °C
RNQ56.CT913	14.0 °C	RNQ56.CT916	22.0 °C

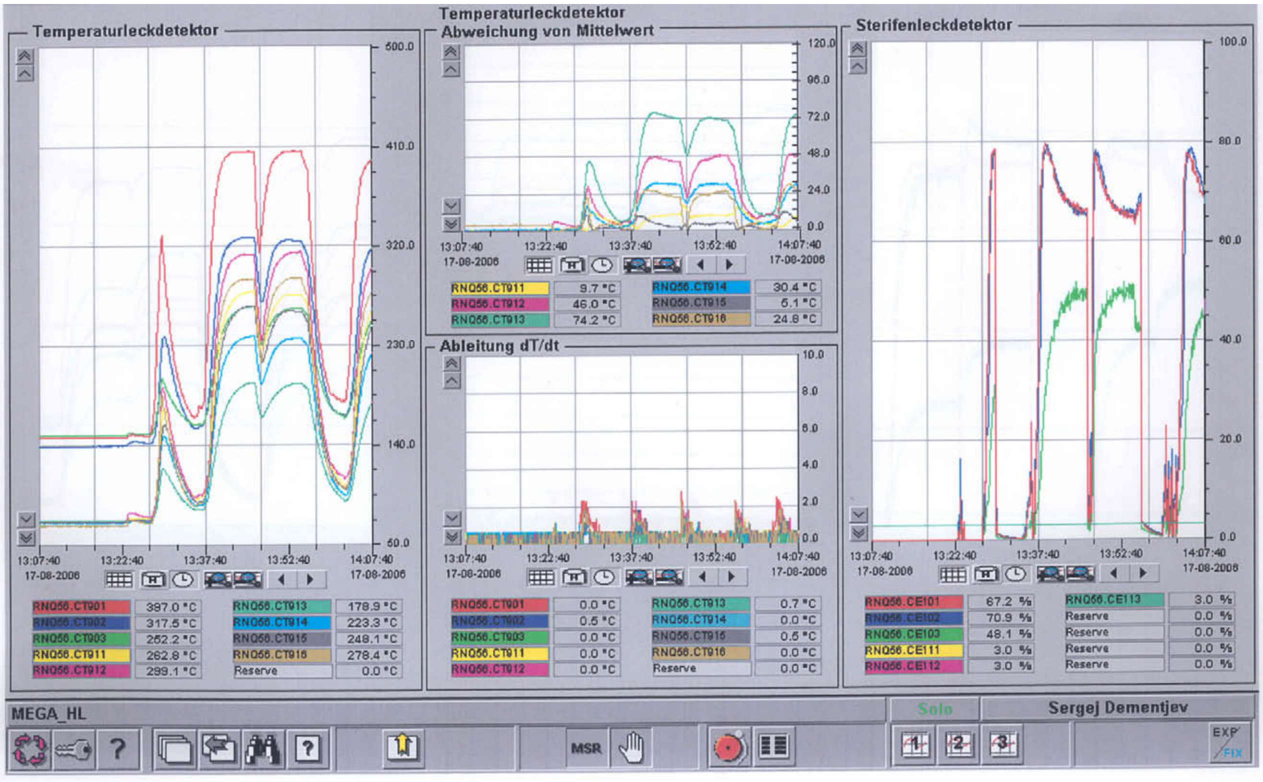
RNQ56.CT901	0.0 °C	RNQ56.CT913	0.0 °C
RNQ56.CT902	0.0 °C	RNQ56.CT914	0.0 °C
RNQ56.CT903	0.0 °C	RNQ56.CT915	0.0 °C
RNQ56.CT911	0.0 °C	RNQ56.CT916	0.0 °C
RNQ56.CT912	0.0 °C	Reserve	0.0 °C

RNQ56.CE101	3.0 %	RNQ56.CE113	3.0 %
RNQ56.CE102	3.0 %	Reserve	0.0 %
RNQ56.CE103	3.0 %	Reserve	0.0 %
RNQ56.CE111	3.0 %	Reserve	0.0 %
RNQ56.CE112	3.0 %	Reserve	0.0 %

Fig. 4. At about 100 μA beam, still on the second day of irradiation, the rising transients of the impedance signals appeared to follow a thermal time course, whereas the trailing edge continued to be much sharper than the temperature decay; compare with temperatures on the left and Fig. 2.

continued to be much sharper than the temperature decay; compare with temperatures on the left and Fig. 2. The impedance sensors had to be temporarily switched off in order to avoid false leak alarms. Before the stripe sensors were enabled again, their sensitivity had been reduced by about a factor of 8.

Fig. 5 shows that at approximately 850 μA on 17 August two of the Ohmic channels indicated fast signal increases and subsequent decay of the readings when the beam was switched on, whereas the third resistive sensor exhibited slower and continued rise in conductivity. The AC-impedance channels had been switched off



RNQ56.CT901	387.0 °C	RNQ56.CT913	178.9 °C
RNQ56.CT902	317.5 °C	RNQ56.CT914	223.3 °C
RNQ56.CT903	252.2 °C	RNQ56.CT915	248.1 °C
RNQ56.CT911	262.8 °C	RNQ56.CT916	278.4 °C
RNQ56.CT912	299.1 °C	Reserve	0.0 °C

RNQ56.CT911	9.7 °C	RNQ56.CT914	30.4 °C
RNQ56.CT912	46.0 °C	RNQ56.CT915	5.1 °C
RNQ56.CT913	74.2 °C	RNQ56.CT916	24.8 °C

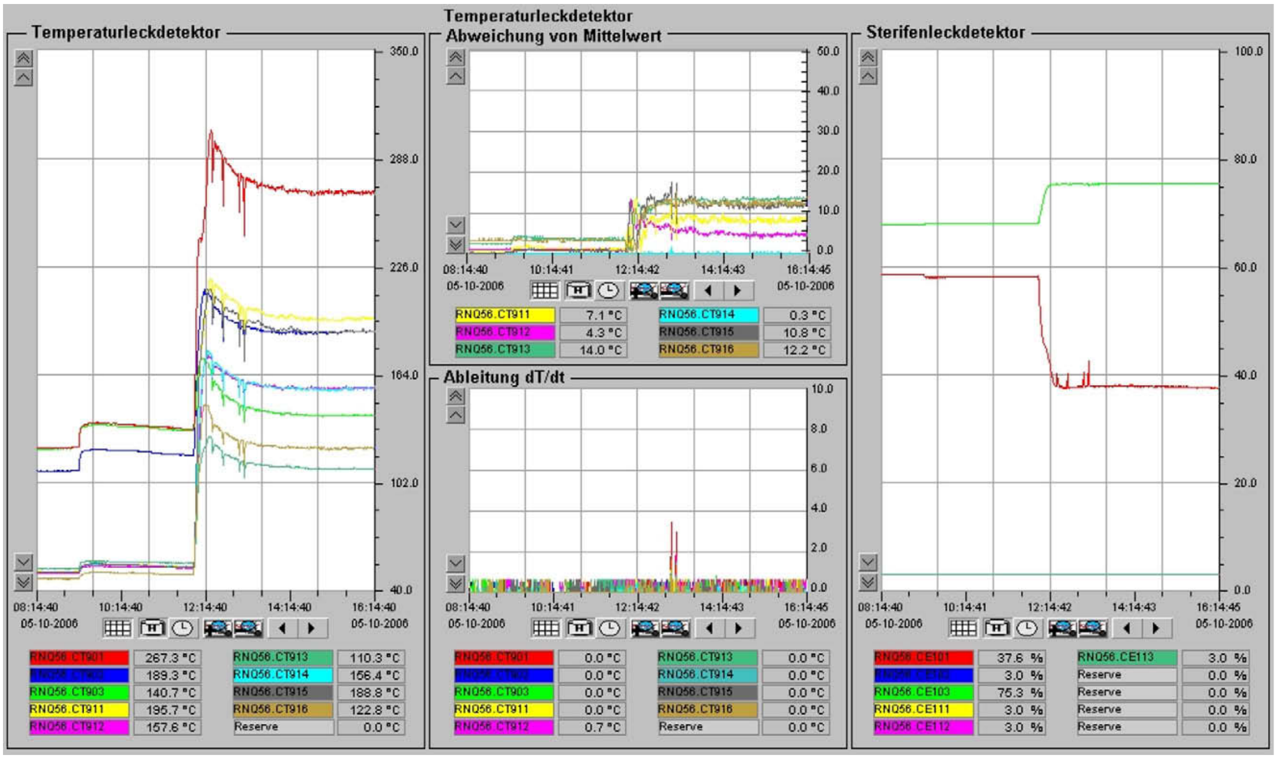
RNQ56.CT901	0.0 °C	RNQ56.CT913	0.7 °C
RNQ56.CT902	0.5 °C	RNQ56.CT914	0.0 °C
RNQ56.CT903	0.0 °C	RNQ56.CT915	0.5 °C
RNQ56.CT911	0.0 °C	RNQ56.CT916	0.0 °C
RNQ56.CT912	0.0 °C	Reserve	0.0 °C

RNQ56.CE101	67.2 %	RNQ56.CE113	3.0 %
RNQ56.CE102	70.9 %	Reserve	0.0 %
RNQ56.CE103	48.1 %	Reserve	0.0 %
RNQ56.CE111	3.0 %	Reserve	0.0 %
RNQ56.CE112	3.0 %	Reserve	0.0 %

Fig. 5. At approximately 850 μA on 17 August two of the Ohmic channels showed fast signal increases and subsequent decay of the readings when the beam was switched on, whereas the third resistive sensor exhibited slower and continued rise in conductivity.

at this time. No correspondence between the temperature distribution around the tip of the nose and the location of the stripes could be found.

Fig. 6 shows that after about 2 months of operation a rather steady behavior of the remaining strip leak detectors emerged. The above example is taken from 5 October during evacuating



RNQ56.CT901	267.3 °C	RNQ56.CT913	110.3 °C
RNQ56.CT902	189.3 °C	RNQ56.CT914	156.4 °C
RNQ56.CT903	140.7 °C	RNQ56.CT915	188.8 °C
RNQ56.CT911	195.7 °C	RNQ56.CT916	122.8 °C
RNQ56.CT912	157.6 °C	Reserve	0.0 °C

RNQ56.CT911	7.1 °C	RNQ56.CT914	0.3 °C
RNQ56.CT912	4.3 °C	RNQ56.CT915	10.8 °C
RNQ56.CT913	14.0 °C	RNQ56.CT916	12.2 °C

RNQ56.CT901	0.0 °C	RNQ56.CT913	0.0 °C
RNQ56.CT902	0.0 °C	RNQ56.CT914	0.0 °C
RNQ56.CT903	0.0 °C	RNQ56.CT915	0.0 °C
RNQ56.CT911	0.0 °C	RNQ56.CT916	0.0 °C
RNQ56.CT912	0.7 °C	Reserve	0.0 °C

RNQ56.CE101	37.6 %	RNQ56.CE113	3.0 %
RNQ56.CE102	3.0 %	Reserve	0.0 %
RNQ56.CE103	75.3 %	Reserve	0.0 %
RNQ56.CE111	3.0 %	Reserve	0.0 %
RNQ56.CE112	3.0 %	Reserve	0.0 %

Fig. 6. After about 2 months of operation a rather steady behavior of the remaining strip leak detectors emerged.

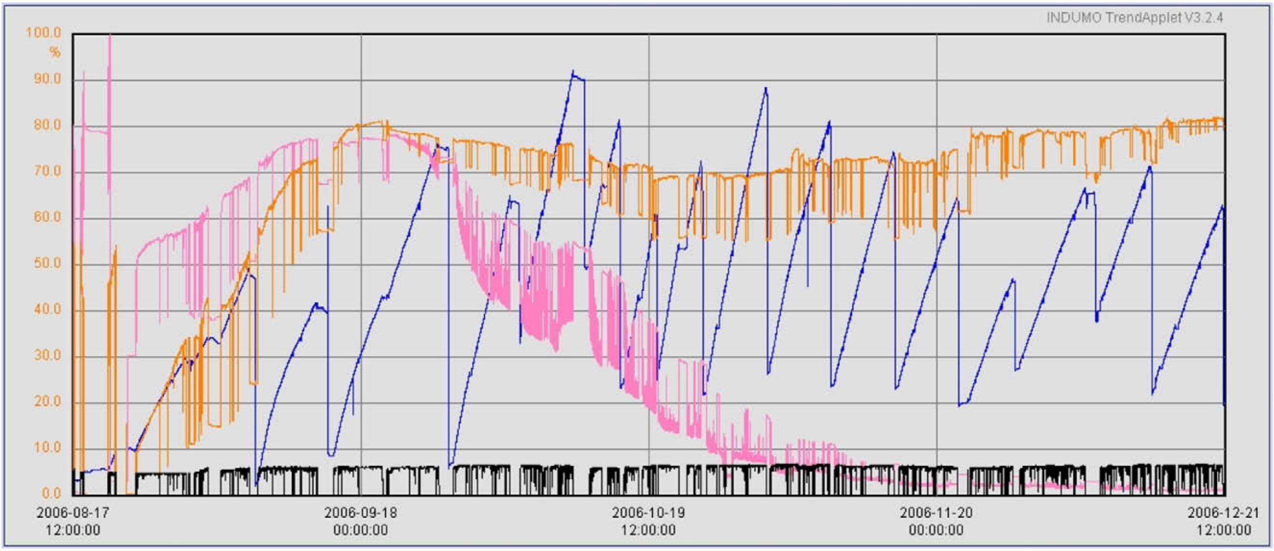


Fig. 7. In an overlay of beam current (black), IG pressure (blue) and the two Ohmic stripe sensor channels, which were operational during the whole irradiation period, the long term evolution of the signals for the whole period of Megapie irradiation. (For interpretation of color mentioned in this figure the reader is referred to the web version of the article.)

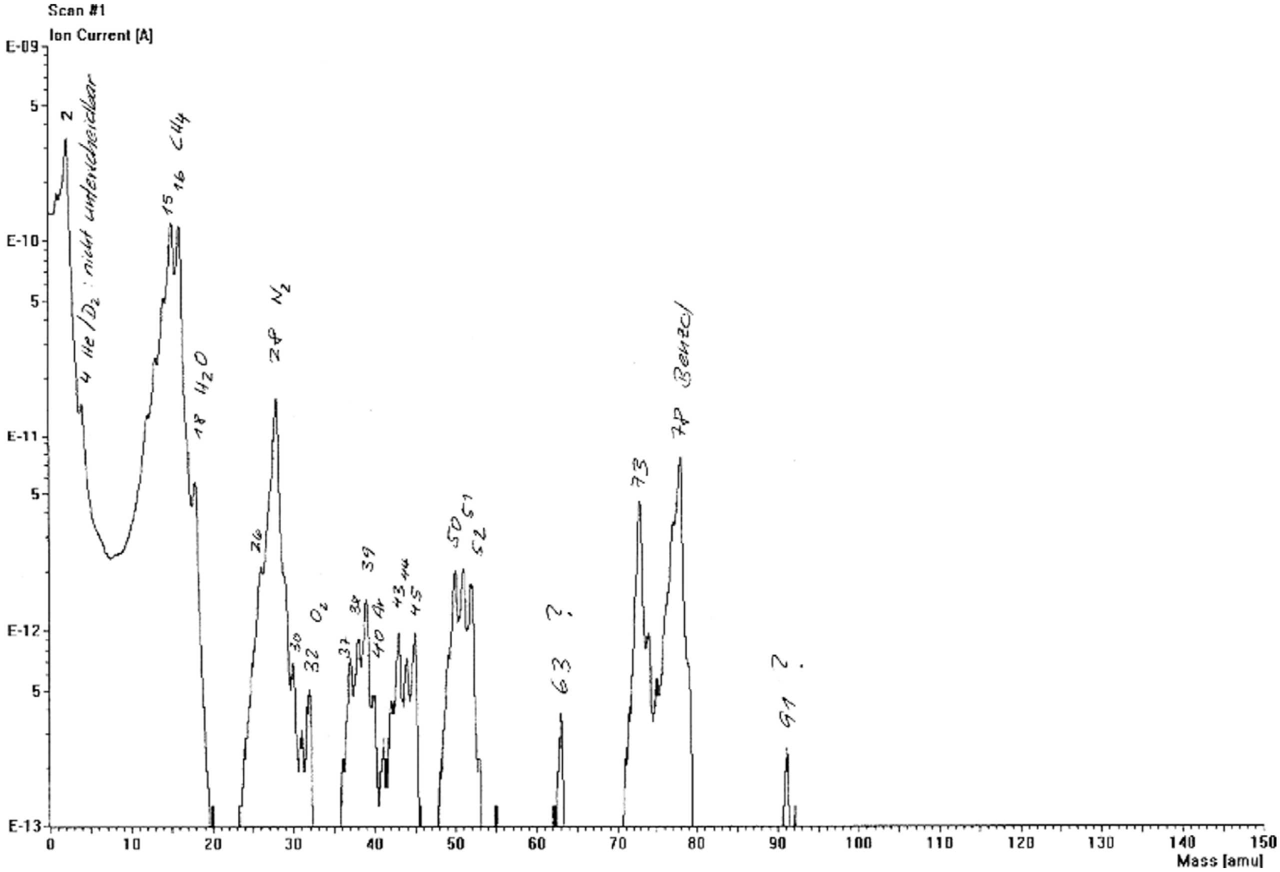


Fig. 8. The mass spectrum of a probe from the IG.

the insulation gas, IG, and switch-on of the beam. Reducing the pressure in the IG volume lead to higher temperatures, especially for the heated thermocouples, as is clearly seen in the leftmost panel (first small step). After irradiation commenced again, a sharp rise in all temperatures was followed by a slow decrease. Later, this has been attributed to effective gas pressure build up, purportedly due to radiolysis of heat transfer oil from a tiny leak of the heat removal system into the IG volume. The then two remaining Ohmic channels of the stripe leak detector featured high baseline conductivity and exhibited inverse responses to the proton beam. Whereas in one channel (green)[1] the conductivity and signal

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[1] For interpretation of color in Figs. 6 and 7, the reader is referred to the web version of this article.

increased under irradiation, the values for the second channel (red) decreased in step.

Fig. 7 shows the long term evolution of the signals for the whole period of Megapie irradiation. In an overlay of beam current (black), IG pressure (blue) and the two Ohmic stripe sensor channels, which were operational during the whole irradiation period. No useful and evident relations were visible other than that with beam on, IG gas was continuously produced. The observed gas production in the IG volume was not anticipated and made the installation of a dedicated gas handling system necessary. At regular intervals the built up IG had to be evacuated into a decay tank. The most likely origin of this IG is radiolysis of heat transfer oil.

Fig. 8 shows the mass spectrum of a probe from the IG. It was determined several times. The observed species were consistent with radiolysis from Dyphil heat transfer oil with a lot of hydrocarbons at masses between 37 and 52.

4. Conclusion

Contrary to expectations, the stripe sensor signals were not only sensitive to leaked liquid metal. Starting immediately with the irradiation of the Megapie target, the purported insulator material ZrO_2 showed significant responses to:

- temperature transients,
- irradiation level & dose,
- gas composition,
- and combinations of the above.

This material is not recommended for use in applications demanding low conductivity in spallation radiation environments, especially not in ill-defined gases. For applications which actually use the response of the ceramics to measure gases, i.e. gas sensors, it appears necessary to verify their behavior in a realistic radiation environment if relevant for the final application.

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[2] K. Thomsen, NIMM A 592 (2008) 476–482.