USE OF NUCLEAR EXPLOSIONS FOR THE STUDY OF RADIATION DAMAGE PROCESSES*

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Introduction

The large flux of neutrons which accompanies chain fission reactions in nuclear reactors first led to extensive concern with the changes in the structure and properties of material subject to bombardment by energetic neutrons. Wigner's prediction[1] of crystalline changes as a consequence of neutron irradiation led to intense engineering interest in possible modifications of fuel and moderator materials in thermal reactors. It was found that a large number of materials showed substantial changes in electrical, thermal, and chemical properties, along with profound distortions of crystalline microstructure following fast neutron irradiations of the intensities and for the times encountered in nuclear reactors. The obvious applications interest was coupled with the interest of solid-state physicists and physical metallurgists in the phenomenology of changes thus wrought in materials.

The changes brought about in the microstructure of a well-ordered crystal by the introduction of energetic neutrons are of interest because of the opportunity to test models of atomic interactions within the crystal. Stable configurations can be deduced through the use of x-ray and neutron diffraction techniques, and by small angle scattering experiments. Another powerful tool is lent by the study of thermal annealing phenomena, wherein the relaxation of the crystalline defects are observed to accompany an increase in temperature to a value substantially above the temperature at which the material was held during creation of the defects by neutrons.[2]

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It should be pointed out that an almost equally interesting facet of such studies of microscopic changes which can be produced and healed in crystals of a material is the opportunity afforded to link the various physical properties of the bulk material to such controlled changes within the crystals. Changes occurring within such complex agglomerates as artificial graphite are not only of great practical interest; they permit a comparison of the roles played by the relatively orderly crystalline material and the disordered filler phase insofar as the various measurable properties are concerned.

An overly simplistic but useful model of neutron-induced radiation damage in a crystalline material may be envisaged as follows. The regular array of the perfect crystal is disrupted by collisions of fast neutrons and energetic displaced "recoil" atoms from the crystal, to yield a material with lattice vacancies ("Schottky" defects) and with displaced atoms wedged into high-energy locations among the remaining ordered atoms of the crystal ("Frenkel" defects). This model obviously is valid only when a relatively small fraction of the atoms have been displaced. If the displaced atoms are not in positions of chemical stability, they will continue to diffuse, provided that the lattice temperature is not too low to permit such diffusion. Such diffusing atoms constitute a sort of constrained gas, the partial pressure of which depends upon the intensity of the causitive neutron irradiation. Among the more likely targets for sorption of these gas atoms are crystal vacancies, crystal defects, crystal edges, or other gas atoms. Indication of the importance of the last sorption mechanism is believed to be provided by the phenomenon of "radiation annealing."

The bulk of the evidence for the radiation annealing phenomenon has come from studies of changes in the physical properties of graphite moderator in the long-lived production reactors at Hanford.[3] A reproducible quantity of data had been collected describing changes in the properties of graphite occurring as a consequence of irradiation at moderate temperature followed by long-term thermal annealing at a given elevated temperature. It was clearly demonstrated that long-term annealing at the same elevated temperature, while simultaneously irradiating with neutrons, resulted in a markedly greater degree of recovery from the effects of irradiation damage, than did thermal annealing alone. Besides being of great practical usefulness in extending the useful lifetime of nuclear reactors utilizing graphite as a neutron moderator, this discovery put at the disposal of the solid-state physicist a powerful tool for studying the formation and destruction of interstitial complexes of displaced atoms within an irradiated material. The effects of such foreign structures upon thermal conductivity, specific heat, and electrical properties of crystalline materials are of great interest.

Neutron irradiation rate would be expected to play at least two roles in affecting the nature of defects remaining in a material following the termination of irradiation. The higher the irradiation rate, the higher the steady state density of diffusing displaced atoms, and the greater the probability of a second-order process leading to the formation of a stable two-atom complex. Often times, such a complex may serve as a "seed" for the subsequent condensation of other interstitial atoms into a larger complex. On the other hand, very high intensity irradiation with disruptive particles increases the probability that such complexes

will fall victim to disruptive encounters. The effectiveness of such disruptive encounters in destroying complexes might be expected to increase with the diffusion rate, and hence the temperature. However, experimental demonstration of the validity of such an assumption, as well as the measurement of the physical parameters governing the dynamics of the process awaits the availability of sources permitting a wide range of irradiation rates. The present paper explores the possible usefulness of nuclear explosive tests for this purpose.

Experiments Using Explosive Nuclear Sources

The intense bursts of neutrons obtained from underground nuclear detonations at the Nevada Test Site of the U. S. Atomic Energy Commission have been used to perform cross section measurements upon a variety of nuclides, and the results have been reported in the open literature.[4] In Fig. 1 we see an underground enclosure for the explosive nuclear device centered at the "working point" 185 meters underground. The working point is connected by means of a large evacuated pipe with a collimator and experimental area at the surface of the ground. The energy spectrum of neutrons drifting up the pipe is broadened by means of a polyethylene moderator placed 40 cm from the neutron source adjacent to the 180-meter-long baffled pipe leading to the experimental area. The neutron source function for neutrons of less than 320 eV leaving the moderator is shown in Fig. 2. Note that the half-width of the pulse is less than 5 μsec.

The short duration of the neutron pulse at the source serves as a beam chopper, and the 180 meter drift tube serves to separate neutrons of varying energy in time at the experimental area. A simple calculation shows that neutrons of 1 MeV energy require 13 μsec to traverse the tube, those of 10 keV require 135 μsec, and those of 100 eV require 1.35 msec. Energy dependence of fission cross section of material deposited on thin foils exposed to the beam of neutrons is readily obtained by observing time dependence of fission particles emitted by the foil deposit and collected on a solid state detector.

Character of a Nuclear Bomb as a Neutron Source

There is a dearth of material pertinent to the evaluation of a nuclear bomb as a source of radiation for experimentation in the open literature. As a starting point I quote from the introductory portion of the book "Neutrons from an Atomic Explosion," by P. A. Yampol'skiy. This is available in English translation from the Office of Technical Services, U. S. Department of Commerce.[5]

> "The energy released by the explosion of one gram of TNT amounts to ~1000 calories. [Therefore, the energy released by 1 kilo tonne TNT equivalent is ~10^{12} calories.]... An explosion equivalent to one kilo tonne of TNT requires the fission of 1.5×10^{23} atoms, that is, about 60 grams of uranium or plutonium.... The fission process occurs within a very short period of time;... the time required for an explosion equivalent to 10 kilo tonnes of TNT equals about 3.5×10^{-7} second. In view of the exponential nature of the reaction development, the number of fission generations

required for a 1,000 kilo tonne explosion differs by only 10% from the number of generations required for a 10 kilo tonne explosion. It may therefore be assumed that the duration of the explosions of different power is several tenths of a microsecond.

"Judging from press reports, the bombs exploded during tests vary a great deal in their power (from one kilo tonne to ~10 mega tonnes). But regardless of the power of the explosions, their basic processes and the nature of their effect on the surrounding medium are the same. This applies not only to atomic explosions but also to hydrogen-bomb explosions.

"In a hydrogen-bomb explosion the thermonuclear reaction is used as an intermediate process whereby neutrons are initiated and bring about the [fast] fission of ^{238}U atoms. The energy of uranium nuclear fission therefore accounts for a considerable portion of the power of the thermonuclear bomb.

"The number of neutrons released by an explosion of a purely thermonuclear [fusion] bomb may be several times greater than the number of neutrons released by an explosion of similar power based on fission reactions. If in a thermonuclear bomb use is made of the fusion reaction of tritium and deuterium, each reaction releases one neutron and an energy equal to 17.6 MeV. [Momentum considerations give the neutron 14.1 MeV of this energy and the helium recoil 3.5 MeV.] This energy is one tenth of the uranium or plutonium fission energy. Therefore, the number of neutrons per unit of released energy in such a bomb is considerably larger than in a bomb based on the fission reaction. Moreover, the neutron energy of a thermonuclear explosion is greater than the average neutron energy of an atomic fission explosion....

"The enormous energy of an atomic explosion is released within a bomb [assembly] weighing about one tonne in less than 10^{-6} second. It follows from [the preceding] that in the explosion of a 20 kilo tonne bomb the average energy density equals $\sim 10^{7}$ cal/g. Such a colossal energy density must result in the heating of matter within the bomb [assembly] to stellar temperatures in the neighborhood of tens of millions of degrees. Matter heated to such high temperature releases energy in the form of electromagnetic radiation of a very broad spectrum ranging from x-rays to longwave electromagnetic radiation."

Simple application of the Stefan-Boltzmann radiation law to a blackbody at (say) 2×10^{7} °K gives a radiation rate of 2.2×10^{17} cal/cm^2 sec. Material of moderate or large atomic number, at such a temperature, is ionized on the average to a degree of about +8. The number of particles is therefore increased nine-fold, and the average specific heat may be

taken to be nine times the low temperature value of about 0.3 cal/g ^{o}C. This will give a heat capacity for a 1 ton assembly of about 2.7 x 10^{6} cal/^{o}K. If the bomb assembly be assumed to be a 1 meter diameter sphere, its surface area is 31,000 cm^{2}. The electromagnetic radiation rate at 2 x 10^{7} ^{o}K is 6.8 x 10^{21} cal/sec, under the assumption of a blackbody radiation law. These two numbers permit the calculation of an initial cooling rate of 2.5 x 10^{15} ^{o}K/sec. Of course, the emissivity of any plasma in the x-ray region is far less than one. Also, reflection and back radiation from the surrounding medium will further reduce the cooling rate. Nevertheless, it is evident that thermal quenching due to radiative energy losses is exceedingly rapid, and a steady state temperature will tend to be reached which depends upon the instantaneous rate of energy generation during the peak of the explosion.

Clearly the width of the intense pulse of blackbody electromagnetic radiation from the initial fireball will be determined by the length of the pulse of nuclear energy generated. This was above indicated to be of the order of a few tenths of a microsecond. The electromagnetic radiation (including prompt gammas) arrives at an experimental station distance D meters from the working point in a time

$$T_{e.m.} = (3.3 \times 10^{-9})\ D \quad \text{seconds} \tag{1}$$

after the nuclear explosion. In the evacuated tube there is no dispersion, quanta of x-rays arriving at the same instant as quanta of infra-red radiation.

The radiant cooling rate of 6.8 x 10^{21} calories per second previously estimated may be used to determine the electromagnetic radiation falling upon an experimental target a distance D meters from the source. If isotropy and no reflection be assumed, we obtain a peak radiation intensity of

$$I_{e.m.} = \frac{5.4 \times 10^{16}}{D^{2}} \text{ cal/cm}^{2} \text{ sec.} \tag{2}$$

If radiation emission is not a blackbody continuous spectrum, the intensity of electromagnetic radiation will be correspondingly lessened.

As was pointed out earlier, a range of neutron energies emitted leads to a separation of neutrons along the drift path following the explosion, so that neutrons of energy E (electron volts) arrive at a position D (meters) from the working point at a time

$$T_{neut} = \frac{7.2 \times 10^{-5} D}{\sqrt{E}} \text{ seconds} \tag{3}$$

after the nuclear explosion.

If an excess neutron yield of 1.8 neutrons per fission be assumed, a 20 kt device will yield 2.7 x 10^{24} neutrons. If these are distributed isotropically (no scattering assumed) the integrated neutron flux in the

pulse at a distance D meters from the working point will be

$$\phi_{neut} = \frac{4.5 \times 10^{19}}{D^2} \text{ neutrons/cm}^2. \quad (4)$$

As was pointed out above, the neutron output of a purely thermonuclear source of the same energy yield may be several times greater than the above figure for a fission source.

Concommitant Mechanical Problems

An abrupt ground shock follows the detonation of the nuclear device beneath the ground, the time delay after the explosion and the acceleration experienced being a function of the properties of the earth intervening, and of the device yield.

For a line-of-sight tube extending from the working point to the experimental area, there will, of course, be a gaseous shock wave and particles of solid matter projected from the explosion at high velocities. The feasibility of detonation of underground nuclear explosions without atmospheric contamination by radioactive debris necessitated the development of fast-acting closures. Such closures seal off the gases evolved from the nuclear bomb itself and from surrounding materials completely, shortly after the passage of the neutrons generated within the device.

The final environmental problem is the collapse of the pocket in the earth following the cooling of gases formed in the explosion. The size of the collapse crater, and the time required for collapse to occur depend upon the device yield, and upon the characteristics of the soil within which the explosion is set off. Typically, the surface of the ground above the working point will accelerate at 0.8 g within an hour after shot time for a distance of thirty feet, where it will rapidly decelerate at 50 g or more. The crater remaining after the collapse may be about 200 feet in diameter.

High Intensity Radiation Damage and Radiation Annealing Experiments

Passive experiments, with samples whose physical properties are to be measured before and after exposure, may be performed at distances from the working point ranging from about 50 meters to 200 meters or more. For the reference source we have developed expressions for, the resulting fast neutron exposures will range from 2×10^{16} to 10^{15} cm^{-2}. For a thermonuclear source of equal yield, fluxes of 14-MeV neutrons will be several-fold higher in intensity. For metal or ceramic targets, about 1% of the neutron energy will be dissipated per millimeter traversed. For a 2×10^{16} cm^{-2} burst of 1-MeV neutrons, approximately 80 cal/cm^3 will be dissipated, resulting in a temperature rise from neutron heating of about 80°C in a material having a specific heat of 0.2 and a density of 5 g/cm^3.

The heating due to the flash of electromagnetic radiation preceding the neutron burst must be considered. If the duration of the light flash

is taken as 10^{-7} sec, use of the preceding expression for intensity gives a flash energy density of 2×10^{6} cal/cm^2 at a distance of 50 meters from the source. The seriousness of the heating effect depends upon the spectrum of the radiation. High energy quanta will penetrate the sample with little energy absorption. Low energy quanta will dissipate their energy in the first few microns of the exposed surface of the sample. Selective filtering may be employed to reduce the intensity of troublesome frequencies in the electromagnetic flash, while attenuating the neutron beam by a relatively small factor.

In order to observe property change transients immediately following irradiation, the sample may be instrumented for remote measurement of properties. Techniques developed at North American Aviation (now North American Rockwell) by associates of the author in the early 1950's for radiation damage studies using the Berkeley 60" cyclotron may be adapted.[6] Electrical resistance and thermoelectric properties are easiest to measure, although Hall coefficient, magnetoresistance and thermal conductivity can also be measured remotely. The above-mentioned cyclotron work also successfully provided for helium gas cooling, permitting measurement of properties at controlled low temperatures. In the event that it becomes desirable to measure effects of irradiation intensities requiring exposure at distance less than 50 meters from the source, the difficulty of recovery for post-irradiation measurement necessitates instrumentation for in-place measurement of changes in the desired properties.

Should it be desirable to observe the differential effect of neutrons of various energies, a polyethylene moderator may be placed near the neutron source, and neutrons of energies ranging downward from those produced by the source will be obtained. At a distance of 100 meters from a thermonuclear source, 14-MeV neutrons arrive a little less than 2 μsec after detonation, 1-MeV neutrons require 7 μsec; 100 keV, 23 μsec; and 10 keV, 70 μsec. It is feasible to mount samples to be exposed near the edge of a 1 meter diameter wheel, which is spun up to 1000 rpm just prior to detonation time, The linear velocity of the samples being approximately 0.05 mm/μsec, regions exposed to 1-MeV neutrons will be separated by 0.25 mm from the onset of irradiation by 14-MeV neutrons. Regions irradiated by 100 keV and 10 keV neutrons are 1.1 mm and 3.5 mm displaced, respectively. Since it is not feasible to spin the wheel with a known phase relationship to the explosion time, it will be necessary to place samples of the material being studied around the entire perimeter of the wheel, then locate the exposed region by autoradiography of a thin foil placed adjacent to the material sample ring.

Direct Energy Conversion Studies

Many physicists feel that a more elegant procedure for the conversion of the energy of fission particles into useful forms of power should be possible. Efforts to produce large electrostatic potential differences in vacuo have been discouraging because of the large field emission electron currents which essentially short out the fission particle currents.[7] High fission particle irradiation rates occurring for very short times will permit the transient study of elemental processes in plasmas and in solids. A fission particle pulse can be initiated by a fast neutron burst

impinging upon a fissionable material. Intense fission particle bursts of less than microsecond duration may be obtained by using a plate of a fissionable material having suitable cross section for fast fission. Vacuum or semiconductor experimental arrays can be designed which permit a study of the dynamics of secondary electron production, and the measures which may be used to suppress or utilize these in energy conversion. Also, the rearrangement of cyclotron resonance state populations in materials within a magnetic field will shed light on the possible usefulness of the fission particle pumped cyclotron resonance maser concept.

The production of excited states by forbidden transitions induced by fission particle collisions within chemical systems could be explored at very high intensity fission particle irradiation intensity within a fluid containing a relatively small quantity of fissile material. Such fission recoil synthesis of chemical species has recently received attention[8] and is closely related to the new field of Plasma Chemistry.

Accessibility of the N.T.S. Facility to Independent Researchers

To the writer's knowledge, no provisions have been made for independent investigators to undertake research unrelated to the A.E.C. programmatic projects at the Nevada Test Site. However, there are a number of contractors carrying out research, some of which has given results of sufficiently general interest to warrant open publication. Opportunities for university faculty members and graduate students to participate in such programs, under sponsorship of one of the contractors, exist. The preceding paper has set forth the thesis that investigations of the effects of high intensity neutron irradiation on the physical properties of solids, which can be conducted at the Nevada Test Site will yield useful, publishable data.

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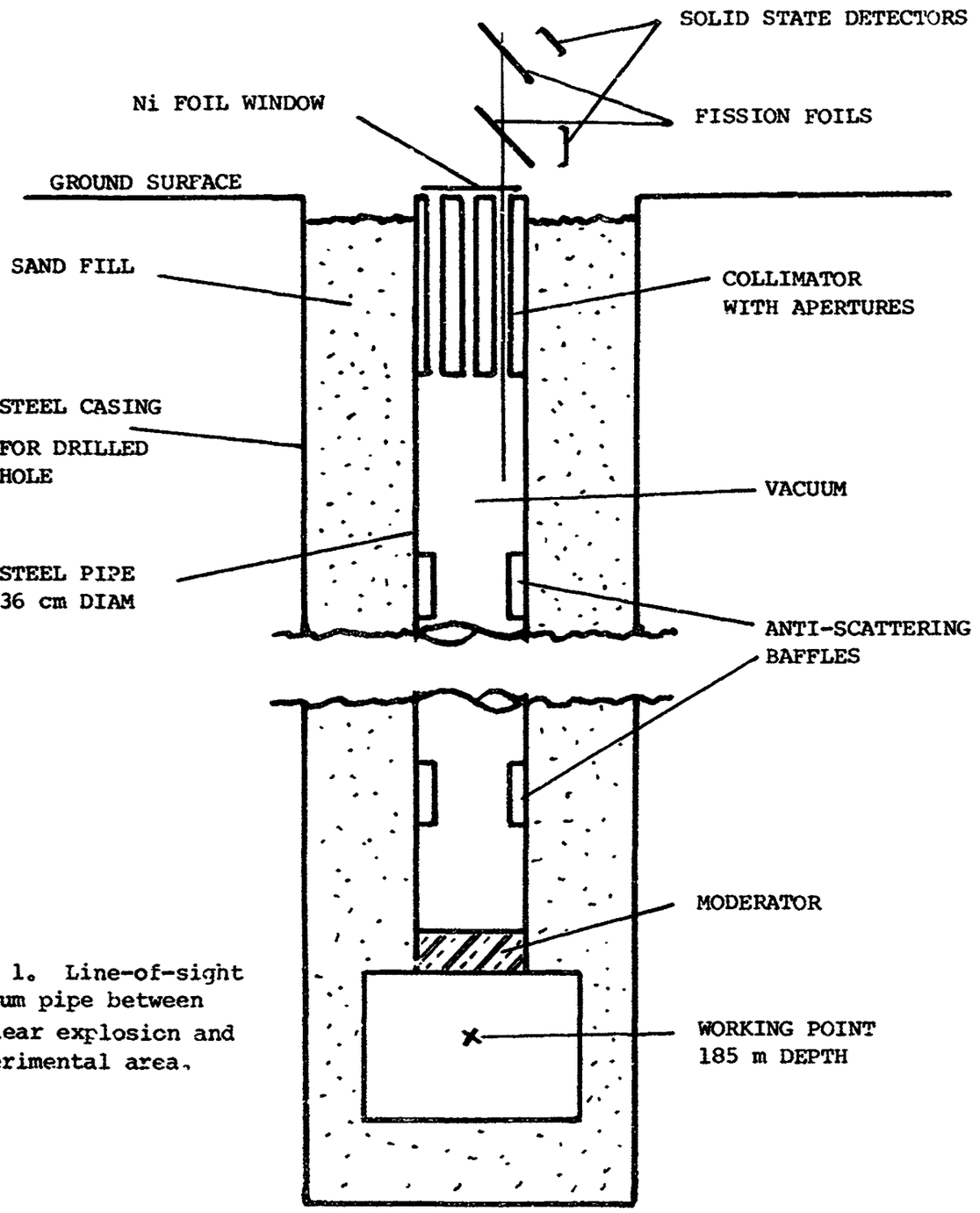

Fig. 1. Line-of-sight vacuum pipe between nuclear explosion and experimental area.

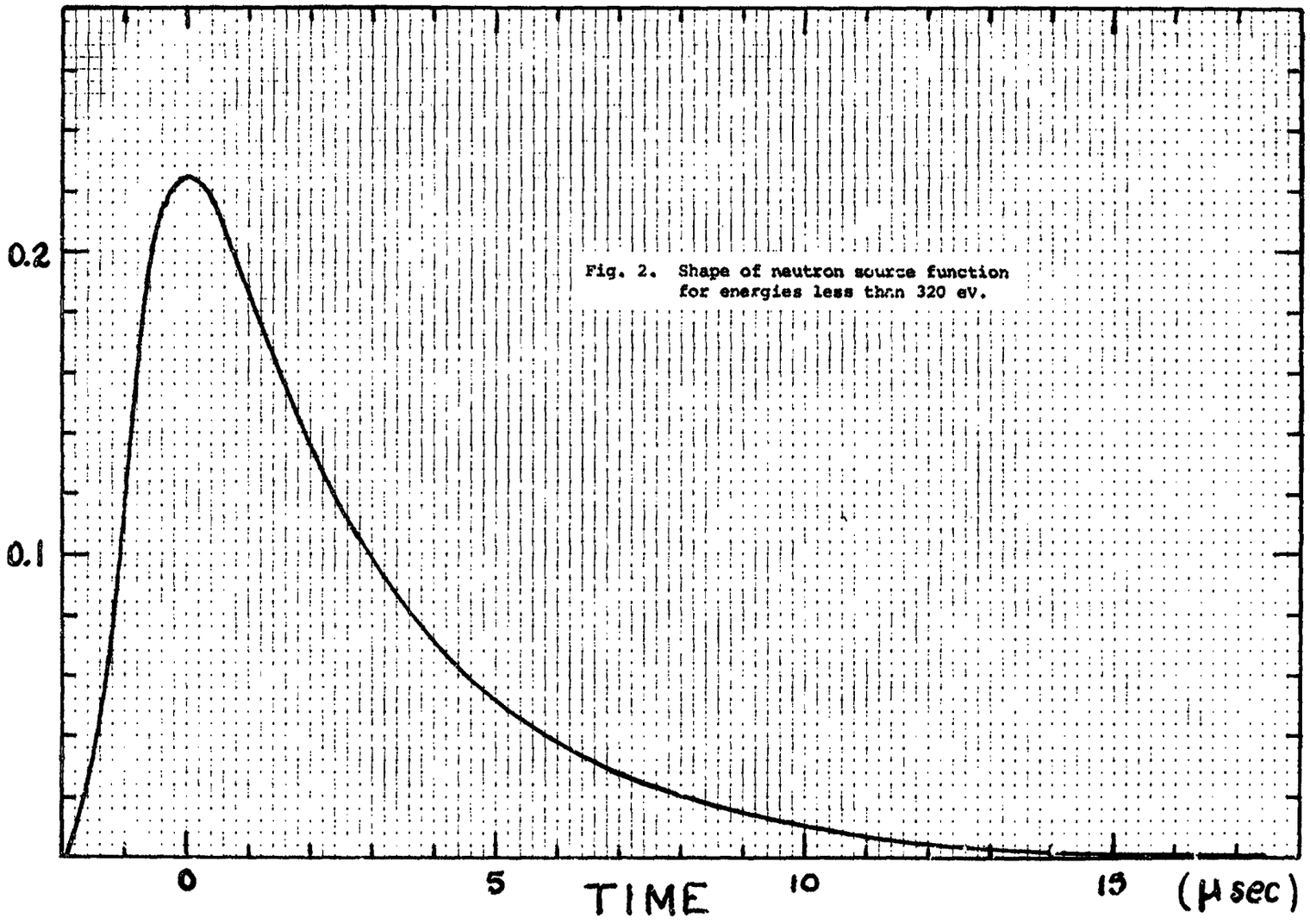

Fig. 2. Shape of neutron source function for energies less than 320 eV.