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NEW LIMITS ON 2β PROCESSES IN ^{40}Ca AND ^{46}Ca BY USING LOW RADIOACTIVE $\text{CaF}_2(\text{Eu})$ CRYSTAL SCINTILLATORS

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The development of highly radiopure $\text{CaF}_2(\text{Eu})$ crystal scintillators has been performed aiming at a substantial sensitivity enhancement of the 2β decay investigation and of the search for dark matter particles with spin-dependent (SD) interaction. The results of $\text{CaF}_2(\text{Eu})$ background measurements and simulation are presented. New and highly improved $T_{1/2}$ limits on the 2β decay of ^{46}Ca and the double electron capture of ^{40}Ca are obtained.

Совершенствование сверхчистых кристаллических сцинтилляторов $\text{CaF}_2(\text{Eu})$ велось с целью повышения чувствительности регистрации процессов 2β -распада и поиска частиц темной материи со спинзависимым взаимодействием. Представлены результаты моделирования и измерения фона сцинтилляторов $\text{CaF}_2(\text{Eu})$, а также новые экспериментальные пределы для 2β -распада ^{46}Ca и двойного электронного захвата ^{40}Ca .

INTRODUCTION

The investigations of the neutrinoless (0ν) double β decay (which violates the lepton number conservation) is a powerful tool to search for information leading to possible new physics beyond the standard model [1, 2]. Moreover, at present, the $0\nu 2\beta$ decay is also considered a powerful test for different extensions of the standard model (including several SUSY models), which could offer not only complementary but — in some cases — competitive and superior results than other running or forthcoming accelerator and nonaccelerator experiments [2–4].

The most sensitive results in the search for the 2β decay have been obtained by the so-called «active source» technique; in this case, the detector, which contains the 2β -decay candidate nuclei, serves itself as source [1]. This approach provides much higher detection efficiency and density of the candidate nuclei than the «passive source» technique, where a thin source is separated from the detector. For instance, we can compare the 100% efficiency quoted for Ge semiconductor detectors in Ref. 5 with the 4% efficiency of the NEMO2 tracking detector searching for the $2\nu 2\beta$ decay of ^{100}Mo [9] and of ^{116}Cd [10].

As examples of the results achieved with the «active source» technique, we recall the highest half-life limit $T_{1/2} > 1.6 \cdot 10^{25}$ y ($m_\nu < 0.5$ eV) established for ^{76}Ge (enriched HP

Ge detectors) [5] and the $T_{1/2}$ limits in the range $3 \cdot 10^{22} - 4 \cdot 10^{23}$ y ($m_\nu < 2-4$ eV) obtained for ^{136}Xe (high pressure Xe TPC) [6], for ^{130}Te (low-temperature bolometers) [7] and for ^{116}Cd (enriched $^{116}\text{CdWO}_4$ scintillators) [8].

Simple considerations can easily demonstrate that progress in the 2β decay research is directly connected with the development of «active» detectors.

Suitable detectors for 2β decay studies are the well-known calcium fluoride scintillators [11, 12]. In fact, they have a relatively large light output ($\simeq 60\%$ of NaI(Tl)) and offer excellent operational characteristics (nonhygroscopicity, inertness, insolubility, good mechanical properties, etc.) when used as β particles or low-energy radiation detectors [12–14]. In particular, they can be usefully utilized to search for the 2β decays of ^{46}Ca (natural abundance: $\delta = 0.004\%$; $Q_{\beta\beta} = 990.4$ keV) and of ^{48}Ca ($\delta = 0.187\%$; $Q_{\beta\beta} = 4272$ keV) besides the double electron capture of ^{40}Ca ($\delta = 96.941\%$; $Q_{2\text{EC}} = 193.8$ keV). The first experiment of this kind regarded the 2β decay of ^{48}Ca [15]; let us also mention the latest ones on the 2β decay of ^{48}Ca [16], of ^{46}Ca [17] and on the double electron capture of ^{40}Ca [18].

Moreover, the $\text{CaF}_2(\text{Eu})$ detectors have been considered as promising detectors for the direct search of Dark Matter particles with SD coupling [18, 19].

All the considerations mentioned above motivated the R&D program for the development of radiopure calcium fluoride detectors carried out by the DAMA collaboration with the Bicorn company [17, 18]; the aim was to enhance the sensitivity of searches for 2β decay and dark matter particles with SD interaction. We present here the new step of development performed in the framework of this R&D program and devoted mainly to the study of the $\text{CaF}_2(\text{Eu})$ background and to the understanding of its origins. New results on 2β decay processes are presented. More details on the experiment can be found in [20].

1. MEASUREMENTS

Two $\text{CaF}_2(\text{Eu})$ crystals grown by the Bicorn company were used in the present work. Each crystal has 3" diameter by 1" length (370 g mass). These crystals are labelled in the following as Bicorn-1 and Bicorn-2.

The background measurements have been performed in the Gran Sasso underground laboratory inside the R&D DAMA set-up described in Ref. 18 for the Bicorn-1 test. Here we only recall the main features of the apparatus for the measurements performed with Bicorn-2. The crystal was coupled to a low background PMT EMI9265B53/FL through a 10 cm long TETRASIL-B light guide; it was wrapped by a teflon diffuser ($\simeq 100$ μm thick) as also the light guide. The detector and the PMT were inside about 10 cm of low radioactive copper shaped in order to contain them tight. We note that in this measurement no NaI(Tl) anti-Compton detector was used as instead done in the Bicorn-1 data taking [18]. The detector and the $\simeq 10$ cm Cu envelop were closed inside a sealed low-radioactive Cu box, in one turn surrounded by a shield made of 10 cm of low-radioactive copper and 15 cm of low radioactive lead followed by 1.5 mm Cd foil and about 4/10 cm of polyethylene/paraffin. To avoid any contact with environmental air (which can contain Radon in trace), the Cu box was continuously flushed with high purity (HP) nitrogen gas (long stored deep underground) and maintained at about 1 mbar overpressure. Finally, the whole shield was sealed in a plexiglass box also maintained in HP nitrogen atmosphere.

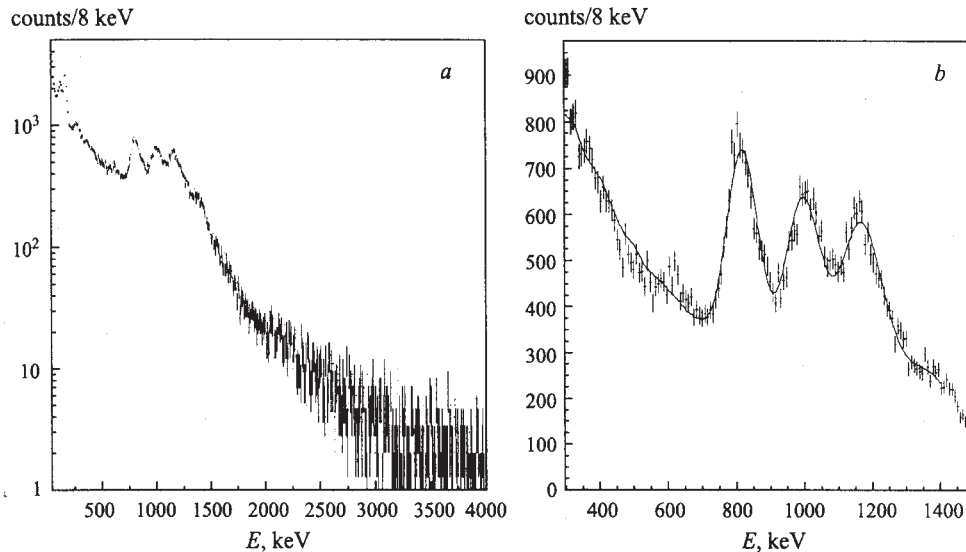


Fig. 1. Measured background spectrum of the Bicron-2 crystal for the energy region 300–4000 keV. The three peaks at about 0.81, 1.01 and 1.15 MeV can be ascribed to α particles from ^{232}Th and ^{238}U chains internal impurities of the crystal, the fitting of the α peaks is shown (b)

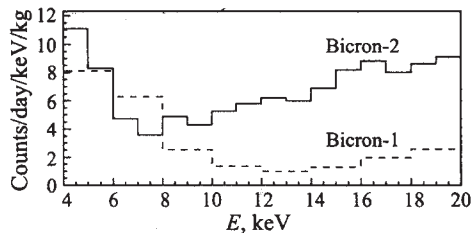


Fig. 2. Measured background spectra of the Bicron-2 and Bicron-1 crystals for the energy region 4–20 keV

applying the same cuts to the source data (collected in the same experimental conditions and energy range).

The background spectrum of the Bicron-2 crystal was measured in this apparatus during 1906.3 h in the three energy regions: 4–20, 40–230 and 200–4000 keV. Data previously collected with Bicron-1 were also available for analysis; the measuring time was 631.4 h for the energy interval 4–24 keV and 260.9 h for the energy region 200–3300 keV [18].

During the measurement, the energy scale and resolution of the detector were calibrated periodically with different sources: ^{109}Cd ($E_\gamma = 22$ keV); ^{137}Cs ($E_\gamma = 32; 662$ keV); ^{22}Na

The rejection of the residual noise just above the software energy threshold (4 keV here)¹ profits by the different timing structure between the noise (PMT fast signals with decay times of the order of tens ns) and the $\text{CaF}_2(\text{Eu})$ «scintillation» (signals with decay times of the order of 940 ns) pulses, whose shapes were recorded over 3125 ns by a Lecroy Transient digitizer. The residual noise can be rejected from the production data by applying software cuts; the corresponding software cut efficiency for each energy bin can be evaluated by apply-

¹We recall that in any kind of experiment requiring an energy threshold at keV level, software cuts or — sometimes — hardware procedures near threshold have to be applied, although generally the corresponding procedures are not explicitly quantified by the authors.

($E_\gamma = 511; 1275$ keV) and ^{60}Co ($E_\gamma = 1173; 1332$ keV). In addition, background peaks ($E_\gamma = 128$ and 169 keV) from ^{152}Eu contamination in the Bicorn-2 crystal were used for calibration in the energy region 40–230 keV.

The measured background spectrum of Bicorn-2 crystal in the energy region 300–4000 keV is shown in Fig. 1, while the low energy part (4–20 keV) and the middle energy (40–400 keV) are given in Figs. 2 and 3, respectively. Three peaks at the energies of about 0.81, 1.01 and 1.15 MeV are present; considering the typical α/β ratio for the light output of the $\text{CaF}_2(\text{Eu})$ crystals, these peaks can be attributed to α particles due to ^{232}Th and ^{238}U internal impurities. Moreover, in the lower energy region, three clear peaks at energies $\simeq 47$, $\simeq 129$ and $\simeq 169$ keV are also present; as was shown by simulation (see below), such peaks could be explained by ^{152}Eu ($T_{1/2} = 13$ y) activity created in the $\text{CaF}_2(\text{Eu})$ crystal by the neutron activation at Earth surface.

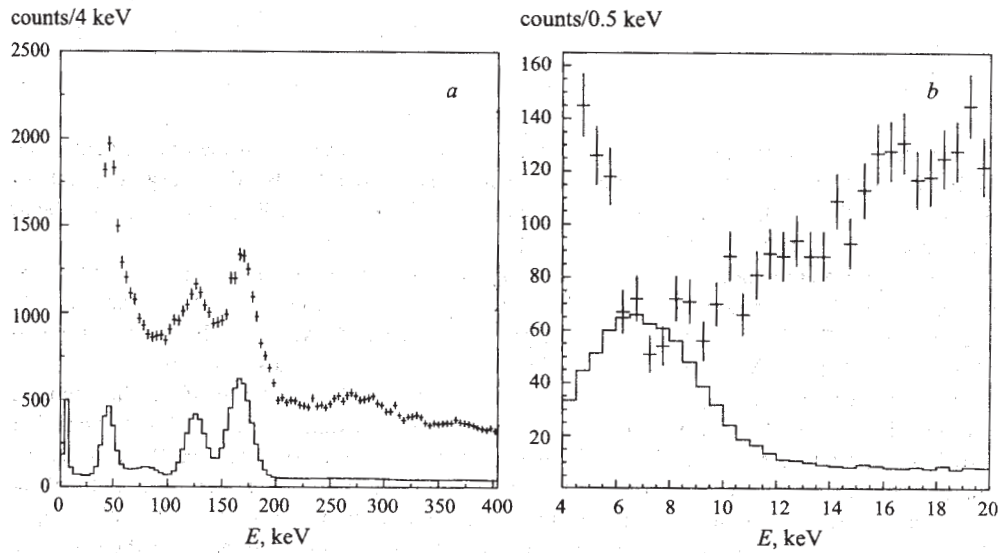


Fig. 3. Experimental spectrum of the Bicorn-2 crystal up to the energy of 400 keV. The three peaks at $\simeq 47$, $\simeq 129$ and $\simeq 169$ keV can be ascribed to ^{152}Eu ($T_{1/2} = 13$ y) activity created in the $\text{CaF}_2(\text{Eu})$ crystal by the neutron activation at Earth surface. The model distribution from ^{152}Eu contamination is also shown (see text for details)

2. BACKGROUND SIMULATION AND ESTIMATE OF THE 2β DECAY HALF-LIFE LIMITS

The locations and amounts of the radioactive contaminations have been estimated by simulating the background spectra of the $\text{CaF}_2(\text{Eu})$ crystal by the GEANT3.21 package [22] and the event generator DECAY4 [23], which describes the initial kinematics of the events. The considered background model includes external (from the ^{40}K , ^{232}Th and ^{238}U contaminations in the PMT) and internal (from ^{40}K , ^{152}Eu , ^{232}Th and ^{238}U intrinsic impurities

in the $\text{CaF}_2(\text{Eu})$ crystal) contributions; it was supposed that equilibrium in U and Th chains may be broken. The fit of the experimental spectrum in the energy interval 40–2700 keV following this model gives the activities of the possible impurities in PMT and in crystal; these are shown in Table 1. As evident, the equilibrium in U and Th chains for the $\text{CaF}_2(\text{Eu})$ contaminations is broken, as often in detectors.

Table 1. **Radioactive impurities of the PMT and $\text{CaF}_2(\text{Eu})$ crystal (mBq/kg)**

Source of background	^{232}Th -chain		^{238}U -chain				^{152}Eu	^{40}K
	^{232}Th	^{228}Ra	^{238}U	^{230}Th	^{226}Ra	^{210}Pb		
PMT	< 100	—	—	—	400(200)	—	—	$4(2) \cdot 10^4$
$\text{CaF}_2(\text{Eu})$ Bicron-2	1.2(5)	11(5)	0.5(2)	4.5(8)	2.4(10)	0.5(2)	7.5(20)	5(2)

The fit in the energy interval 300–1500 keV is shown together with the experimental data in the insert of Fig. 1, where a reasonable agreement is achieved. Also the energy dependence of the α/β ratio for the $\text{CaF}_2(\text{Eu})$ crystal was obtained: $\alpha/\beta = 0.130 + 0.017 E_\alpha$, E_α being the energy of α particle in MeV.

The experimental spectrum up to the energy of 400 keV is given in Fig. 3. There is also shown the model distribution from ^{152}Eu contamination, which consists of four peaks at the energies: a) $\simeq 7$ keV (L -shell binding energy of Sm); b) $\simeq 47$ keV (K -shell binding energy of Sm); c) $\simeq 128.8$ keV (sum of L -shell binding energy with the most intensive $\simeq 121.8$ keV γ ray or conversion electron from ^{152}Eu decay); d) $\simeq 168.8$ keV (sum of K -shell binding energy with the $\simeq 121.8$ keV γ ray). The absence in the data of the $\simeq 7$ keV peak (see also Fig. 2) could be explained by the known cross sections uncertainties ($\pm 10\%$) in the low energy region 40–120 keV, which were used for simulation. The estimated activity of the ^{152}Eu in the Bicron-2 crystal — considering the higher energy peaks — is $7.5(20)$ mBq \cdot kg $^{-1}$. Such activity could be reached after approximately five months of activation of the $\text{CaF}_2(\text{Eu})$ crystal (in our case the content of Eu is $\simeq 0.5\%$ [24]) at the Earth surface, where the thermal neutron flux is about $5 \cdot 10^{-3}$ neutrons/cm 2 /s.

The background model described above was used to estimate half-life limits for the double EC capture of ^{40}Ca ($Q_{2\text{EC}} = 193.8$ keV) and the neutrinoless double β decay of ^{46}Ca ($Q_{\beta\beta} = 990.4$ keV). The number of candidate nuclei in each $\text{CaF}_2(\text{Eu})$ crystal is equal to $2.766 \cdot 10^{24}$ and $1.141 \cdot 10^{20}$ for ^{40}Ca and ^{46}Ca , respectively. The double EC capture of ^{40}Ca has to be followed by the emission of two X rays with 3.2 keV energy each and it is expected that an inner bremsstrahlung photon will carry off the energy released in the neutrinoless process. Efficiencies for the different double β decay processes were calculated by using the GEANT3.21 package [22] and the event generator DECAY4 [23].

The simulated response functions of $\text{CaF}_2(\text{Eu})$ crystals and the calculated values of efficiencies for the different 2β decay processes were used together with the corresponding background rates, the measuring times and the numbers of ^{40}Ca and ^{46}Ca nuclei in order to estimate half-life limits for the effects under investigation. These limits are summarized in Table 2 and were obtained in two different ways. Firstly, they have been obtained by using

Table 2. Half-life limits for different 2β processes of ^{40}Ca and ^{46}Ca

2 β process (Peak energy)	$T_{1/2}$ limit, y (one σ approach) 68 % C.L.		$T_{1/2}$ limit, y (χ^2 fit procedure) 68 % (90 %) C.L.		$T_{1/2}$ limit, y (χ^2 fit procedure) 68 % (90 %) C.L. (Combined from Bicron-1+Bicron-2)
	Bicron-2	Bicron-1	Bicron-2	Bicron-1	
$0\nu 2\text{EC}$ of ^{40}Ca (6.4 keV)	$4.4 \cdot 10^{21}$	$2.7 \cdot 10^{21}$	$4.1(2.5) \cdot 10^{21}$	$2.4(1.4) \cdot 10^{21}$	$4.9(3.0) \cdot 10^{21}$
$0\nu 2\text{EC}$ of ^{40}Ca (193.8 keV)	$5.5 \cdot 10^{20}$	—	$3.4(1.8) \cdot 10^{20}$	—	$3.4(1.8) \cdot 10^{20}$
$2\nu 2\text{EC}$ of ^{40}Ca (6.4 keV)	$8.8 \cdot 10^{21}$	$5.3 \cdot 10^{21}$	$8.2(5.0) \cdot 10^{21}$	$4.8(2.8) \cdot 10^{21}$	$9.8(5.9) \cdot 10^{21}$
$0\nu 2\beta$ of ^{46}Ca (990 keV)	$1.8 \cdot 10^{17}$	$9.1 \cdot 10^{16}$	$1.5(0.9) \cdot 10^{17}$	$8.3(4.9) \cdot 10^{16}$	$1.7(1.0) \cdot 10^{17}$

the so-called «one σ approach», in which the excluded number of signal events is determined simply as the square root of the number of background counts in a given energy window ΔE . Notwithstanding its simplicity, this method gives the right scale of the experiment's sensitivity. Moreover, the $T_{1/2}$ limits were improved by using the standard procedure of the least square fit of the experimental data by the sum of the background model and of the simulated response function of the detector for the 2β decay process under investigation. From such a fit, the number of events in the expected signal peak (and the uncertainty) were determined, usually giving no statistical evidence for the effect. Then using these values, the excluded — at 68 % (90 %) C.L. — numbers of events for the different 2β decay processes were calculated in accordance with the Particle Data Group procedure [25].

As an example, part of the experimental spectrum of the crystal Bicron-2 in the energy interval 4–20 keV and the fitting curve are shown in Fig. 4, where the excluded — at 90 % C.L. — peak from the $0\nu 2\text{EC}$ decay of ^{40}Ca with half-life $2.5 \cdot 10^{21}$ y is also shown.

In the last column of Table 2 the half-life limits obtained by combining the results for both crystals: Bicron-2 and Bicron-1 [18] are shown. In this case the procedure was as following. The numbers of events in the expected peak — determined for each crystal by χ^2 fit as described above — were simply added: $S = S_1 + S_2$, while their error bars were

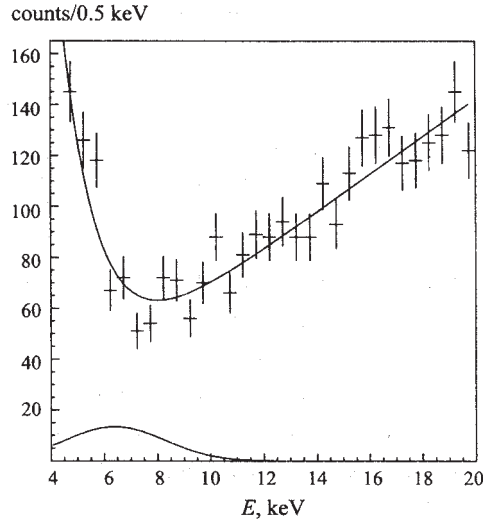


Fig. 4. The 4–20 keV experimental energy spectrum of the Bicron-2 crystal and the fitting curve, where the excluded — at 90 % C.L. — peak from $0\nu 2\text{EC}$ of ^{40}Ca with half-life $2.5 \cdot 10^{21}$ y is also shown

added quadratically: $\sigma(S) = [\sigma^2(S_1) + \sigma^2(S_2)]^{1/2}$. The values of S and $\sigma(S)$ were used to calculate the combined excluded numbers of events for different 2β decay processes.

The $T_{1/2}$ limit obtained for $0\nu 2\beta$ decay of ^{46}Ca is higher than the best result previously available [17]. Restriction for the two neutrino 2EC of ^{40}Ca is also improved from value $4.6 \cdot 10^{21}$ y [18] to $\simeq 10^{22}$ y. It should be stressed that the last one is the highest half-life limit obtained up-to-date for the $2\beta^+$ decay processes and, in particular, for the double electron capture (see reviews [1] and the last work [26] for comparison). The same is true for the neutrinoless 2EC of ^{40}Ca , which $T_{1/2}$ limit is established for the first time.

CONCLUSIONS

The new and highly improved $T_{1/2}$ limits on the 2β decay processes of ^{46}Ca and ^{40}Ca achieved here have surely demonstrated the feasibility and perspectives of developed $\text{CaF}_2(\text{Eu})$ scintillators for high sensitive investigations of the 2β decay. The use of these scintillators in the search for dark matter particles with SD coupling is also another interesting possibility. Further efforts are in progress.

REFERENCES

1. *Moe M., Vogel P.* // Ann. Rev. Nucl. Part. Sci. 1994. V. 44. P. 247;
Tretyak V. I., Zdesenko Yu. G. // At. Data Nucl. Data Tables. 1995. V. 61. P. 43;
Suhonen J., Civitarese O. // Phys. Rep. 1998. V. 300. P. 123.
2. *Klapdor-Kleingrothaus H. V. et al.* // J. Phys. G: Nucl. Part. Phys. 1998. V. 24. P. 483.
3. *Bednyakov V. A. et al.* // Mod. Phys. Lett. A. 1997. V. 12. P. 233.
4. *Hirsch M., Klapdor-Kleingrothaus H. V.* // Prog. Part. Nucl. Phys. 1998. V. 40. P. 323.
5. *Baudis L. et al.* // Phys. Rev. Lett. 1999. V. 83. P. 41.
6. *Luescher R. et al.* // Phys. Lett. B. 1998. V. 434. P. 407.
7. *Alessandrello A. et al.* // Phys. Lett. B. 1998. V. 433. P. 156.
8. *Danevich et al. F. A.* // Phys. Rev. C. 2000. V. 63. P. 045501.
9. *Dassie D. et al.* // Phys. Rev. D. 1995. V. 51. P. 2090.
10. *Arnold R. et al.* // Z. Phys. C. 1996. V. 72. P. 239.
11. *Knoll G. F.* Radiation Detection and Measurement. 2nd ed. N.Y., 1989.
12. *Colmenares C. et al.* // Nucl. Instr. Meth. 1974. V. 114. P. 277.
13. *Campbell M. et al.* // Nucl. Instr. Meth. 1976. V. 137. P. 235.
14. *Pouliot J. et al.* // Nucl. Instr. Meth. 1988. V. 270. P. 69.
15. *Der Mateosian E., Holdhaber M.* // Phys. Rev. 1966. V. 146. P. 810.
16. *You K. et al.* // Phys. Lett. B. 1991. V. 265. P. 53.
17. *Bernabei R. et al.* // Nuovo. Cim. A. 1998. V. 111. P. 347.
18. *Bernabei R. et al.* // Astropart. Phys. 1997. V. 7. P. 73.
19. *Bacci C. et al.* // Astropart. Phys. 1994. V. 2. P. 117.

20. *Belli P. et al. // Nucl. Phys. B. 1999. V. 563. P. 97.*
21. *Ellis J., Flores R. A. // Phys. Lett. B. 1991. V. 263. P. 259.*
22. *Brun R. et al. CERN Program Library Long Write-Up W5013. CERN, 1994.*
23. *Ponkratenko O. A. et al. // Phys. Atom. Nucl. 2000. V. 63. P. 1282.*
24. *Krus D. Bicron Co. Private communication.*
25. *Particle Data Group: Review of Particle Physics // Phys. Rev. D. 1996. V. 54. P. 1.*
26. *Belli P. et al. // Astropart. Phys. 1999. V. 10. P. 115.*