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Perspective

The effects of Total Ionizing Dose irradiation on supercapacitors deployed in nuclear decommissioning environments☆

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HIGHLIGHTS

- Experimental tests on supercapacitors were carried out using a ^{60}Co irradiator.
- Evaluated if gamma radiation has effect on electrical properties of supercapacitors.
- Testing showed that supercapacitors exhibited no observable effect from irradiation.
- Tested devices are suitable for use in two specific decommissioning environments.

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ABSTRACT

The effects of Total Ionizing Dose (TID) on electrical components is a key parameter to evaluate the life span of wireless sensor nodes for possible deployment in nuclear decommissioning environments. The aim of this study was to experimentally evaluate the effects of TID on capacitance, internal resistance and the self-discharge characteristic of 100 F supercapacitors. An automated test circuit was designed and assembled to charge and discharge the supercapacitors. The supercapacitors were irradiated using a Co-60 γ ray radiation source and the voltage across the supercapacitor terminals, charging current and discharging current were monitored and logged to calculate the capacitance during the irradiation process. Measurements of internal resistance and self-discharge characteristic were performed before and after the irradiation to examine the effects of exposure to γ radiation on these electrical properties. The experimental results show negligible effects on the capacitance of supercapacitors exposed to a maximum dose of 40 kGy. The internal resistance and self-discharge characteristics were not affected by TID up to 89 kGy. These results demonstrate that supercapacitors are a suitable technology to design an Energy Storage System to be deployed in the majority of nuclear decommissioning environments.

1. Introduction

One of the main challenges associated with the decommissioning of nuclear facilities is to reduce the cost associated with monitoring and surveillance operations. For example, the use of sensor networks to monitor waste storage facilities can reduce the cost and time of projects and dose exposure of the workers. In Ref. [1] the need to develop remote techniques to support the monitoring and characterization of areas with high radiation levels were identified. Wireless sensing systems can be deployed in nuclear environments to monitor the plant during decommissioning operations and to replace legacy instrumentation. Early deployments of Commercial Off The Shelf (COTS) wireless instrumentation on the Sellafield site to monitor the pressure and temperature of steam lines have demonstrated the benefits associated with this technology [2]. The use of Wireless Sensor Networks (WSNs) in harsh environments is well documented in the literature [3–5], and [6].

Much research in recent years has focused on energy harvesting systems to reduce the battery replacement operations and hence extend the lifetime of WSNs powered by scavenging energy from the environment [7,8]. However, the solutions presented in the literature can

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provide only an intermittent and limited power output, which is not adequate to meet the power requirements of typical sensor nodes [9]. In Refs. [10] supercapacitor technology was identified as a better energy storage device for WSN applications compared to battery technologies. Supercapacitors address a specific requirement to limit the use of rechargeable lithium batteries in nuclear decommissioning environments, and hence, reduce fire and explosion hazards [11,12]. Few researchers have investigated the effects of high radiation levels on the electrical properties of supercapacitors for deployment in nuclear decommissioning environments. Their tolerance to radiation is a fundamental parameter in providing a sufficient and predictable operational life span. However, several studies have investigated the damage that radiation can have on general electronic components [13]. Recent works have focused on the damage caused by Total Ionizing Dose [14–16]; Neutron Displacement [17–19] and Single Event Effects [17,20,21]. Whilst there may be some neutron emissions in a nuclear decommissioning environment, it is the effects of Total Ionizing Dose (TID), produced by the energy deposited in the electronics by γ radiation, that will produce the most significant damage to electronic components [14].

Laghari et al. (1990) evaluated the radiation effects on the electrical properties of chemical double-layer capacitors exposed to γ radiation (TID of 75 kGy), thermal neutrons (fluence of 4.5×10^{14} n/cm^2), and fast neutrons (fluence of 1.75×10^{13} n/cm^2) in the reactor core. This study found that whilst the electrical properties of the capacitors were temporarily affected during irradiation, these properties fully recovered post-irradiation [22].

Further research has focused on the effects that radiation has on the capacitance and equivalent series resistance of supercapacitors. For example, Shojah-Adalan et al. described the effects of γ radiation and proton irradiation on three different supercapacitors, manufactured by Maxwell Technologies, with capacitance values of 10, 100, and 1200 F [23]. The capacitance and equivalent series resistance of these capacitors were measured following γ exposure of up to 2 kGy TID and proton TID of up to 20 kGy and found to be unaffected. However, the charging mechanism of one of the supercapacitors was damaged after exposure to 10 kGy.

Although this study demonstrated that capacitance and equivalent series resistance are not affected by TID effects, there has been limited research on capacitance measurements during irradiation and self-discharge properties of supercapacitors exposed to radiation levels typically experienced in nuclear decommissioning radioactive environments. The self-discharge characteristic is particularly relevant when the supercapacitor is being used for a long-term energy storage device in a wireless sensor system. In the present study, irradiation experiments were performed on Maxwell supercapacitors to evaluate the effect of TID on the capacitance during the irradiation. Also, the self-discharge and internal resistance characteristics were monitored, before and after the irradiations. The results provide valuable information to guide the design of energy storage systems for WSN. In particular, this work verifies the use of supercapacitors as a suitable technology for deployment in nuclear decommissioning environments.

2. Experimental methodology

The current investigation involves the experimental evaluation of TID effects on capacitance, internal resistance and self-discharge properties of COTS supercapacitors. BCAP0100 supercapacitors manufactured by Maxwell Technologies were selected to perform the experiments, using supercapacitors from lot number W170410702. To simulate the conditions that would be present in a nuclear decommissioning environment, the experiments were carried out at the Dalton Nuclear Facility using a ^{60}Co irradiator. The irradiator is a FTS Model 812 self-contained γ irradiator and emits two γ rays with energies of 1.17 and 1.33 MeV resulting in an average beam energy of 1.25 MeV [24].

Whilst in a decommissioning environment other γ emitters, such as ^{137}Cs are likely to be present, the energies of these γ rays will be lower, and hence they will have a slightly less damaging effect on electronic components. Consequently, ^{60}Co irradiators have become a standard γ ray radiation source to perform experiments to evaluate the effects of TID [25] and was therefore considered to be a suitable testing environment for this study. The MIL-STD-883K standard identifies uniform methods, controls and procedures for testing microelectronic devices suitable for use within military and aerospace environments. This standard suggests using a ^{60}Co γ ray source with a dose rate between 0.5 and 3 Gy/s [26].

Studies presented in Ref. [27] have shown that electronic components in nuclear decommissioning environments are exposed to different dose rates and total dose rates compared to other industrial applications. These findings need to be taken into account during the selection of dose rate and total dose to evaluate the effects of TID on components deployed in nuclear decommissioning environments.

The work presented in this paper focuses on two real-life scenarios: a medium level sludge storage facility with a dose rate of 4.5×10^{-1} Gy/h and hot spots in a Pressurized Water Reactor with a dose rate up to 2 Gy/h [28]. For both these cases γ ray radiation sources are the main challenge, and the contribution of neutron displacement damage is negligible.

A total of 9 supercapacitors were irradiated and analysed, by adapting the test method described by the British Standards (BS EN 62391) to evaluate electrical characteristics of fixed electric double-layer capacitors [29]. The availability of the irradiation facility limited the sample size of the experimental investigation.

The block diagram, displayed in Fig. 1, shows the automated measurement system that was designed to measure the capacitance and internal resistance whilst the supercapacitor under test is charged and discharged, following the adapted testing method. The constant current load was implemented using IRF510 MOSFET and LM10C operational amplifier/voltage reference. This sub-circuit was isolated from the supercapacitor's terminal block via relay 2. The charging and discharging current was monitored using two LT6105 precision current sense amplifiers. Both the voltages from the current sense amplifiers and the supercapacitor were connected to three different operational amplifiers OPA140 to buffer the signal.

Fig. 2 shows the block diagram of the circuit that was designed to perform the self-discharge experiments. The components to charge the supercapacitors is the same as described above. In addition, relay 4 was included between the supercapacitor and the operational amplifier OPA140 to isolate the supercapacitor from the data acquisition unit during the experiments and avoid any further discharge.

The methodology to evaluate the electrical characteristics used in this work was a modified version of BS EN 62391 adapted to the specific case of a supercapacitor of 100 F capacitance and maximum operating voltage of 2.7 V. A National Instruments USB-6009 data acquisition unit was connected to the charging and discharging circuit and to a laptop running LabVIEW software. LabVIEW software was designed to control the relays during the experiments and the logging of the voltage across the supercapacitor under test, charging current, and discharging current, was performed using a National Instruments Technical Data Management Streaming (TDMS) file. The data was graphically shown in real time with a sample period of 100 ms.

The measurement consists of three main stages: set the maximum voltage to 2.7 V and start to charge the supercapacitor with 1 A constant current, when the maximum voltage value is reached continue the constant voltage charging for 300 s and then discharging the supercapacitor using a constant current load set at 1 A.

3. Experimental results and discussion

Three supercapacitors were used in each experiment to evaluate the TID effects on capacitance and were mounted on a PCB and positioned

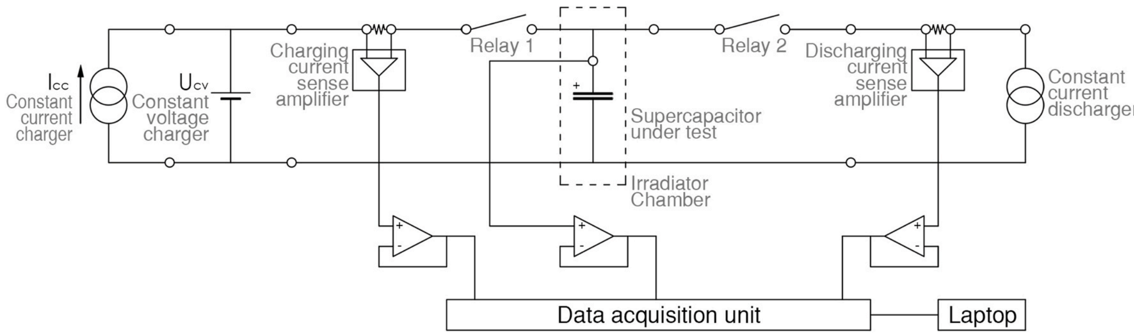

Fig. 1. Block diagram of capacitance and internal resistance measurement circuit.

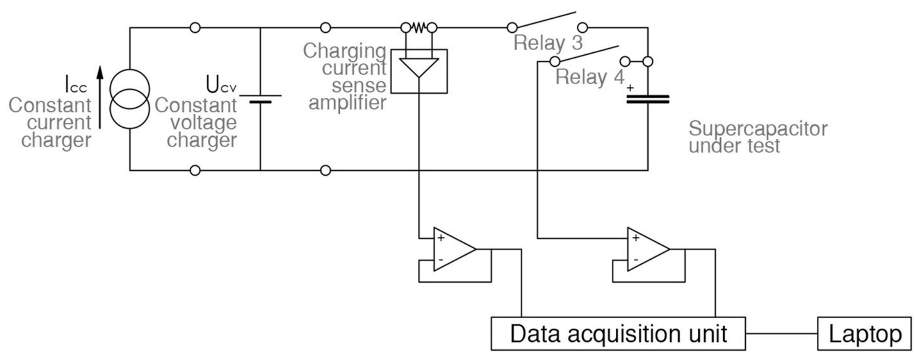

Fig. 2. Block diagram of self-discharge measurement circuit.

inside the irradiator chamber.

As shown in Fig. 3 (a) the device under test was connected to the charging and discharging circuit using two wires feeding through the irradiator access port. The advantage of having the charging and discharging circuit outside the irradiator is to limit the effects of TID on other components. For example, it has been demonstrated that operational amplifiers [30], power MOSFETs [31], and voltage regulators [32] are affected by γ radiation. Before mounting the supercapacitor inside the ionizing chamber, the dose rate was experimentally measured using an ionization chamber, with a result of 2.07 Gy/s, which is within the range suggested by the MIL-STD-883K standard.

Fig. 4 illustrates the voltage-time characteristics, charging and discharging current during the experiments to evaluate capacitance and internal resistance.

Capacitance was calculated according to the BS EN 62391 using the following equation:

$$C = I_d \frac{t_2 - t_1}{U_1 - U_2} \tag{1}$$

where:

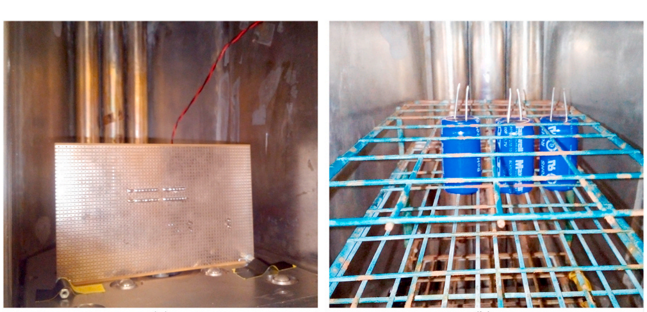

Fig. 3. Supercapacitor positioned inside the irradiator during capacitance test (a) and supercapacitors positioned using irradiator's rack (b).

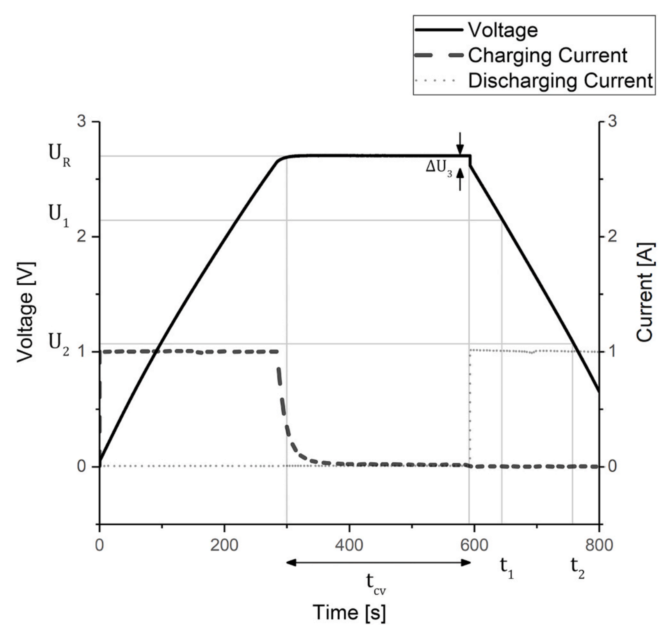

Fig. 4. Experimental measurement of voltage/time characteristics between supercapacitor terminals in capacitance and internal resistance experiments.

C is the capacitance of supercapacitor [F]
I_d is the discharge current [A]
U_1 is the measured start voltage [V]
U_2 is the measured end voltage [V]
t_1 is the time at which the terminal voltage of the capacitor reaches the value U_1 from the start of the discharge [s]
t_2 is the time at which the terminal voltage of the capacitor reaches the value U_2 from the start of the discharge [s];

Experiments to evaluate the internal resistance and self-discharging characteristics before and after irradiation were performed on two different groups of supercapacitors for a total irradiation time of 16 h and 4 h. Supercapacitors 4 to 9 were irradiated during two different irradiation tests, as shown in Fig. 3 (b).

Fig. 5 presents the results for capacitance measurements obtained using equation (1) to evaluate the effects of TID during the irradiation. As seen, similar behaviour was observed in all cases, with negligible effects up to a total dose of 40 kGy.

Although it is evident that the results are in good agreement with the manufacturer's nominal value of 100 F with a 20% tolerance [33], additional experiments were carried out to estimate other potential sources of measurement error during the experiments. For example, the temperature of the irradiation chamber was measured to increase from ambient room temperature to 40 °C during the first two hours of irradiation. As a result, the effect of temperature on the capacitance of supercapacitor was investigated. Supercapacitors 1 to 3 connected to the charging and discharging circuit by two wires were placed inside an oven, following the same experimental arrangement developed for irradiation experiments. The voltage across the supercapacitors, the charging current and discharging current were monitored for 5 h to evaluate capacitance changes during the test. A second evaluation was completed to assess if the results of capacitance measurements were affected directly by the tolerances and self-heating effects of the components employed in charging and discharging circuits. Supercapacitors 1 to 3 were charged and discharged in the laboratory environment at ambient temperature for five hours, and then the capacitance was calculated to evaluate if the initial calculated value had undergone any change during the experiment.

Fig. 6 illustrates the comparison between the effects of TID, temperature and test circuit components. As can be seen in the figure, the three plots are very similar. All the experiments show a variation of the initial capacitance value compared to the value calculated after 5 h within the range 1.3%–2.2%. This result demonstrates the small changes in the capacitance values were unlikely to be the result of irradiation. To the best knowledge of the authors, the results of TID effects on capacitance during the irradiation are novel. The 3 supercapacitors did not show any temporary changes to the capacitance during the irradiation test, this result could suggest that the dielectric constant of the electrolyte is not affected.

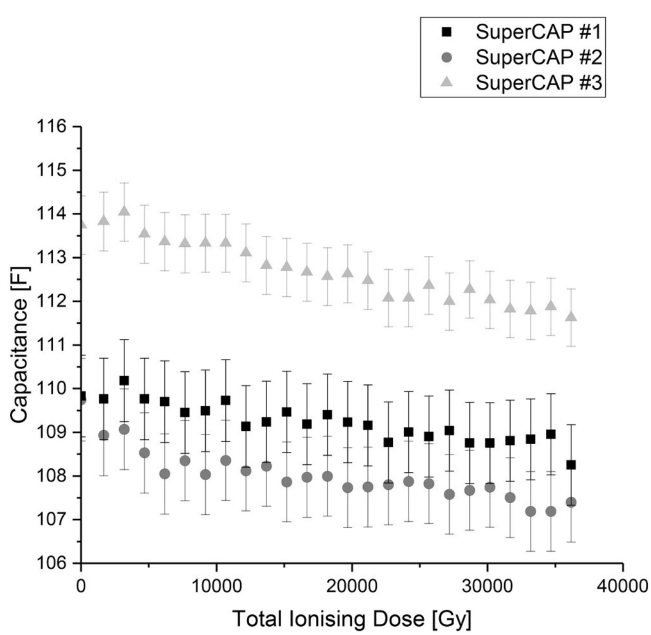

Fig. 5. Total Ionizing Dose effects on capacitance characteristic.

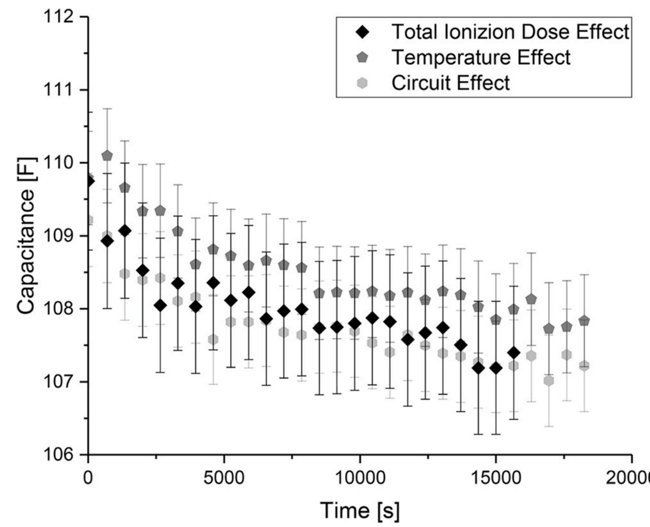

Fig. 6. Comparisons of TID, temperature and circuit effects on capacitance characteristic.

Internal resistance was calculated according to BS EN 62391 using the following equation:

$$R = \frac{\Delta U_3}{I_d} \tag{2}$$

where:

R is the internal resistance of supercapacitor [Ω]
ΔU_3 is the difference of voltages between the calculation start voltage and the set value of constant voltage charging [V]

Table 1 shows the results for the internal resistance irradiation test with a TID up to 29 kGy and 89 kGy. The findings on irradiation effects for the internal resistance of supercapacitors extends to those in literature, confirming that no effects have been observed up to a dose of 89 kGy.

Fig. 7 shows the voltage between the supercapacitor terminals measured during this study to evaluate the self-discharge characteristic of the supercapacitors. The supercapacitor was charged at 1 A constant current up to the rated voltage of 2.7 V, then continuing to charge at 2.7 V constant voltage for 300 s and at the end disconnecting the supercapacitor from the circuit. During the experiments, the voltage across the terminals of supercapacitor was monitored every 12 h for 72 h.

The self-discharge parameter was calculated according to the following equation:

$$U_{s-d} = U_{end} - U_R \tag{3}$$

where:

U_{s-d} is the self-discharge characteristic [V]

Table 1
Internal resistance before and after irradiations.

Supercapacitor under test	TID[kGy]	R[Ω]	
		Irradiated	Non Irradiated
SuperCAP#4	27	0.045	0.046
SuperCAP#5	29	0.045	0.047
SuperCAP#6	29	0.045	0.046
SuperCAP#7	83	0.045	0.045
SuperCAP#8	89	0.047	0.047
SuperCAP#9	89	0.047	0.048

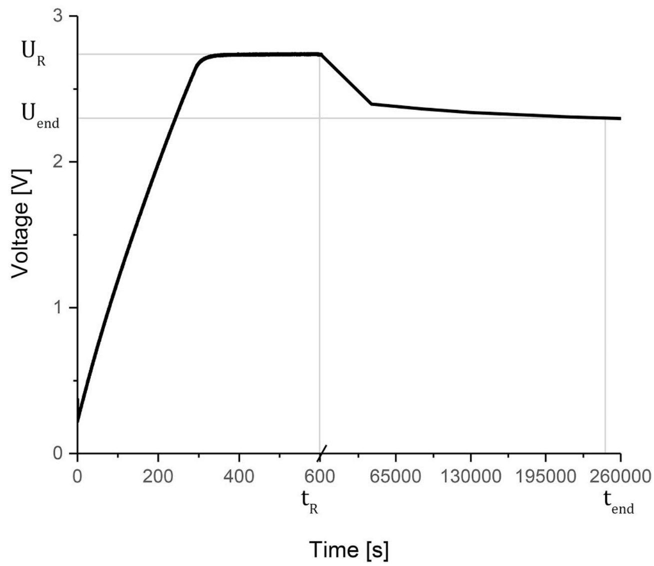

Fig. 7. Experimental measurement of voltage/time characteristics between supercapacitor terminals in self-discharge characteristic experiments.

Table 2
Self-dischage characteristic before and after irradiations.

Supercapacitor under test	TID [kGy]	Self-Discharge[V]	
		Irradiated	Non Irradiated
SuperCAP#4	27	0.0496	0.481
SuperCAP#5	29	0.0442	0.413
SuperCAP#6	29	0.0450	0.396
SuperCAP#7	83	0.554	0.531
SuperCAP#8	89	0.583	0.518
SuperCAP#9	89	0.565	0.523

U_{end} is the voltage between open capacitor terminal at the end of the experiment [V]
U_R is the rated voltage [V]

Finally, the results on the self-discharge characteristic of supercapacitors are reported in Table 2. The self-discharge characteristic is a fundamental parameter to evaluate the energy stored by the supercapacitor exposed to γ radiation during deployment. The values present the changes in voltage before and after the irradiation calculated with equation (3). The voltage increase in the supercapacitor after irradiation at 27 kGy and 89 kGy ranged from 0.015 V to 0.065 V.

4. Conclusion

This paper has investigated the effect of TID on the electrical properties of supercapacitors to determine their suitability for use in two specific nuclear decommissioning environments. The first set of experiments evaluated the effects of gamma radiation on capacitance when the devices were irradiated with a dose of 40 kGy, with a second experiment examining whether the internal resistance and self-discharging abilities of the supercapacitors changed before and after being irradiated with a dose of 89 kGy. The results of these experiments indicated that the electrical properties of the supercapacitors showed no observable effects both during and after irradiation.

The TID that the supercapacitors were exposed to in the experiments is relatively high for a nuclear decommissioning environment, and it can therefore be concluded that the particular supercapacitors that were tested could have an operational life span up to 10 years within a medium level sludge storage facility and up to 2 years if deployed close to a hot spot in a Pressurized Water Reactor under decommissioning.

CRediT authorship contribution statement

Antonio Di Buono: Conceptualization, Methodology, Investigation, Writing - original draft. **Neil Cockbain:** Conceptualization, Supervision, Writing - review & editing. **Peter R. Green:** Conceptualization, Methodology, Supervision, Writing - review & editing. **Barry Lennox:** Supervision, Writing - review & editing, Funding acquisition.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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