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GAMIDEN: A Program to Aid in the Identification of Unknown Materials by Gamma-Ray Spectroscopy

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Foreword

The UCRL-50400 series, *An Integrated System for Production of Neutronics and Photonics Calculational Constants*, describes an integrated, computer-oriented system for the production and application of neutronics and photonics calculational constants.

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- Vol. 1, Part A, Rev. 3, *ECSIL: A System for Storage, Retrieval, and Display of Experimental Neutron Data*, September 1976.
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- Vol. 2, Rev. 2, *A Bibliography of the Experimental Data of Neutron-Induced Interactions*, July 1976.
- Vol. 3, Rev. 2, *An Index of the Experimentel Data of Neutron-Induced Interactions*, July 1976.
- Vol. 4, Rev. 1, *Evaluated Nuclear Data Library*, September 1981.
- Vol. 5, Part A, Rev. 1, *CLYDE: A Code for the Production of Calculational Constants from Nuclear Data*, September 1975.
- Vol. 5, Part B, Rev. 1, *Relativistic Transformations between Center-of-Mass and Laboratory Systems for Two-Body Nuclear Reactions*, April 1978.
- Vol. 6, Rev. 2, *Tables and Graphs of Photon-Interaction Cross Sections from 1 keV to 100 MeV*, December 1978.
- Vol. 7, Part A, Rev. 1, *Major Neutron-Induced Interactions (Z ≤ 55): Graphical, Experimental Data*, July 1976.
- Vol. 7, Part B, Rev. 1, *Major Neutron-Induced Interactions (Z > 55): Graphical, Experimental Data*, July 1976.
- Vol. 8, Part A, Rev. 1, *Supplemental Neutron-Induced Interactions (Z ≤ 35): Graphical, Experimental Data*, July 1976.
- Vol. 8, Part B, Rev. 1, *Supplemental Neutron-Induced Interactions (Z > 35): Graphical, Experimental Data*, July 1976.
- Vol. 9, *Thresholds of Nuclear Reactions Induced by Neutrons, Photons, Deuterons, Tritons, and Alpha Particles*, September 1970.
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- Vol. 15, Part F, *Experimental and Evaluated Elastic Nuclear Plus Interference Cross Sections for Light Charged Particles*, July 1980.
- Vol. 16, Rev. 2, *Tabular and Graphical Presentation of 175 Neutron-Group Constants Derived from the LLL Evaluated-Nuclear-Data Library (ENDL)*, October 1978.
- Vol. 17, Part A, Rev. 2, *Program LINEAR (Version 79-1): Linearize Data in the Evaluated-Nuclear-Data File/Version B (ENDF/B) Format*, October 1979.
- Vol. 17, Part B, Rev. 2, *Program SIGMA1 (Version 79-1): Doppler Broaden Evaluated Cross Sections in the Evaluated-Nuclear-Data File/Version B (ENDF/B) Format*, October 1979.

- Vol. 17, Part C, *Program RECENT: Reconstruction of Energy-Dependent Cross Sections from Resonance Parameters in the ENDF/B Format*, October 1979.
- Vol. 17, Part D, *Program GROUPIE: Calculation of Self-Shielded Cross Sections and Multiband Parameters from Evaluated Data in the ENDF/B Format*, 1980.
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- Vol. 17, Part F, *DOWNER (Version 79-1): Group Collapse Cross Section and Transfer Matrices*, January 1979.
- Vol. 18, *ACTL: Evaluated Neutron Activation Cross-Section Library*, October 1978.
- Vol. 19, *Neutron-Induced Angular and Energy Distributions: Graphical Experimental Data*, April 1977.
- Vol. 20, *Bonderenko Self-Shielded Cross Sections and Multiband Parameters Derived from the LLL Evaluated-Nuclear-Data Library (ENDL)*, July 1978.
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- Vol. 21, Part C, *Program SIGMAL (Version 79-1): Doppler Broaden Evaluated Cross Sections in the Livermore-Evaluated Nuclear Data Library (ENDL) Format*, March 1979.
- Vol. 22, Rev. 1, *GAMIDEN: A Program to Aid in the Identification of Unknown Materials by Gamma-Ray Spectroscopy*, June 1982.
- Vol. 22, Rev. 2, *GAMIDEN: A Program to Aid in the Identification of Unknown Materials by Gamma-Ray Spectroscopy*, May 1983.
- Vol. 23, *ENSL and CDRL: Evaluated Nuclear Structure Libraries*, February 1981.
- Vol. 24, *Thresholds and Q Values of Nuclear Reactions Induced by Neutrons, Protons, Deuterons, Tritons, ^{3}He Ions, Alpha Particles, and Photons*, March 1981.

Contents

GAMIDEN: A Program to Aid in the Identification of Unknown Materials by Gamma-Ray Spectroscopy

Abstract

The intent of the computer code GAMIDEN is to help identify isotopes by their gamma-ray emissions and thus to assist in the nondestructive assay of unknown materials. From both radioactive decays and neutron captures, GAMIDEN searches GAMTOT83, a file of gamma-ray spectra, for matches with observed photon energies. This report describes the search procedure, outlines the use of the code, and gives an example.

The code is designed to operate on the CRAY 1 computer at Lawrence Livermore National Laboratory (LLNL). It is written in standard Fortran (ANSI) for the most part but contains some LRLTRAN instructions to make use of the Livermore time-sharing system (LTSS). The code uses about 545,000 words of memory. Typical problems run in about 45 s. The source program and the data file are available on request.

Introduction

The gamma rays emitted by an isotope undergoing radioactive decay or neutron capture have energies and intensities that are characteristic of that isotope. Such a set of gamma rays is often referred to as the signature of the isotope. A useful method for the nondestructive assay of unknown materials is based on identifying the signature of the isotope.

Since there are thousands of known gamma rays, we have developed an automated search procedure. GAMIDEN, a computer code, searches GAMTOT83, a disk file of gamma-ray spectra, for isotopes that match observed gamma energies. GAMIDEN lists the matching isotopes, eliminates isotopes that are not likely to be the source of the observed photons, and then lists the isotopes that are left—the likely candidates. The user makes any further analysis required to identify the actual source isotopes.

Search Procedure Used in GAMIDEN

The search procedure used in GAMIDEN is outlined as follows:

1. Given a set of observed photon energies, search the tabulated data for photons with the same energies, within experimental uncertainty. Isotopes that emit such photons form the initial list of possible sources of the observed photons.
2. Examine the tabulated spectrum of each isotope for gammas that have a higher probability of emission than the observed gamma has; presumably, such gammas should have been observed as well.
3. Examine energies of the other observed photons to see if these higher-probability gammas, if any, were, in fact, observed.
4. If the matching isotope's spectrum contains a higher-probability gamma and it was not observed, remove this isotope from the list of possible sources. Isotopes that survive this test constitute the final list of possible sources.

Limiting the Search

If in the search procedure just described we include other information available to the user, the search will be much faster and other improbable isotopes can be eliminated from the final list. These additional constraints on the search are described below.

1. **Ignore tabulated photons below a chosen minimum energy.** There are several reasons why this may be desirable: (a) The commonly used lithium-drifted germanium (Ge-Li) detectors are usually biased in such a way that photons below some minimum energy are not recorded. (b) There may be other unknown materials between the source and the detector—containers or shielding, for example. Such materials will alter the photon spectrum of the source, especially at lower energies (since photon-interaction cross sections rise rapidly with decreasing energy). (c) There may be a very large number of observed photons, and the user may want to limit the volume of output from the program. He can do this by choosing an energy cutoff that leaves a reasonable number of higher-energy photons as input values. If there are interposed materials, higher-energy photons are most likely to represent the spectrum of the unknown source.

2. **Ignore isotopes outside some chosen range.** The user may know that the source does not contain isotopes outside some range of atomic number Z and mass number A, or he may simply be uninterested in isotopes outside of some such range. The user combines the Z and A of these limiting isotopes into numbers equal to $1000Z + A$, and enters these to indicate the limits chosen.

3. **Search only prompt gammas, or only those from radioactive decays, or both.** If the source is not emitting prompt gammas or gammas from decays with very short half-lives, the user can specify a minimum half-life as a criterion for searching. He can set this value to zero if he wants to search both prompt and decay gammas. (In GAMTOT83 prompt gammas are assigned a half-life of 10^{-14} s.)

(Of course, the capture process may result in the formation of a radioactive product, which will decay with its own characteristic spectrum and half-life. The user may need to consider this in deciding whether to search prompt gammas only.)

4. **Do not consider gammas with a low probability of emission.** Clearly low-probability photons, even if present in the unknown spectrum, may not be detected. (The tabulated probabilities, also called multiplicities, are actually probabilities of emission per decay or per neutron capture.)

The user will also have to estimate the uncertainty to be associated with each observed photon energy. The Ge-Li detector has an accuracy of plus or minus a few keV for photon energies in the range from 100 keV to 10 MeV. This uncertainty arises mainly from the nonlinearity of the relationship between photon energy and detector output. (A Ge-Li detector operated at liquid-nitrogen temperature has a precision of about 0.1 keV for photons in this energy range.) Unknown energies are determined by interpolating or extrapolating detector output values from known photons; typically, only a few such calibration photons are used. To take account of asymmetry in the calibration, the program permits the user to specify different values for the uncertainties above and below the observed energies.

Program Output

GAMIDEN provides four kinds of output information:

1. The input data (search constraints and photon energies).
2. A list of each match of an observed photon giving the identity of the matching isotope and a list of all the photons in the spectrum of the matching isotope that satisfy the search constraints.
3. A list of the isotopes that have no unobserved higher-probability photons. These isotopes remain active candidates.
4. A list of the eliminated isotopes, with the multiplicity and energy of both the observed and the unobserved higher-multiplicity photon.

Gamma Spectra in the Data File (GAMTOT83)

There are 44,810 entries in GAMTOT83, taken from two compilations[1,2] of gamma-ray spectra. The compilation of radioactive-decay gammas[1] includes the half-life for each isotope and the energy and probability of emission per decay for each gamma emitted by that isotope. We have included in GAMTOT83 all photons from Ref. 1 for which multiplicities are tabulated. The compilation of gammas induced by neutron capture[2] gives the energy and probability per capture for each photon accompanying the formation of the product nucleus. We have included all photons from Ref. 2 that have probabilities greater than 0.01. The structure of GAMTOT83 is described in Appendix A.

Using GAMIDEN

This section deals with the use of GAMIDEN per se; Appendix B details the use of GAMIDEN at LLNL.

Most likely the user will want to enter the search constraints from his console so that he can alter them after seeing the program output. However, if there are many observed photon energies, he may want to enter photon energies from a user-created disk file. GAMIDEN allows the user to enter his data from a console or from a disk file. The disk file contains the observed photon energies in format 10 F7.3. The last record of the data file should be blank. The program will request the name of the disk file, which must be less than or equal to eight characters.

The program prompts the user as follows:

ENTER NAME OF SOURCE FILE IN A8, E.G., GAMTOT83

This gives the user the option of using a data file other than GAMTOT83 if he wishes to do so. Of course, any other data file must be in the same format as GAMTOT83. (See Appendix A for a description of the required format.)

ENTER DETECTOR SHIFTS LOWER, THEN UPPER IN 2F7.3

Note that these uncertainties, although of the order of a few keV, are entered in MeV. The user may use either a 2F7.3 format or a field-free, floating-point format and delimit the two variables with a comma.

ENTER LOWER ENERGY LIMIT FOR GAMMAS IN F7.3

This suppresses the search for energies less than the entered value. The unit of energy is MeV.

ENTER LOWER AND UPPER LIMITS OF ISOTOPES IN 2I6

These two numbers, entered as 1000Z + A, limit the range of isotopes searched.

ENTER LOWER BOUND of HALF-LIFE IN SEC. (F10.3)

This suppresses the search for unreasonably short half-lives. Enter zero if all isotopes are to be searched.

IF CAPTURE GAMMAS ONLY ENTER YES; OTHERWISE LF

A "YES" causes the program to ignore the previous entry and search only for gammas from Ref. 2 associated with neutron capture.

ENTER INPUT DEVICE FOR OBSERVED GAMMAS: 06 IF FILE; 59 IF TTY

If TTY is chosen, the following prompt will appear:

ENTER UP TO 10 GAMMA ENERGIES IN 10F7.3

You may use format 10F7.3 or you may enter the energies (in MeV) in field-free format and delimit the variables by commas. If there are 10 or fewer energies to enter, you should put them all on this line. If there are more than 10 energies, you should put exactly 10 on this line. The program will prompt for additional energies (ENTER NEXT 10 GAMMA ENERGIES IN 10F7.3) until it encounters a blank record or a line with fewer than 10 entries.

ENTER LOWER BOUND OF MULTIPLICITIES IN F7.3

This suppresses the search for low-probability photons.

After output is completed the user is prompted for further problems:

IF ANOTHER PROBLEM ENTER 1; IF NOT LF

The program will request another set of observed photon energies.

Example Input

- Assume that observed photon energies are 0.058, 0.826, 1.173, 1.332, and 2.158 MeV.
- Consider all tabulated isotopes between ${}_{20}Ca^{40}$ and ${}_{82}Pb^{207}$ with half-lives greater than 660 s (11 min).
- Consider all tabulated photons with energies greater than 0.80 MeV and probabilities greater than 10%.
- Accept as a match any photon that satisfies the criteria above and is within plus or minus 0.005 MeV (5 keV) of an observed photon.

The user would enter these data from his console, as shown in the dialogue below.

ENTER NAME OF SOURCE FILE IN A8, E.G., GAMTOT83
GAMTOT83

ENTER DETECTOR SHIFTS LOWER THEN UPPER IN 2F7.3
.005,.005

ENTER LOWER ENERGY LIMIT FOR GAMMAS IN F7.3
.80

ENTER LOWER AND UPPER LIMITS OF ISOTOPES IN 2I6
20040 82207

ENTER LOWER BOUND OF HALF-LIFE IN SEC., (F10.3)
660.

IF CAPTURE GAMMAS ONLY ENTER YES; OTHERWISE LF

ENTER LOWER BOUND OF MULTIPLICITIES (F7.3)
.1

ENTER INPUT DEVICE FOR OBSERVED GAMMAS: 06 IF FILE, 59 IF TTY
59

ENTER UP TO 10 GAMMA ENERGIES IN 10F7.3
.058, .826, 1.173, 1.332, 2.158

IF ANOTHER PROBLEM ENTER 1; IF NOT LF

Sample Output

HOUT, a disk file of results, is generated when GAMIDEN is run. The results from the sample problem are listed and explained below.

1. The input variables are printed out (Fig. 1).

```
detector shifts are: lower =  0.005 upper =  0.005
lower limit for photons is:  0.800
lower and upper limit for isotopes are: 20040  82207
lower limit for half-lives is: 6.600e+02 seconds

photon energies observed are:
       0.058  0.826  1.173  1.332  2.158
lower bound of multiplicities is:  0.100
```

Figure 1.

2. The GAMTOT83 table is searched for isotopes that satisfy the input criteria. Each such isotope is printed out, accompanied by a list of other gamma emissions from the isotope that one might expect to see. The list below (Fig. 2), for the sample problem, gives the possible isotopes found in this way. The sym..ols used in the column labeled SOURCE are described in Appendix A.

```
1   nucleus   photon   multiplicity   half-life          source

  27-co- 60   1.1732   0.9986000   5.272e+00 y  nth co 59,nfa cu 63,nfa ni 60   a
  27-co- 60   1.3325   0.9998000   5.272e+00 y  nth co 59,nfa cu 63,nfa ni 60   a
               1.1732   0.9986000
               1.3325   0.9998000
  27-co- 62   1.1720   0.9790000   1.391e+01 m  nfa ni 62,cha ni 64,cha co 59   a
               1.1635   0.6810000
               1.1720   0.9790000
               2.0037   0.1860000
  29-cu- 60   0.8264   0.2174000   2.320e+01 m  cha ni 60,cha ni 58             a
  29-cu- 60   1.3325   0.8800000   2.320e+01 m  cha ni 60,cha ni 58             a
               0.8264   0.2174000
               1.3325   0.8800000
               1.7916   0.4541000
  35-br- 82   0.8278   0.2420000   1.470e+00 d  nth br 81,nfa kr 82,nfa rb 85   a
               0.8278   0.2420000
               1.0440   0.2800000
               1.3174   0.2700000
               1.4749   0.1660000
  37-rb- 82   0.8278   0.2100000   6.200e+00 h  cha kr 82,cha br 79             a
               0.8278   0.2100000
               1.0440   0.3300000
               1.3175   0.2600000
               1.4748   0.1700000
```

Figure 2.

1 nucleus	photon	multiplicity	half-life	source	
41-nb- 98	1.1[illegible]	0.17[illegible]	5.110e+01 m	nfi 0.17,nfa mo 98	e
	0.[illegible]	0.1[illegible]			
	1.1[illegible]	0.17[illegible]			
45-rh-100	0.[illegible]	0.2[illegible]	2.060e+01 h	cha ru100,cha ru 98,cha rh103	e
	0.[illegible]	0.[illegible]			
	1.1071	0.1[illegible]			
	1.[illegible]21	0.1[illegible]			
	1.[illegible]4	0.2100000			
	1.[illegible]7	0.1240000			
	2.3781	0.[illegible]70000			
45-rh-106	0.[illegible]	0.11[illegible]2000	2.200e+00 h	nfi 2e-5,nfa pd106,nfa ag109	e
	0.[illegible]40	0.11[illegible]2000			
	0.[illegible]	0.11[illegible]2000			
	1.0[illegible]7	0.2[illegible]76000			
	1.1[illegible]1	0.106[illegible]000			
	1.5274	0.1344000			
46-pd- 98	1.3330	0.1600000	1.800e+01 m	cha ru 96,cha cd	e
	0.8379	0.1400000			
	1.3330	0.1600000			
47-ag-106	0.8247	0.1543000	8.500e+00 d	pho ag107,nfa cd106,cha rh103	e
	0.8043	0.1244000			
	0.8247	0.1543000			
	1.0458	0.2972000			
	1.1280	0.1182000			
	1.1994	0.1129000			
	1.5277	0.1641000			
51-sb-120	1.1713	1.0000000	5.800e+00 d	cha sn120,nfa te120,nfa sb121	e
	1.0231	0.9900000			
	1.1713	1.0000000			
57-la-143	1.1700	0.5700000	1.430e+01 m	nfi 5.900	r
	1.0700	0.2600000			
	1.1700	0.5700000			
	1.5800	0.2800000			
	1.7000	0.1900000			
	1.9800	0.3500000			
	2.4600	0.1300000			
	2.5600	0.2700000			
	2.8500	0.1500000			
60-nd-139	0.8278	0.1025000	5.500e+00 h	cha pr141	e
	0.8278	0.1025000			
	0.9822	0.2618000			
65-tb-160	1.1780	0.1550000	7.210e+01 d	nth tb159,nfa dy160	e
	0.8764	0.3000000			
	0.9624	0.1000000			
	0.9662	0.2550000			
	1.1780	0.1550000			
67-ho-166	0.8306	0.1000000	1.200e+03 y	nth ho165,nfa tm169,nfa er166	e
	0.8103	0.5970000			
	0.8306	0.1000000			
68-eu-161	0.8265	0.6150000	3.240e+00 h	nfa er162,pho er162,cha dy161	e
	0.8265	0.6150000			
69-tm-168	0.8211	0.1114000	8.690e+01 d	nfa tm169,nfa yb168,cha er168	e
	0.8160	0.4627000			
	0.8211	0.1114000			
79-au-189	0.8277	0.1000000	2.830e+01 m	cha au197	r
79-au-189	1.1778	0.1700000	2.830e+01 m	cha au197	r
	0.8126	0.5900000			
	0.8277	0.1000000			
	0.9020	0.1000000			
	1.0715	0.2600000			
	1.1606	0.3300000			
	1.1778	0.1700000			
81-tl-200	0.8283	0.1100000	1.088e+00 d	cha hg,cha au197,cha tl203	e
	0.8283	0.1100000			
	1.2057	0.3040000			

Figure 2. (Continued)

3. The elimination criterion (no unobserved higher-multiplicity photon) is applied, and those isotopes that survive this test are listed (Fig. 3).

Of these candidates $_{27}Co^{60}$ is most probable since it has two gammas that meet the search criteria and both were observed. The other isotopes, for which only one of one or more possible gammas were observed, are less-probable candidates.

s u m m a r y

isotopes that cannot be eliminated because of photons that should be present and are not

isotope	no. of matches
27-co- 60	2 of 2
27-co- 62	1 of 3
41-nb- 98	1 of 2
46-pd- 98	1 of 2
51-sb-120	1 of 2
57-la-143	1 of 6
66-eu-161	1 of 1

Figure 3.

4. The isotopes eliminated in the preceding step are listed (Fig. 4).

photons that should be observed and are not

isotope	least mult. observed	observed photon	unobserved photon	multiplicity
29-cu- 60	0.2174	0.8264	1.7916	0.4541
35-br- 82	0.2420	0.8278	1.0440	0.2800
			1.3174	0.2700
37-rb- 82	0.2100	0.8278	1.0440	0.3300
			1.3175	0.2600
45-rh-100	0.2060	0.8225	1.5534	0.2100
			2.3761	0.3570
45-rh-106	0.1152	0.8245	0.8040	0.1152
			1.0457	0.2976
			1.5274	0.1344
47-ag-106	0.1543	0.8247	1.0458	0.2972
			1.5277	0.1641
60-nd-139	0.1025	0.8278	0.9822	0.2618
65-tb-160	0.1550	1.1780	0.8764	0.3000
			0.9662	0.2550
67-ho-166	0.1000	0.8306	0.8103	0.5970
69-tm-168	0.1114	0.8211	0.8160	0.4627
79-au-189	0.1000	0.8277	0.8126	0.5900
			0.9020	0.1000
			1.0715	0.2600
			1.1606	0.3300
81-tl-200	0.1100	0.8283	1.2057	0.3040

Figure 4.

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2. M. A Lone, R. A. Leavitt, and D. A. Harrison, *Atomic Data and Nuclear Data Tables,* 26, 511 (1981).

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Appendix A. File Structure of GAMTOT88

GAMTOT88 or a similar file must be present to run with GAMDEN. Each photon tabulated in GAMTOT88 takes one record in the file. Table A1 describes the format of a single record.

Table A1. Format of a single record in GAMTOT88

Bytes	Variable name	Format	Description
1	IFL	I1	EOF indicator: blank on data card; 1 (or other digit) on last card in file, which is otherwise blank.
2-7	IZA	I6	Combined atomic number (Z) and mass number (A), in the form 1000Z + A.
8-12	Blank	5X	
13-19	GAM	F7.4	Observed photon energy (in MeV).
20-24	Blank	5X	
25-33	AI	F9.7	Probability (same as multiplicity) of this photon per decay or per capture.
34-38	Blank	5X	
39-48	TIME	E10.3	Half-life of isotope, numerical value only (unit is next variable), is equal to 1.000E-14 if this is a prompt (n-capture) photon.
49	Blank	1X	
50	IUNIT	A1	Unit is used for half-life: second (S), minute (M), day (D), or year (Y).
51-55	Blank	5X	
56-95	SOURCE	5A8	Source[a]: N,GAM, prompt n-capture; CHA, charged particle-induced reaction; NTH, thermal or epithermal neutron-induced reaction; NFA, fast neutron-induced reaction; NFI,[b] thermal neutron-induced fission of ^{235}U; NAT,[c] naturally occurring radionuclides; PHO, photonuclear reaction. Miscellaneous symbols: A, absolute intensity; R, relative intensity; X, x-ray line; D, doublet line; C, complex unresolved lines; W, weak intensity; <, the intensity is less than the value given.

[a]For all symbols except NFI and NAT the nuclide designator that follows refers to the target nuclide.
[b]For NFI the number following the symbol refers to the cumulative yield of the fission product.
[c]For NAT the number following the symbol is the percentage of natural isotopic abundance.

Appendix B. Running GAMIDEN at LLNL

At LLNL both a source deck (GAMIDENS) and a controllee (a compiled, loaded, ready-to-execute version called GAMIDEN) are available from photostore. They may be accessed by the XPORT command

RD.415000 : GAMDATA [GAMIDENS GAMTOT83 GAMIDEN].

GAMIDENS is the source deck, prefaced by control cards that instruct the computer to compile and load the program on a CRAY I computer. Since GAMIDEN is already available, recompilation of GAMIDENS is necessary only if it is changed by the user.
To compile GAMIDEN, type

CIVIC GAMIDENS GAMIDEN B H (Box no.) / T V,

where T and V are appropriate time and value numbers.
GAMIDEN is the executable controllee. To run a problem, type

GAMIDEN / T V.

The time and value numbers can be adjusted to the status of the computer. A typical problem requires less than 0.75 min.
Please note that disk file GAMTOT83 must be present.