Journal of Radioanalytical Chemistry, Vol. 61, No. 1–2 (1981) 295–305

MEASUREMENTS OF HARD BETA EMITTERS BY CERENKOV DETECTOR

E. HEDRICH, F. GRASS

Institut für Anorganische Chemie der Universität Wien und Atominstitut Wien (Austria)

(Received August 6, 1980)

Boron and lithium content in various glass fiber-optics, enamel, and lake sediments were determined by measurement of the Cerenkov-radiation of ^{12}B and ^{8}Li using the fast rabbit-system of the Atominstitute. The decay-curves were also measured using a dead time correction unit (LFC) and evaluated with respect to interferences of other short-lived nuclides. Simultaneously, γ-spectra of short-lived nuclides were measured with the LFC-system. Main interferences in Cerenkov-counting were observed from ^{28}Al and ^{16}N. High count-rates were measured correctly with the LFC-system. The halflives of ^{8}Li and ^{12}B were determined as well as their detection limits in glass and lake-sediments.

Introduction

Nondestructive analytical methods, such as NAA, are especially important for materials which are difficult to dissolve, e.g. glasses and geological samples. The determination of Li and B by NAA is only possible by means of their short-lived hard β-emitting isotopes ^{8}Li and ^{12}B – preferably measured by Cerenkov counting[1,2] – as there are no longer-lived isotopes or isomeric states. To demonstrate the applicability of this method, some examples of Li- and B-measurements in glass and lake sediments are given. The determinations were of practical interest. The boron content of glass and enamel is essential for their quality, and we felt that the determination of Li in lake sediments might permit rapid characterization of the layers.

Nuclear reactions

Besides ^{12}B and ^{8}Li, other hard β-emitters are also produced from matrix-elements of glass and geological samples and have to be considered as possible interferences in the B- and Li-determinations. The nuclear data are summarized in Table 1 and show the β-interferences, of which all but ^{24m}Na can be avoided by ap-

Table 1
Nuclear data of B, Li and interfering elements

Reaction	σ_{th}, b	σ_{fast}, b	τ	E_β, MeV	β-Decay, %
$^{11}B(n,\gamma)^{12}B$	0.0055	–	20.41 ms[10]	13.37, 4.44, 7.65	97.1, 1.33, 1.27[3]
$^{23}Na(n,\gamma)^{24m}Na$	0.4	–	20.12 ms[8]	6	weak[4]
$^{10}B(n,\alpha)^{7}Li$	3837	–	–	–	–
$^{7}Li(n,\gamma)^{8}Li$	0.037	–	844 ms	12.5	~100%[5]
$^{11}B(n,\alpha)^{8}Li$	–	0.14	844 ms	12.5	~100%[5]
$^{16}O(n,p)^{16}N$	–	0.039 (14 MeV)	7.13 s	4.3, 10.4	68, 26[6]
$^{27}Al(n,\gamma)^{28}Al$	0.23	–	2.246 m	2.9	100[4]

propriate discrimination or corrected by computation after the measurement, owing to the differences in halflife. ^{24m}Na has the same halflife as ^{12}B and is also a hard β-emitter, so that a correction would be difficult to apply, if this transition were not emitted with low probability. Though we have not yet studied the cross-over sensitivity for ^{24m}Na on a systematic basis, we were able to show that in certain experimental setups we had only a negligible correction for Na. This is somewhat odd, as we definitely have one for ^{16}N and ^{28}Al. We assume that owing to the special setup of the Cerenkov counter used, the cross-over sensitivity of ^{28}Al is mainly due to the Compton effect of the γ-transitions[7] and not to its β-rays, but for ^{16}N there may be an additional component from the 10.4 MeV β-transition.

Experimental

Activation and measurement

Samples of about 0.1 to 0.3g and standards are sealed in PE-vials and activated for 1 or 2s at a neutron flux of 1.25 10^{12} $cm^{-2} \cdot s^{-1}$ and a Cd-ratio of 12. Owing to the short halflife, it is necessary to use the fast rabbit system of the Atominstitut with a transfer time of 20 ms. The Cerenkov detector system consists of a large acrylic glass block 100 X 100 X 250 mm which concentrically surrounds the slowing down device for the sample and acts as a radiator. The Cerenkov light is registered by a photomultiplier equipped with a high-current voltage divider, feed-

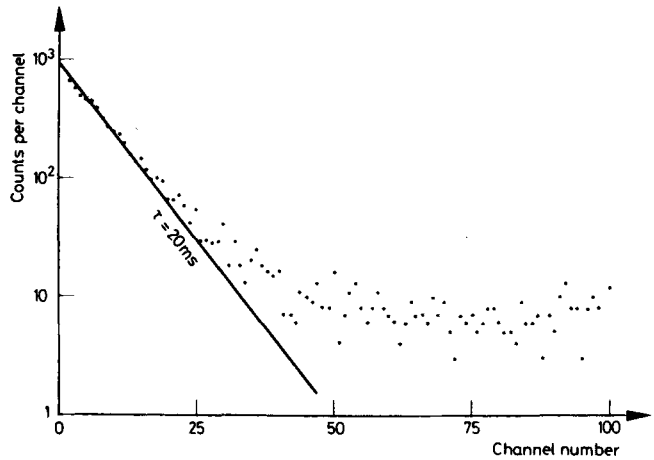

Fig. 1. Decay-curve of boron standard, 6.57 mg B, t_{irr} = 2 s, t_m = 4 ms/Ch, no LFC

ing the negative high voltage to the cathode. To prevent light emitted from the sample and sample container[8] from reaching the photocathode, the outer surface of the slowing down device is blackened to make an effective absorber of 4 to 5 mm acrylic glass before a β-particle can enter the radiator. The pulses from the photomultipliers (EMI 9514B, 6097B, 9778B, RCA 4525, RCA 8055) are fed via an amplifier and a discriminator to a 4000 channel analyzer working in the multi-scaling mode. Alternatively, a Loss Free Counting system in the shift register version[9] allows measurement of high activity samples. In general, the counting time per channel is about 1/5 – 1/10 of the halflife of the resp. nuclide and decay-curves are detected over 2000 or 4000 channels. Simultaneously, the γ-spectra can be measured with a Ge(Li)-detector and the LFC system in the last version.[10]

Results

Decay-curves and half-lifes

Some typical decay-curves of B- and Li-standards and of samples of glass containing a high amount of B and of a lake sediment containing Li are shown in Figs 1–5. The B-standard-curve shows (Fig. 1) a rapid decrease at the beginning of the curve according to a half-life of 20 ms, after that there remains a small but significant increase over background level, caused by ^{8}Li from the ^{11}B(n, α)^{8}Li reaction. Similarly the glass-sample (Fig. 2) at first decays very rapidly due to

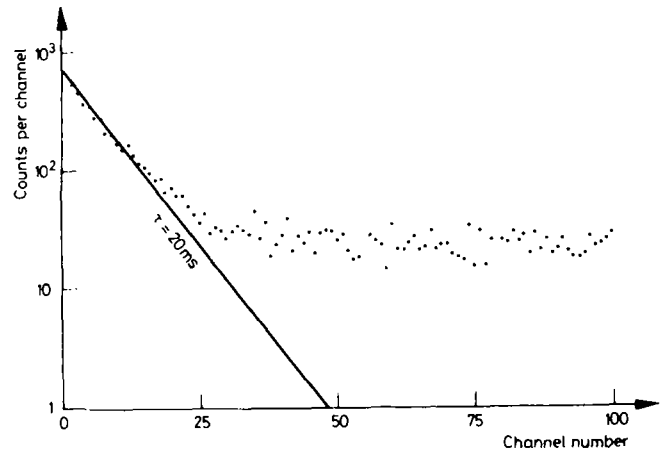

Fig. 2. Decay-curve of fibre optic glass containing boron, 141mg glass, t_{irr} = 2 s, t_m = 4 ms/Ch, no LFC

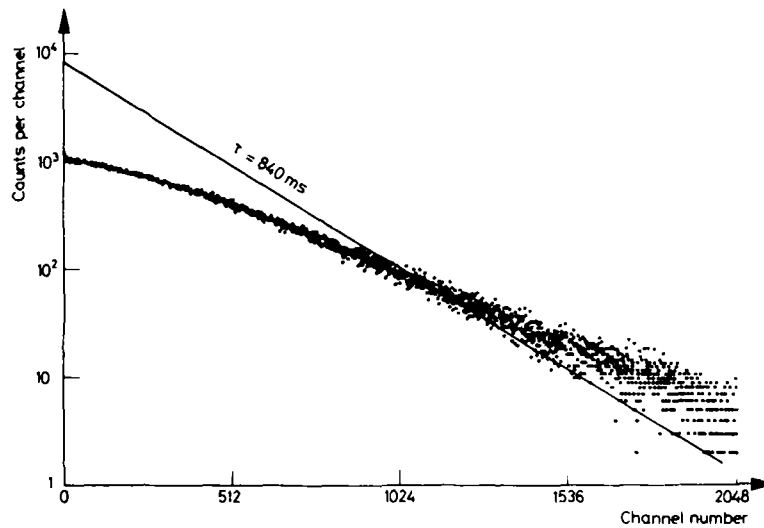

Fig. 3. Decay-curve of lithium standard, 1.03 mg Li, t_{irr} = 2 s, t_m = 4 ms/Ch, no LFC

^{12}B. ^{28}Al is detected despite a high discriminator level and obscures that existance of ^{8}Li. The Li-standard curve of about 1 mg Li (Fig. 3) shows strong deadtime effects at the beginning of the curve when measured without the LFC-system whereas no deadtime-effects can be observed in the curve measured with it (Fig. 4).

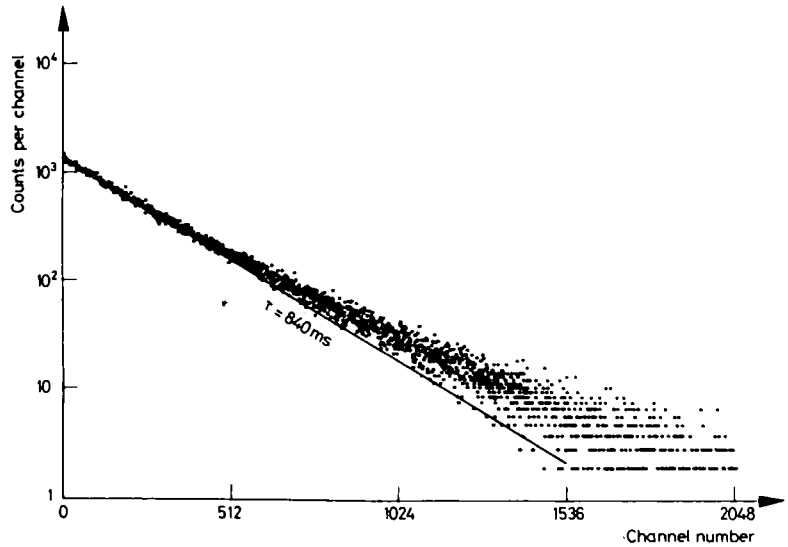

Fig. 4. Decay-curve of lithium standard, 1.03 mg Li, t_{irr} = 2 s, t_m = 4 ms/Ch, with LFC

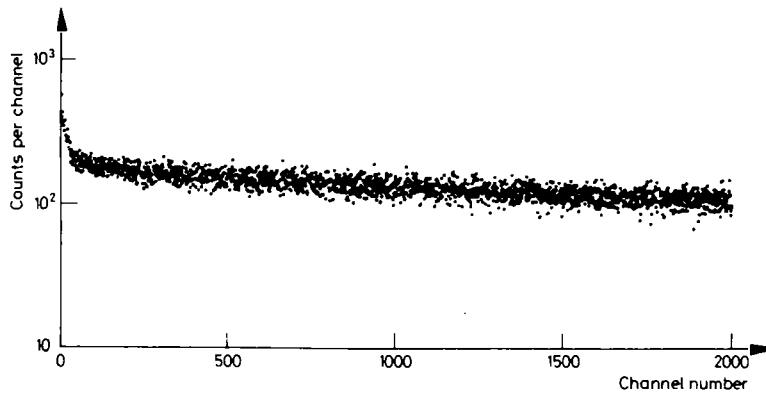

Fig. 5. Decay-curve of lake sediment, 362 mg sediment, t_{irr} = 2 s, t_m = 80 ms/Ch, no LFC

The halflife of ^{8}Li determined graphically from curves with 100 μg Li-content and without deadtime effects is 840 ms and application of an exponential curve-fitting programme for the calculation of halflives gives a value of 844 ms.[11] The curve of the lake sediment (Fig. 5) shows the decay of ^{8}Li followed by the slower decay of ^{28}Al which again obscures the part with ^{16}N. The fact that ^{16}N is produced at our activation position and that it has to be considered in the evaluation of the decay-

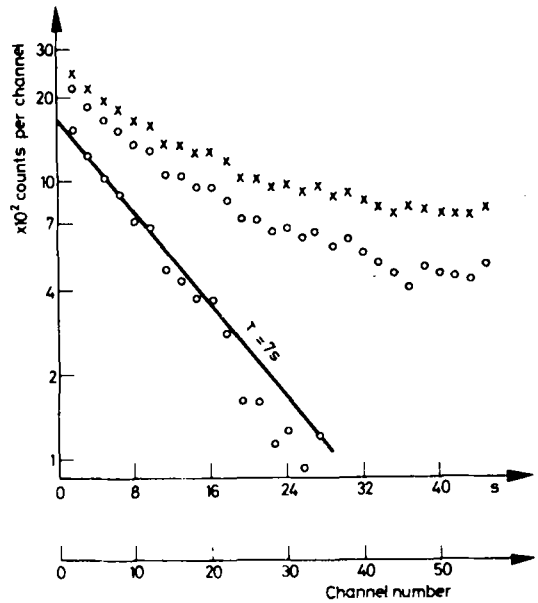

Fig. 6. Decay-curve of quartz, 250 mg quartz, $t_{irr} = 7$ s, $t_m = 800$ ms/Ch, no LFC; x decay-curve of quartz, o corrected for background and ^{28}Al

curves can be shown from the decay-curve of a pure quartz sample (Fig. 6). In this there remains a nuclide with $\tau = 7$ s, i.e. ^{16}N after deduction of background and an ^{28}Al contribution.

Calculation of initial count rates

According to the law of radioactive decay for a multicomponent decay

$$\Sigma N_i = \Sigma N_{oi}\, e^{-\lambda_i t_{di}} (1 - e^{-\lambda_i t_{mi}}) \lambda_i^{-1}$$

where N — count rate,
N_o — initial count rate,
t_d — decay time,
t_m — measurement time,
λ — decay constant,

the initial count rates of a composed decay-curve can be calculated easily provided that the differences of halflives allow a complete decay of the shorter-lived nuclides.

Table 2
Decay-, measurement-times, typical count rates and initial count-rates

	Boron			Lithium		
	^{28}Al	^{8}Li	^{12}B	^{28}Al	^{16}N	^{8}Li
t_d(s)	14	0.24	0.004	72	8.4	0.24
t_m(s)	2	0.42	0.080	24	7.2	0.80
Typical counts	7200	950	3500	6900	580	3000
Correction for Al		1600	300		3000	360
f. Li resp. N (counts)		–	250		–	190
Initial count rate (cps)	3700	3300	$1.46 \cdot 10^5$	450	254	6200

As this applies to the case of our glasses and sediments, a computer programme HEDPR was established, where for selected decay and measurement times the respective initial count rates as well as the correction necessary are computed. An example for boron and lithium is given in Table 2.

Calibration

Standards of boric acid and Li-citrate were used to avoid interfering elements. A boron calibration curve is shown in Fig. 7. Linearity in the range of 0.4 to 10 mg B is typical for measurements without LFC. The lithium calibration curves are linear up to 2 mg Li if measured with LFC. Without LFC, correct measurements are possible only up to about 100 μg. A typical curve is shown in Fig. 8.

Gamma-Spectra

Gamma-spectra were measured simultaneously with the Cerenkov measurement. An example of a lead-containing enamel for pottery and of a sediment sample from Neusiedler See is shown in Figs 9 and 10. 4 consecutive spectra with increasing measurement times were measured in both cases. The enamel clearly shows large amounts of ^{207m}Pb in the first two spectra, in the two others increasing amounts of ^{28}Al and ^{56}Mn. Nuclides determined in the sediment sample were ^{24m}Na which decays completely during the first measurement, ^{46m}Sc and ^{179m}Hf in the second and third spectrums and finally ^{52}V, ^{56}Mn and large amounts of Al, which decay only very slowly.

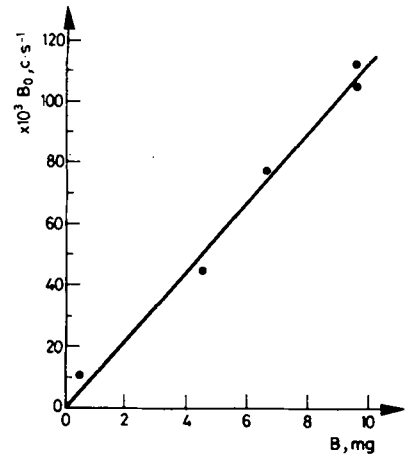

Fig. 7. Calibration-curve of boron, $t_{irr} = 2$ s, $t_m = 4$ ms/Ch

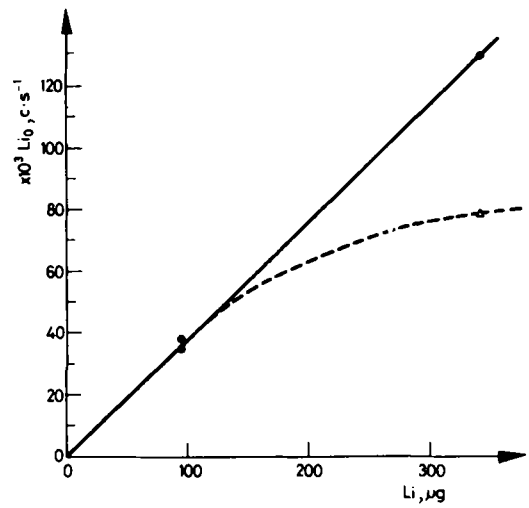

Fig. 8. Calibration-curve of lithium, $t_{irr} = 2$ s, $t_m = 4$ ms/Ch, • with LFC, △ no LFC, normalized

Concentration of B and Li in samples

Boron was determined in various fibre optic glasses as well as in enamel samples. The values are in the range of 0.6 to 8% B, the detection limit – depending on the contribution of other nuclides to the underground – is about 0.1 to 0.3 mg B,

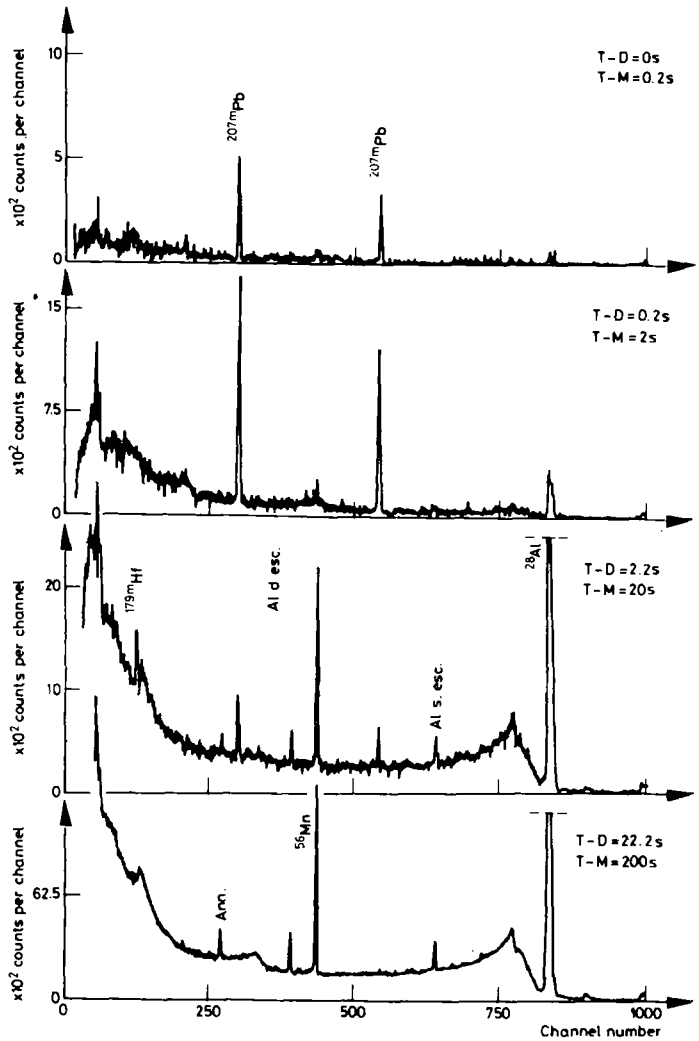

Fig. 9. γ-Spectra of short-lived nuclides of pottery enamel $t_{irr} \approx 2$ s

the standard deviation of replicate analyses is about 0.2 mg B, if the samples are activated in steady-state operation.

Lithium was determined in a large number of sediment samples of Neusiedler See as well as in 2 geological standard materials. The concentration range of the sediment is 20–90 ppm Li, the values of the geological standard material correspond

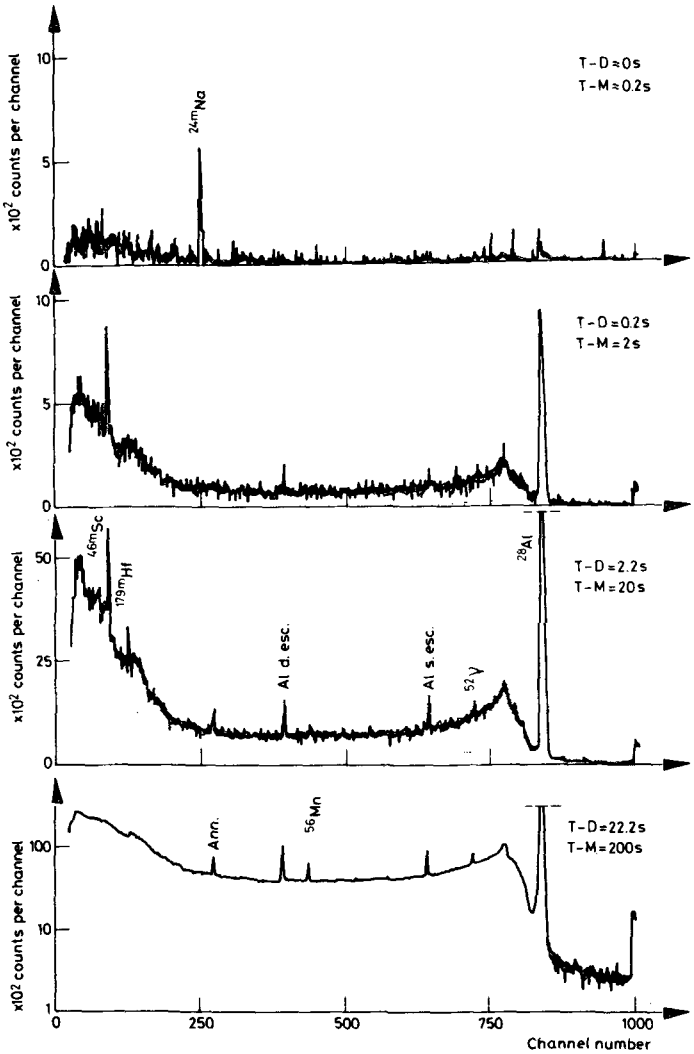

Fig. 10. γ-Spectra of short-lived nuclides of a lake sediment $t_{irr} = 2$ s

well with the reported values, the detection limits in lake sediments calculated as standard deviation of replicate sample determinations is 2–3 ppm Li due to the contribution of Al to the underground using steady-state irradiation. The boron and lithium values are given in Table 3. Pulsing the reactor would enhance the sensitivity by a factor of 640 for boron and 34 for Li.

Table 3
Concentration of boron and lithium in glass, enamel and sediments

Sample	Concentration	Standard deviation	Other short lived nuclei
Fibreoptics			
AO	0.7% B	0.1–0.3 mg B	Pb, Na
AR	1.7% B		–
CE	7.8% B		–
D	2.8% B		–
SS	<0.2% B		n.d.
Quartz	<0.2% B		n.d.
Enamel			
LS	0.6% B	0.1–0.3 mg B	Na, Pb
SG	5.2% B		–
DB	1.6% B		Na, Pb
WM	2.3% B		–
Sediments of Neusiedler See	20–90 ppm Li	2–3 ppm Li	^{24m}Na, ^{179m}Hf, ^{46m}Sc
Geological standard			
UG GS G2	33 ppm Li	3 ppm Li	reported value: 34.9 ppm[12]
UG GS GSP1	28.5 ppm Li	2 ppm Li	reported value: 32 ppm

*

We wish to express our thanks to E. SEYMANN for writing the computer programme HEDPR.

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