Criticality aspects of actinide nuclides and their relevance to the long-lived-fission-waste problem

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From systematic analysis, carried out by us earlier, of the criticality parameters of over twenty fissile and fertile isotopes of eight transthorium actinide elements, it is observed that the infinite neutron multiplication factor K_{∞} increases and critical mass decreases monotonically with the 'fissility parameter' Z^2/A of the nuclides. This implies that each and every isotope of transuranic elements such as neptunium, americium, curium, etc., which are produced as byproducts during reactor operation, is a more valuable nuclear fuel than the corresponding fissile/fissible isotopes of plutonium. This finding has a profound bearing on the long-lived-fission-waste problem and supports the view that the byproduct actinide elements should be separated from the 'high-active waste' stream and recycled back into fission reactors, thereby eliminating one of the commonly voiced concerns regarding the acceptability of nuclear fission power.

CONSIDERABLE quantities of actinide elements such as protactinium (Pa), neptunium (Np), americium (Am) and curium (Cm) have accumulated in the spent fuels of nuclear fission reactors over the last four decades. The quantum of nuclides formed decreases near-exponentially with the mass number A of the nuclides[1]. Even though some of these nuclides have important applications in the fabrication of radioisotope electric generators, neutron sources, space reactors[2], etc., these byproduct actinide nuclides have generally been looked upon as 'nuclear waste' and are proposed to be disposed off along with high-active wastes in deep-geological repositories.

The actinide nuclides can be classified on the basis of criticality properties[3] into three categories, namely fissile, fissible and fertile nuclides. The fertile nuclides are those nuclides such as thorium-232 (^{232}Th) and uranium-238 (^{238}U), which, even though fissionable by fast neutrons, cannot sustain a chain reaction. The term 'fissible' was introduced[4] to describe actinide nuclides such as ^{231}Pa, plutonium-240 (^{240}Pu) etc., which, in spite of having sharp thresholds in their fission cross-sections in the 0.3- to 0.6-MeV neutron energy range, are still capable of independently supporting a fast-neutron fission chain reaction. It is observed[5] that, with a few exceptions, all fissible nuclides have an even number of neutrons and all fissile nuclides have an odd number of neutrons.

We have generated afresh criticality parameters of over twenty fissile and fissible isotopes of eight actinide elements, starting from basic point cross-section files. The methodology used and results obtained are discussed in detail elsewhere[6]. We present here only the essential results and conclusions of our study and comment on the implications of our findings on the long-lived-fission-waste problem.

Cross-sections and codes used

Cross-section data from the evaluated nuclear data file ENDF/B-IV (ref. 7) and the evaluated nuclear data library ENDL-82 (ref. 8), which provide absorption, scattering, fission and other basic point cross-section values as a function of energy for various materials, were employed to generate 35-group cross-section sets[9] using a special processing code known as MINX. The ENDL-82 library was used for those nuclides for which data are not available in the ENDF/B-IV library. The cross-section set for ^{231}Pa was derived by combining the Bhabha Atomic Research Centre (BARC)-evaluated data in the 1- to 20-MeV-energy region with the ^{233}Pa data of the JENDL-2 file below 1 MeV.

Multigroup transport theory code DTF-IV, which solves the one-dimensional Boltzmann neutron-transport equation using what is known as the discrete-ordinates method to give neutron density as a function of position, direction and energy inside a given medium, was used to compute infinite multiplication factor K_{∞} and bare critical mass for spherical fast systems of the actinides. K_{∞} is defined as the average number of neutrons produced per neutron absorbed in an infinite-sized neutron-multiplying assembly. The 35-group cross-section sets[9] were employed in these computations. Table 1 summarizes the results. Column 3 of the table gives the $\bar{\nu}$ values corresponding to the neutron energy range 0.6 to 1.1 MeV (9th group of 35-group set). Column 8 gives the critical surface mass density σ_c^b given by $\rho R_c^b/3$, where ρ is the core density and R_c^b the bare critical core radius. The importance of σ_c^b in criticality considerations has been pointed out earlier[6]. The parameter q_c^b is a measure of the critical core size in units of neutron mean free path and therefore is independent of core density. The critical mass M_c^b (column 7 of the table) is obtained from σ_c^b using the relation

$$M_c^b = \frac{36\pi(\sigma_c^b)^3}{\rho^2}. \quad (1)$$

Table 1. Criticality data for bare fast assemblies of actinide nuclides.

				DTF IV Results			
Nuclide	Fissility parameter (Z^2/A)	Neutrons per fission	Density ρ (g cm^{-3})	K_∞	Critical radius R_c^b (cm)	Critical mass M_c^b (kg)	σ_c^b (g cm^{-2})
Fissile (34.10)*							
$^{239}_{92}$U	35.41	2.576	18.90	1.410	12.32	145.65	77.60
$^{235}_{92}$U	36.02	2.528	18.90	2.337	8.18	43.33	51.53
$^{233}_{92}$U	36.33	2.548	18.90	2.524	5.79	15.37	36.48
$^{241}_{94}$Pu	36.66	3.058	19.00	2.912	5.35	12.18	33.20
$^{239}_{94}$Pu	36.97	2.994	19.00	2.956	5.02	10.13	31.84
$^{242m}_{95}$Am	37.29	3.365	11.87	3.238	7.08	17.65	28.01
$^{247}_{96}$Cm	37.31	3.651	13.51	3.658	5.18	7.87	23.33
$^{245}_{96}$Cm	37.62	3.585	13.51	3.430	6.01	12.28	27.07
$^{243}_{96}$Cm	37.93	3.584	13.31	3.487	5.23	8.10	23.55
$^{251}_{98}$Cf	38.26	4.068	13.84	3.978	5.24	8.24	24.17
$^{249}_{98}$Cf	38.57	4.104	13.84	3.993	5.12	7.78	23.62
Fissible (35.70)*							
$^{232}_{90}$Th†	34.91	2.152	11.30	0.084	—	—	—
$^{238}_{92}$U†	35.56	2.851	18.90	0.380	—	—	—
$^{231}_{91}$Pa	35.85	2.598	15.37	2.199	13.61	162.31	69.73
$^{236}_{92}$U†	35.86	2.482	18.90	0.672	—	—	—
$^{234}_{92}$U	36.17	2.478	19.00	1.477	13.70	203.20	86.74
$^{237}_{93}$Np	36.49	2.854	20.45	1.746	8.60	54.48	58.62
$^{242}_{94}$Pu	36.51	2.936	19.00	2.169	8.84	54.98	55.99
$^{240}_{94}$Pu	36.82	3.000	19.00	2.308	7.70	36.33	48.77
$^{238}_{94}$Pu	37.13	2.845	19.84	2.884	5.02	10.54	33.20
$^{243}_{95}$Am	37.14	3.150	11.87	1.926	22.89	596.24	90.56
$^{241}_{95}$Am	37.45	3.090	11.87	2.519	11.47	75.07	45.39
$^{244}_{96}$Cm	37.77	3.343	13.51	3.032	7.21	21.21	32.47

*Threshold value of (Z^2/A) required for attaining finite critical mass.
†Fertile nuclide.

Systematics of criticality data

Correlation between $\bar{\nu}$ and (Z^2/A)

The dependence of $\bar{\nu}$, the average number of neutrons emitted per fission, on Z and A has earlier been studied by Manero and Konshin[10]. Figure 1 shows a plot of the $\bar{\nu}$ values given in Table 1 against the fissility parameter Z^2/A of the actinide nuclides. It is seen that $\bar{\nu}$ increases monotonically with Z^2/A.

Correlation between K_∞ and (Z^2/A)

Figure 2 shows plots of the variation of K_∞ with (Z^2/A) for fast fissile and fissible cores. It can be seen that data points for the fissile and fissible actinides fall into two distinct groups, and the variation of K_∞ with Z^2/A for each group is linear, with a slope different from that of the other. 'Eye-fit' straight lines to the data points yield the relations

$$K_\infty \simeq 1.0 + 0.7\,[(Z^2/A) - 34.1] \text{ (fissile)}, \quad (2)$$

$$K_\infty \simeq 1.0 + 1.1\,[(Z^2/A) - 35.7) \text{ (fissible)}. \quad (3)$$

The above relations indicate that threshold Z^2/A values of at least 34.1 and 35.7 are required for fissile and fissible actinides respectively to sustain a fission chain reaction in unmoderated systems. The data points for ^{239}U and ^{247}Cm, of the fissile actinides, are well away from the line. In the case of fissible actinides there is considerable spread in the data points. The plots indicate that fertile actinides are likely to be more

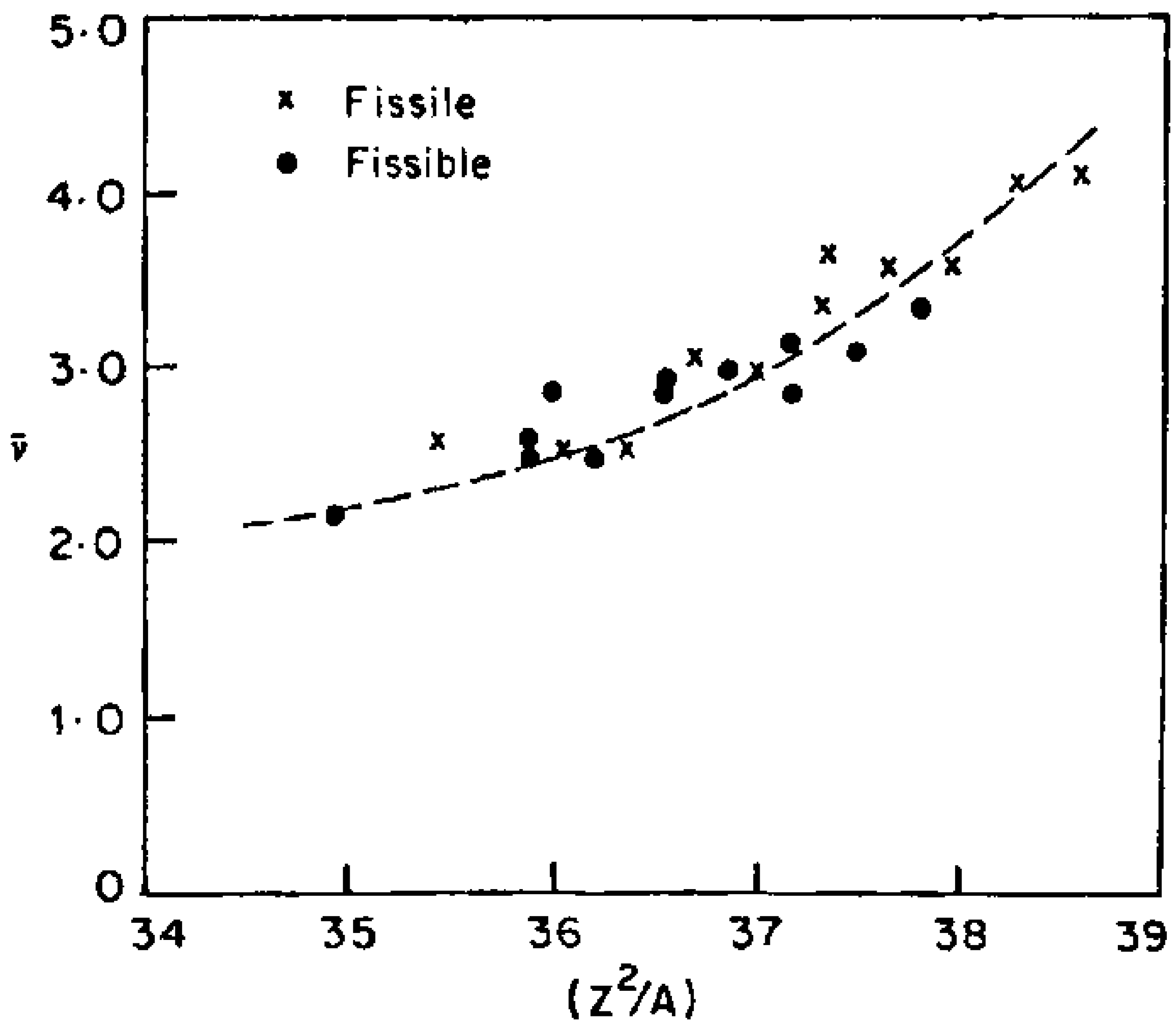

Figure 1. Plot of $\bar{\nu}$ vs (Z^2/A) for actinides.

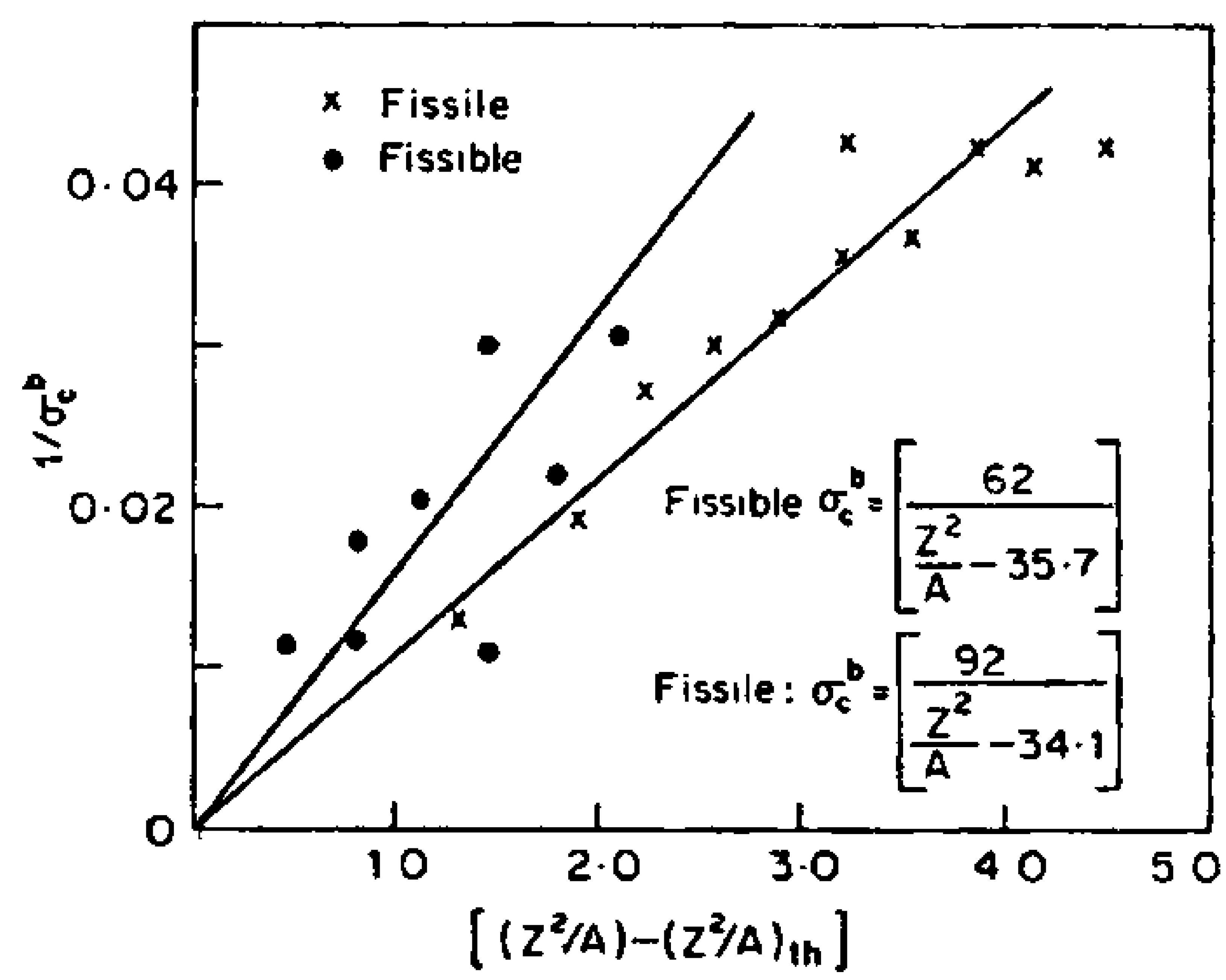

Figure 3. Plot of $(1/\sigma_c^b$ vs $[(Z^2/A)-(Z^2/A)_{th}]$ for fast actinide assemblies.

reactive than fissile actinides beyond a Z^2/A value of ~ 39.5.

Correlation between σ_c^b and (Z^2/A)

It is apparent that σ_c^b, which measures critical size, should tend to infinity when K_∞ reduces to unity. Hence a plot of $1/\sigma_c^b$ vs Z^2/A for fissile nuclides should intercept the Z^2/A axis at 34.1. Figure 3 shows a plot of $1/\sigma_c^b$ vs $[(Z^2/A)-(Z^2/A)_{th}]$, where $(Z^2/A)_{th}$ is taken as 34.1. It is seen that the data points fall on a straight line which passes through the origin. The linear behaviour is surprising since there is no a priori reason why the points should fall on a straight line. It is found that the data points corresponding to fissible nuclides also can be fitted into a straight line provided a threshold Z^2/A value of 35.7 is used for them. Thus all the $1/\sigma_c^b$ data points are seen to fit relations of the form

$$\sigma_c^b \simeq \left[\frac{92}{[(Z^2/A)-(34.1)]}\right] \text{(fissile)}, \qquad (4)$$

$$\sigma_c^b \simeq \left[\frac{62}{[(Z^2/A)-(35.7)]}\right] \text{(fissible)}. \qquad (5)$$

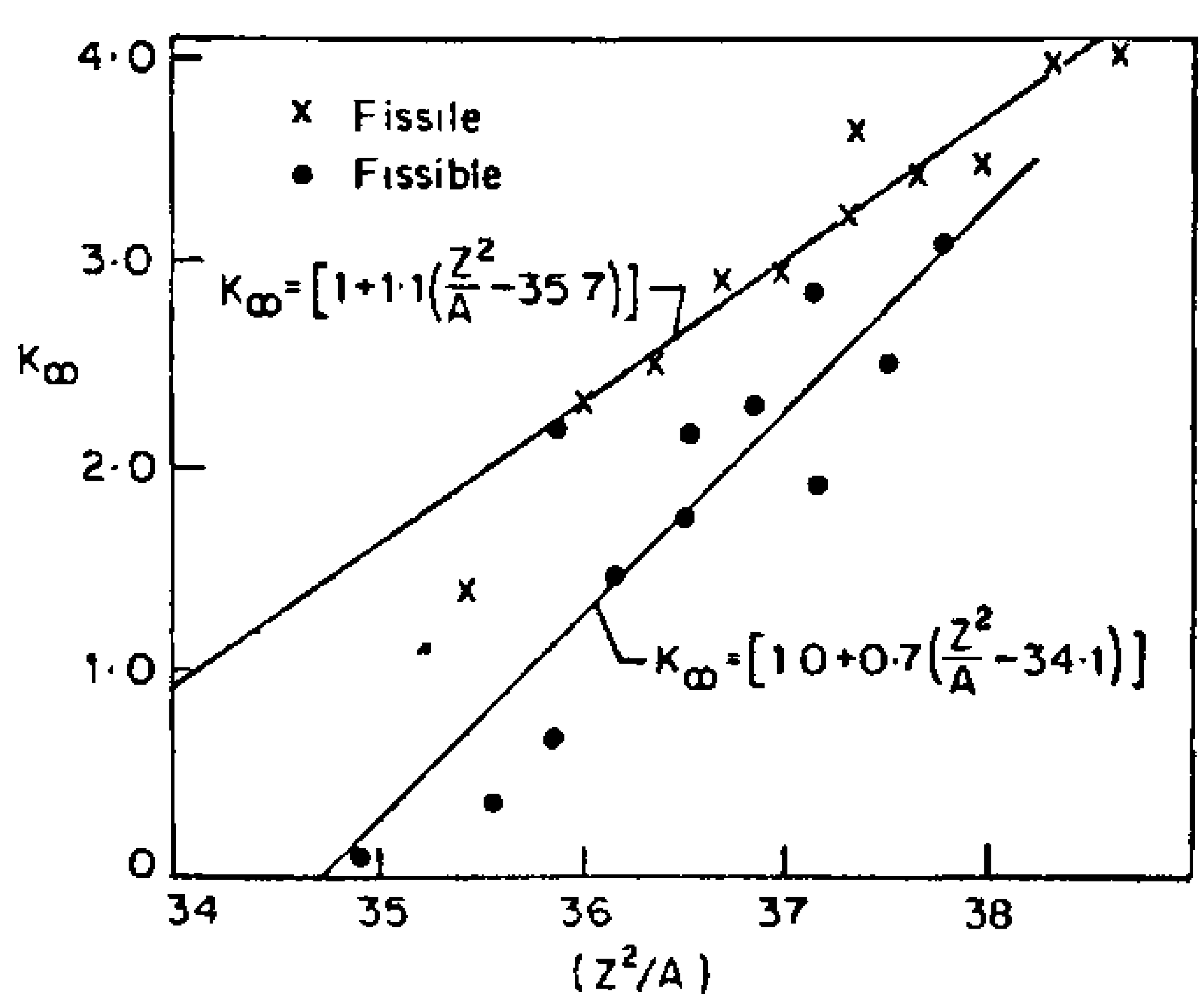

Figure 2. Plot of K_∞ vs (Z^2/A) for fast actinide cores.

Summary and conclusions

The most important conclusion to emerge from this study is that the K_∞ values of both fissile and fissible nuclides increase systematically with their fissility parameter Z^2/A. Correspondingly the critical size measured in terms of the surface mass density σ_c^b also shows a systematic decrease with Z^2/A, indicating that each and every isotope of Np, Am, Cm, etc. generated as byproducts during reactor operation is a better nuclear fuel than the corresponding odd- or even-neutron nuclides of plutonium. If reactor-grade plutonium, along with its higher-mass isotopes such as ^{240}Pu, ^{241}Pu and ^{242}Pu, can be treated as valuable fuel material for both fast and thermal reactors, including light water reactors (LWR), then it is clear that, on the basis of reactor-physics considerations alone, there is no justification for treating the special actinides as 'waste'[11]. The main objective of the present paper is to draw the attention of the scientific community to this feature of the criticality properties of the so-called waste actinides. Wide recognition and acceptance of the special actinide materials as a potential nuclear fuel rather than as long-lived alpha-active waste would help eliminate at least one of the negative aspects of nuclear fission energy from the public-acceptability point of view.

It also follows from the present study that there is no more any justification for pursuing exotic schemes for the neutron transmutation of 'waste actinides' using novel non-fission neutron sources such as spallation targets, superconducting cyclotrons or fusion-reactor blankets, for it is obvious now that all these nuclides are, if at all, better fuels for any type of fission reactor than even plutonium.

The study also has a bearing on the so-called shelf-life of plutonium. There seems to be a widely accepted contention that when 'fissile' ^{241}Pu (half-life of 14.4 years) decays to 'non-fissile' ^{241}Am there is a 'significant loss' of fuel value[12]. In fact this 'belief' has often been used to justify the near-term recycle of the growing stocks of plutonium in LWR while awaiting the commercialization of liquid metal fast breeder reactors (LMFBR), which has been delayed. The systematics of criticality data presented in this paper (see Table 1) clearly indicates that the fissionability properties of ^{241}Am with $K_\infty = 2.519$ and $\sigma_c^b = 45.39\ g\ cm^{-2}$ (Table 1 column 9), seem to be better than those of even ^{235}U, whose K_∞ is 2.337 and σ_c^b is $51.53\ g\ cm^{-2}$ in a hard-spectrum fast reactor.

A study of systematics of the type reported here provides the possibility of identifying those nuclides whose basic nuclear data warrant reexamination. For example, the data points of ^{247}Cm and ^{231}Pa, which fall outside the overall trend, possibly warrant a reappraisal of their input nuclear data. Further, by scrutinizing whether the deviation from the overall trend occurs in the $\bar{\nu}$ plot, the K_∞ plot or the $1/\sigma_c^b$ plot, one can assess which segment of the nuclear data needs refinement.

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RESEARCH COMMUNICATIONS

The role of compilers in computer-system performance

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The compiler is a system code which translates the user's program into machine-executable code and thereby dictates the performance of the computer system. Here we present results obtained while benchmarking various FORTRAN compilers on Motorola 68020-based machines. Our results dramatically show the effect of compilers on the performance of computing systems.

THE Advanced Numerical Research and Analysis Group (ANURAG) is building a parallel-processing system called PACE[1], primarily for computational-fluid-dynamics (CFD) applications. CFD is computationally intensive and therefore the basic aim of the parallel-processing system is to provide a platform that gives very high computational throughput. This requires careful choice of hardware. The hardware and operating-system environment of PACE have been chosen on the basis of several considerations (reported elsewhere, see ref. 2). For a given system configuration, the performance hinges crucially on the efficiency of the compiler used to generate executable code.

Our purpose in this paper is to report some results and observations made while benchmarking several FORTRAN compilers for use on our parallel processor. The study was motivated by the need to maximize computational throughput on PACE. Generally one associates system speed with hardware performance. However, the systems software also has a significant role in determining system speed. The benchmark results reported in the literature do not usually consider this aspect. Therefore, having chosen the specific system and hardware configuration for PACE, ANURAG decided to explore the possibility of fine-tuning the