ESTIMATION OF GAS PURITY IN A CO_2-FILLED PROPORTIONAL COUNTER BY RISE-TIME ANALYSIS

ADAM MICHCZYŃSKI and ANNA PAZDUR

Radiocarbon Laboratory, Institute of Physics, Silesian Technical University
Bolesława Krzywoustego 2, PL-44-100 Gliwice, Poland

ABSTRACT. Since 1994, a new data-acquisition system with rise-time (RT) based background reduction has been in operation at the Gliwice Radiocarbon Laboratory. During long-term stability tests, we observed large shifts in the RT spectra obtained during various measurement series. These shifts caused important changes in the count rate measured with RT reduction. As a result, we have ascertained that the shifts are the consequence of variable concentration of electronegative impurities. A correction method was considered, using average RT of pulses obtained in coincidence with the muon events, so-called ARTL (average rise time of L pulses), for estimation of the RT of the spectrum's shift. However, radiocarbon dates calculated based on count rates with RT discrimination, which were corrected using this method, are not more precise than traditional ^{14}C dates. Nevertheless, the value of the average coincidence pulse RT is a good indication of sample gas purity.

INTRODUCTION

Since 1994, a new data acquisition system with rise-time (RT) based background reduction has been in operation at the Gliwice Radiocarbon Laboratory. The system uses microprocessor-controlled pulse and coincidence analyzers for data acquisition from three proportional counter sets, called L1, L2 and L3. Background is reduced by the rejection of pulses with rise times greater than the optimally selected value (discrimination level). The details of the new system and the background-reduction method were presented in Michczyński *et al.* (1995). We expected that the background reduction by rise-time (RT) discrimination would increase the accuracy of ^{14}C dates, especially for old samples. However, during long-term stability tests we observed large shifts and changes of a shape of the RT spectra obtained from various measurement series. These changes are particularly distinct for the spectra of coincidence events, which are generated by cosmic radiation (high-energy muons). We can also observe similar shifts and changes in the anticoincidence spectra of the background. Due to these changes, the count rate measured with pulse-rise discrimination varies according to the position and shape of the RT spectrum. These variations are so large that they call into question the possibility of a background reduction by RT discrimination.

RESULTS AND DISCUSSION

The most important reason for the changes described above may be differences in the concentration of electronegative impurities. Theoretical analysis and simulations show that an increase in these impurity concentrations causes a shift in the RT spectrum of cosmic muons to the direction of smaller values (Robinson 1994). Figure 1 shows RT spectra of coincidence events obtained by the L1 counter during three series of measurements, when a low value of C—*i.e.,* the counting efficiency and gas purity control parameter (Pazdur, Walanus and Mościcki 1978)—indicated a high concentration of electronegative impurities.[1] Figure 1 also presents four examples of RT spectra of coincidence counts obtained during background measurements, and the spectrum acquired when the high value of C allows us to assume that counting gas was very pure. Table 1 contains values of the parameter C for the series of measurements, which correspond to the spectra presented in Figure 1. The RT spectra for

[1]Parameter C is defined as a quotient of the number N of coincident muon counts between the two sections of guard counters by the number L of coincident counts between the proportional counter and the whole guard (C=L/N). The ratio C does not depend on muon flux and gives information about counting efficiency. In particular, this parameter indicates electronegative impurities when its concentration has an effect on counting efficiency.

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three series, when the impurity concentration was high, are shifted to a shorter RT, whereas the spectrum obtained for very pure gas is shifted to a longer RT. This confirms our statement that variations in gas purity cause the changes in the observed RT spectra.

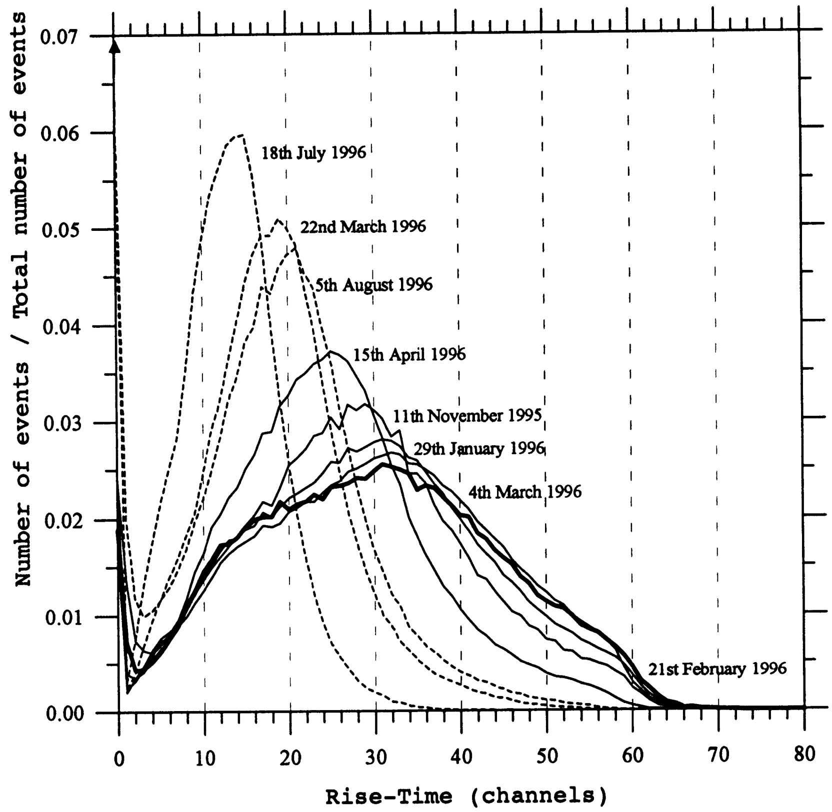

Fig. 1. Rise-time spectra of coincidence events (muon counts) from proportional counter L1. Spectra obtained during the measurement sessions, when a high concentration of electronegative impurities was detected (low value of gas purity and counting efficiency control parameter—C) are represented by dashed lines. The spectrum obtained during the measurement session, when the C-value was high (we can assume that gas was very pure) is represented by a thick line. Thin lines represent some of the background spectra (1 RT channel = 0.4 μs).

To make this conclusion more reliable, we also analyzed the dependence of the changes of RT spectra of coincidence pulses (muon events) upon the corrected voltage, defined as supply voltage divided by working pressure. We described the variation in position and shape of the RT spectra using the average RT (calculated in RT-channels), which we denoted as ARTL (average rise time of L pulses). In our laboratory the supply voltage is automatically changed by computer during the counting period to stabilize the working point of the counter. The working point is controlled by dividing the muon pulse height spectrum into two channels and observing the ratio of pulses recorded in the upper and lower channels. When the impurity concentration in the counter gas increases, the control system also increases the voltage. Hence, the value of the proper voltage gives us information about the concentration of electronegative impurities. Because a similar effect may be caused by changes in counter gas pressure,[2] instead of voltage we use corrected voltage as a measure of the concentration of electronegative molecules. This idea has been applied in our lab for gas purity control (Goslar *et al.* 1990).

[2]The gas pressure differences in the counter are limited to a narrow range of a few percent of the standard working pressure.

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TABLE 1. Values of gas purity and counting efficiency control parameter C for the measurement sessions, which correspond to the spectra presented in Figure 3. Values of average RT for coincidence counts (ARTL) are also given.

Date	C	ARTL
Measurements with low C-value (presence of electronegative impurities)		
18 July 1996	1.402	11.16
22 March 1996	1.611	17.17
5 August 1996	1.584	18.95
Background measurements		
15 April 1996	1.643	23.95
11 November 1995	1.627	27.57
29 January 1996	1.631	29.03
4 March 1996	1.666	30.15
Measurements with high C-value (very pure gas)		
21 February 1996	1.665	29.65

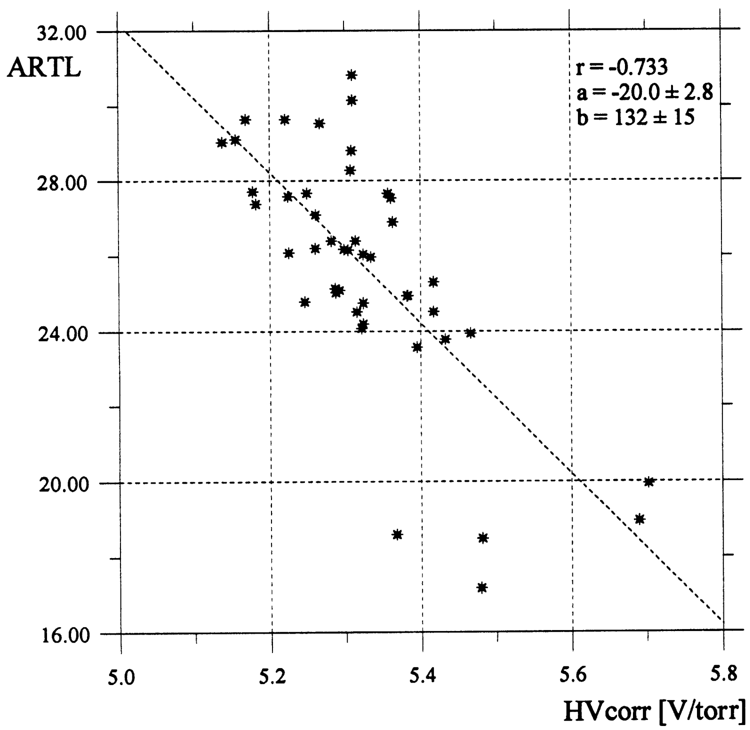

Fig. 2A. Values of average rise-time of coincidence counts (ARTL) *vs.* pressure-corrected voltage

The results of our analysis are presented in Figure 2A, showing the average RT of coincidence counts (ARTL) for counter L1 from measurements of background, modern standard and few contaminated gases. We note an approximate linear dependence of the ARTL value upon the corrected voltage, which confirms that the shifts and the changes in shape of the RT spectra are the consequence of different concentrations of electronegative impurities. Therefore, the ARTL value may be treated as a parameter for gas purity control. Comparison with the parameter currently used (C value) shows that the ARTL better detects the concentration of electronegative impurities (Fig. 2B).

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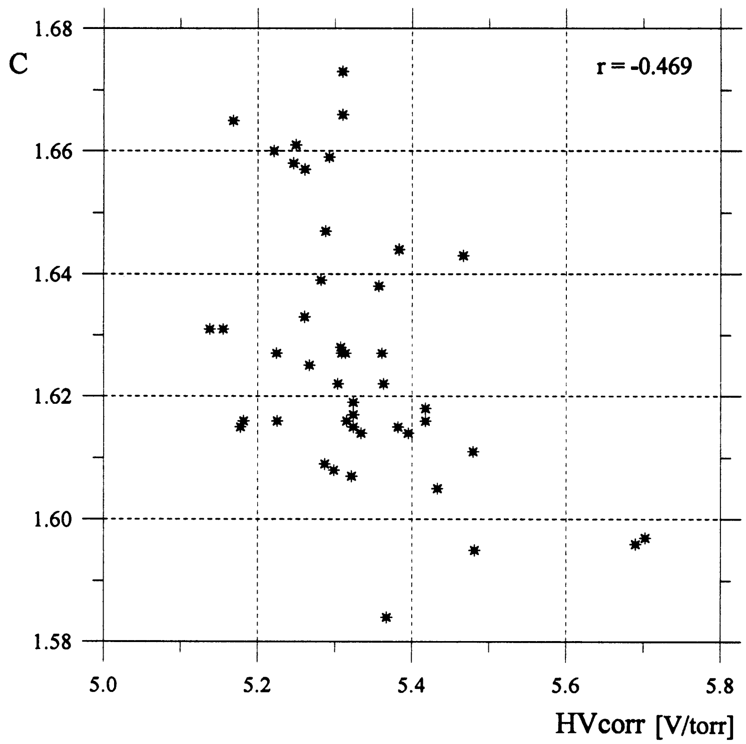

Fig. 2B. Values of C parameter *vs.* pressure corrected voltage

To compensate for the influence of gas purity on the count rate measured with pulse-rise discrimination, we developed a method to correct for observed changes in the count rate. Changes in the background count rate after RT discrimination, as well as changes of the modern standard count rate after RT discrimination, depend linearly on ARTL values (Fig. 3). Therefore, we can assume that the count rates of background and standard after RT discrimination depend linearly on the ARTL value. This gave us the opportunity to correct the count rate of background and standard based on the measured value of ARTL. Hence, for each measured sample, we calculate the corrected values of the count rate of background $B_{red}(ARTL)$ and modern standard $S_{0red}(ARTL)$ using the formulas

$$B_{red}(ARTL) = B_{red0} + a_{B\_ART} \cdot (ARTL - ARTL_0) \quad (1)$$

$$S_{0red}(ARTL) = S_{0red0} + a_{S\_ART} \cdot (ARTL - ARTL_0) \quad (2)$$

where: ARTL = average RT determined for the measured sample
$ARTL_0$ = value of ARTL assumed as a reference value (pure gas)
B_{red0} = background count rate after RT discrimination, meas. for $ARTL=ARTL_0$ (pure gas)
S_{0red0} = count rate of standard after RT discrimination meas. for $ARTL=ARTL_0$ (pure gas)
a_{S_ART}, a_{S_ART} = constants determined during calibration meas. of a proportional counter set

These values are then applied, together with the count rate of a sample, for calculating the ^{14}C age.

Table 2 shows a comparison of ^{14}C dates obtained with and without RT discrimination. Dates calculated based on count rates with RT discrimination were corrected using the new gas-purity control parameter, ARTL, as described above. Table 2 shows that results agree with each other;[3] however, ^{14}C dates calculated with RT discrimination are not more precise than traditional ^{14}C dates.

[3]Differences between dates determined based on count rates with and without RT discrimination are smaller than standard deviations multiplied by 3.

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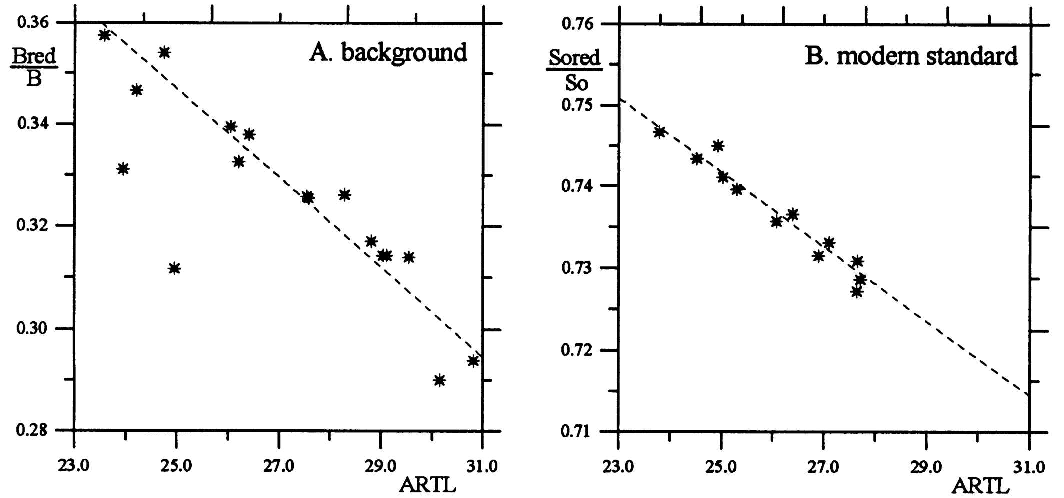

Fig. 3. Relative background reduction (3A) and relative decrease of count rate for a modern standard (3B) after rise-time discrimination *vs.* average RT of coincidence counts (ARTL)

TABLE 2. ^{14}C dates determined based on count rates with and without RT discrimination. Dates calculated based on count rates with RT discrimination were corrected using the ARTL gas-purity control parameter. Distinct differences appear when very low ARTL values indicate relatively high concentration of electronegative impurities (in italics).

Date (yr–mo–dy)	Lab no.	Sample	^{14}C age (yr BP)	^{14}C age determined with RT discrimination (yr BP)
96–03–05	Gd-7742	CAH-94/13	1550 ± 60	1580 ± 60
96–03–06	Gd-7742	CAH-94/13	1520 ± 60	1580 ± 60
96–03–12	Gd-7744	Zat.Pucka W-2	12,000 ± 140	12,180 ± 120
96–03–19	Gd-7746	MORI-1	8880 ± 100	9150 ± 100
96–03–23	*Gd-7750*	*OM24/94*	*34,600 ± 1500*	*35,500 ± 2800*
96–03–26	*Gd-7751*	*Frakcja C18/B1*	*2140 ± 60*	*1960 ± 120*
96–03–27	Gd-7752	Frakcja C18/B2	3300 ± 70	3390 ± 80
96–03–28	Gd-7752	Frakcja C18/B2	3240 ± 70	3460 ± 80
96–03–29	*Gd-7753*	*Frakcja C5/B1*	*110 ± 60*	*220 ± 100*
96–03–30	*Gd-7754*	*Frakcja C31/B1*	*1450 ± 60*	*1570 ± 90*
96–05–09	Gd-7769	BHP (15-20)	1040 ± 50	1010 ± 50
96–05–28	Gd-7778	Pgh-10	200 ± 60	270 ± 60
96–05–31	*Gd-7779*	*BHP-50*	*1450 ± 80*	*880 ± 120*
96–06–15	Gd-7785	Chełmża 6/95	950 ± 50	810 ± 80
96–06–18	Gd-7786	Szynwałd 11/95	1070 ± 60	920 ± 70
96–06–20	Gd-7787	Klasztorek 1/95	750 ± 60	670 ± 80
96–06–21	Gd-7787	Klasztorek 1/95	890 ± 50	850 ± 70
96–07–13	Gd-7791	BG-C6/14-25	2400 ± 60	2360 ± 60
96–07–15	Gd-7791	BG-C6/14-25	2310 ± 40	2340 ± 40
96–07–24	Gd-7793	CHR 10/94 (0.80)	6990 ± 90	7040 ± 110
96–08–06	Gd-7795	TIRI-I	11,190 ± 120	11,130 ± 120

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Conclusion

Although the proposed correction method allows us to compensate for the influence of the gas purity on the count rates with RT discrimination and to obtain proper values of ^{14}C age, the accuracy of the obtained ^{14}C dates does not increase. Therefore, this correction method cannot be used effectively. Rather, we should improve the gas purity system to insure a better and more stable purity of CO_2 for obtaining more precise ^{14}C dates with RT discrimination.

On the other hand, the results show that the value of the average RT is very sensitive to electronegative impurities and the ARTL parameter detects this concentration better than the parameter used up to now. Considering the advantage of the ARTL parameter over C, we will apply ARTL for regular gas purity control.

Acknowledgments

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References

Goslar, T., Pazdur, A., Pazdur, M. F., Walanus, A. and Zastawny, A. 1990 Stanowisko licznika L2 używane w pomiarach ^{14}C o podwyższonej dokładności (Setup of proportional counter L2 for ^{14}C measurements with improved precision). *Zeszyty Naukowe Politechniki Śląskiej, Seria Matematyka-Fizyka, Geochronometria* 6: 83-89.

Michczyński, A., Goslar, T., Pazdur, A. and Pazdur, M. F. 1995 A data acquisition system for proportional counters at Gliwice. *In* Cook, G. T., Harkness, D. D., Miller, B. F. and Scott, E. M., eds., Proceedings of the 15th International ^{14}C Conference. *Radiocarbon* 37 (2): 781–787.

Pazdur, M. F., Walanus, A. and Mościcki, W. 1978 A method of continuous examination of counting efficiency during measurements of natural radiocarbon by CO_2 filled proportional counter. *Nuclear Instruments and Methods in Physics Research* 151: 541–547.

Robinson, S. W. 1994 The response characteristic of gas proportional counters. Poster presented at the 15th International ^{14}C Conference, Glasgow, Scotland, 15–19 August, 1994.

https://doi.org/10.1017/S0033822200017975 Published online by Cambridge University Press