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ON THE CATION EFFECT ON THE DISTRIBUTION OF OXIDATION
STATES FOLLOWING RADIATIVE-NEUTRON CAPTURE REACTIONS ON
AN ANIONIC SPECIES

Vincent C. Anselmo (1)
Department of Chemistry
New Mexico Highlands University
Las Vegas, New Mexico 87701

MASTER

ABSTRACT

Calculations were performed on the influence of the nuclear reactions of the cation on the oxidation states observed following radiative-neutron capture on a particular salt where the anionic species is under scrutiny. Any radiation damage from photons evolved because of the exoergicity of the reaction, or from nuclear decay reactions, including also beta decay, are negligible and should not cause any radiation effect, at least for the irradiation condition described.

INTRODUCTION

There is unquestionable evidence that the cation exerts some influence on the observed oxidation states of an anionic species following radiative-neutron capture. For example, in the phosphate system the retention as orthophosphate increases as one increases the atomic weight of the cation in the group IA alkali metal orthophosphates (2). In this report I show the influence one parameter, the capture reaction on the cation, on the experimental observation mentioned above, via self-irradiation effects. Specifically, the two nuclear reactions in question are the Q of the reaction, and the beta and/or gamma decay of the daughter nucleus. The following assumptions were made: one millimole (approximately 100 mg) was taken as the sample size; a representative 15 minute irradiation at 2.5×10^{12} n/cm² - s was taken as typical; in a deliberate oversimplification of the case, it was assumed that all of the energy of decay was deposited in the sample. This last assumption also permitted a conversion of the energy into rads for comparison with the literature.

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241

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RESULTS AND DISCUSSION

Using these assumptions, and the literature values for cross-sections, the number of nuclear events was first calculated and then, using the calculated Q , the total energy liberated for all the capture reactions in the size sample taken was determined (Table I). The total energy dissipated for the daughter decay reactions was then calculated (also Table I), where it was assumed that the decay was completed during the time of irradiation, handling, and separation of the sample, or else a simplifying assumption was made with respect to the fraction of the original nuclides that had decayed. The energy was finally converted to Mrads with the assumption described above.

For comparison with the literature, Getoff (3) reports on experiments showing retention as phosphate as a function of gamma irradiation (expressed in Mrads) from a ^{60}Co source. He shows that it takes about 1 Mrad to produce any change in the percent orthophosphate in irradiation orthophosphate. Working with this number, and even if one assumes that all the photons from the Q of the capture reaction are absorbed, then only in the case of the (n, α) reaction on ^6Li does the total energy exceed the 1 Mrad limit deduced from Getoff's experiments. However, if the assumption that all the energy is absorbed is now neglected in favor of something more realistic, it may be concluded that there is insufficient gamma energy from this source to produce any radiation annealing.

A similar conclusion may be drawn from the photon emission as a result of decay of the daughter. The most energetic reaction is the decay of ^{86}mRb where the total photon energy is calculated as 4.4×10^{12} Mev (less than the 4.1×10^{14} Mev for the Q of the (n, α) reaction on ^6Li). The energy of the beta emission can also be significant, and here one would expect greater energy absorption in the sample than for gamma emission, but the total energy is still less than the energy received on the assumption that all the energy is absorbed. The daughter isotope, ^{86}Rb , emits a total of 7.0×10^{12} Mev after one half-life (18.8 d), a number that is again comparable to but less

than the energy dissipated in the ${}^6\text{Li} (n, \alpha) {}^3\text{H}$ reaction. A number that may be used for comparison is available from Claridge and Maddock (4) wherein they report on the effect of 1.8 Mev electrons, at doses of 50 Mrad and 100 Mrad, on the distribution of activities in neutron irradiated KPF_6 . Assuming that 1 Mrad are at least necessary to provide sufficient radiation damage to be observed, the same conclusions may be drawn as with the photon irradiations, i. e., the beta ray dose is insufficient to cause radiation damage under the conditions specified.

In conclusion, it can be stated that in the case of nuclear reactions on the cation, the Q of the reaction, as well as the beta and/or gamma decay of the daughter nuclides, do not provide sufficient energy to produce radiation damage, under the conditions specified. It is likely that a much longer irradiation time would be enough to cause damage in the solid. However, there is another effect that was not discussed here that could account for the cation effect, and that is the solution process. It is also possible that other effects, such as particle size and degree of hydration, are related to the cation effect on solution and its ultimate effect on the distribution of products. Further work is planned on the cation effect on the solvent, e. g., by a study of the pH of the solution, or by an investigation into the formation of radiation products, such as H_2O_2 , among others.

Table I

SUMMARY OF THE Q OF THE NUCLEAR REACTION INDICATED, AND
THE TOTAL ENERGY OF BETA AND/OR GAMMA DECAY OF
THE DAUGHTER NUCLIDE

Nuclear Reaction	Number of Product Nuclides (x 10 ⁻¹⁰) ^(a)	Q (Mev x 10 ⁻¹⁰)	Energy ^(b) in Mrads	Total β -Decay Energy ^(c) (Mev x 10 ⁻¹⁰)	Total γ -Decay Energy (Mev x 10 ⁻¹⁰)
⁷ Li (n, γ) ⁸ Li	3.9	2.04	0.01	5.1	0
⁶ Li (n, α) ³ H	8400	4.78	65 ^(e)	(d)	(d)
²³ Na (n, γ) ²⁴ Na	65	6.96	.72	91	180
⁴¹ K (n, γ) ⁴² K	10	7.52	.0018	36	15
⁸⁵ Rb (n, γ) ⁸⁶ Rb	782	8.58	.16	700	0
⁸⁵ Rb (n, γ) ^{86m} Rb	782	-	-	0	440
⁸⁷ Rb (n, γ) ⁸⁸ Rb	4.0 ³	6.3	.12	21	7.6
¹³³ Cs (n, γ) ^{133m} Cs	360	6.8	.054	0	47

(a) Calculated from the percent abundance of the parent nuclide, the cross section for the reaction, and assuming a neutron flux of 2.5×10^{12} n/cm² - s, and a 15 minute irradiation.

(b) Assuming that all of the energy from the Q of the reaction is absorbed in a 100 mg sample.

(c) The total energy was calculated assuming all the daughter nuclides have decayed, except for ⁸⁶Rb where only one-half were assumed to have decayed, with known decay mode and energy.

(d) This does not contribute a significant quantity of energy because of its long half-life.

(e) Equivalent to 4.1×10^{14} Mev.

REFERENCES

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- (4) Claridge, R. F. C. and A. G. Maddock, Radiochim Acta, 6, 80 (1963).